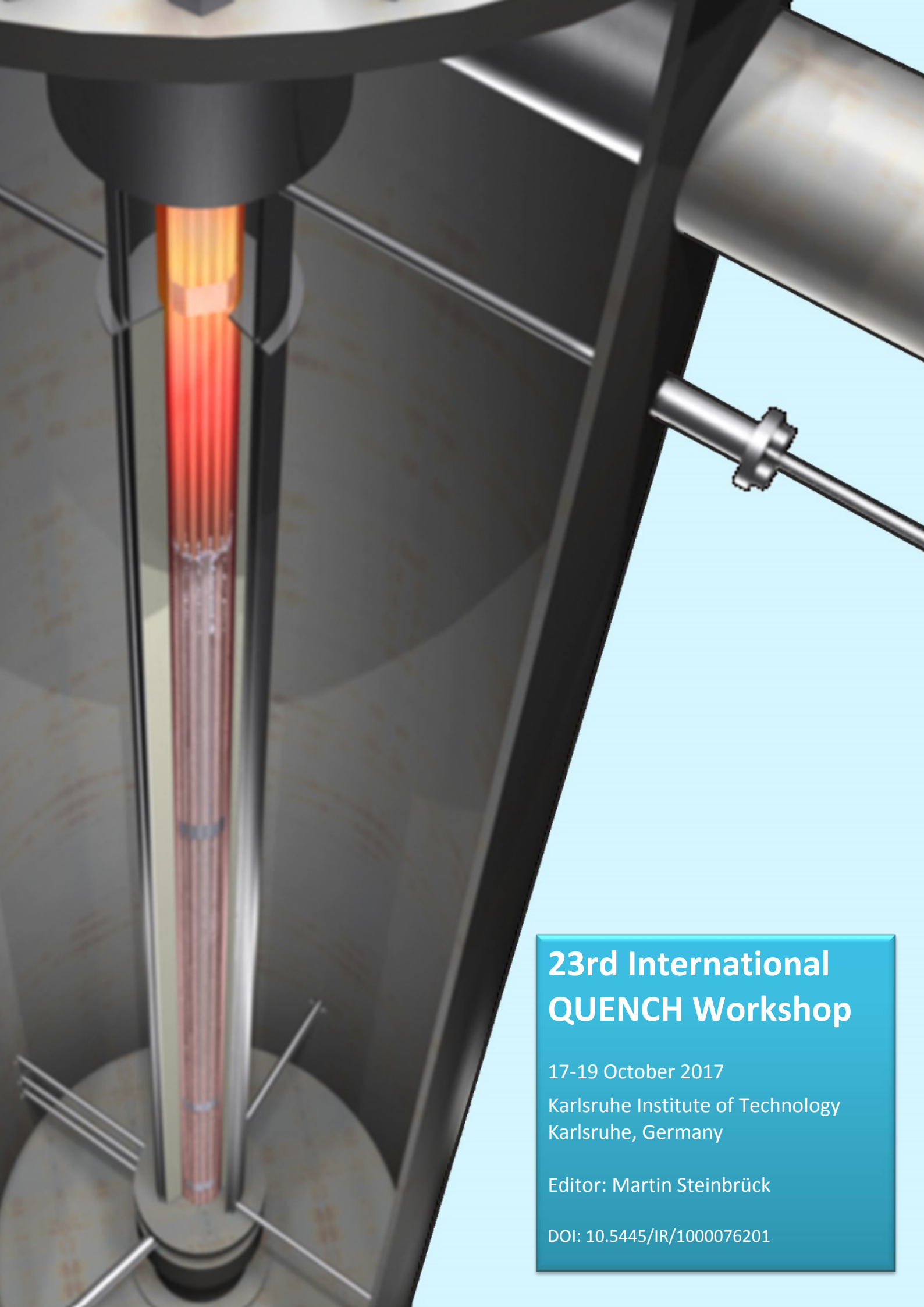


23rd International QUENCH Workshop

17-19 October 2017

Karlsruhe Institute of Technology
Karlsruhe, Germany

Editor: Martin Steinbrück

DOI: 10.5445/IR/1000076201

AGENDA

23rd International QUENCH Workshop*

Karlsruhe Institute of Technology, Campus North, H.-von-Helmholtz-Platz 1, 76344 Egg.-Leopoldshafen, Germany
October 17-19, 2017

Meeting Location: Fortbildungszentrum für Technik und Umwelt (FTU), Auditorium (Aula)

Tuesday, 17 Oct 2017

8:30	**Registration**	
9:00	Welcome	M. Steinbrück, KIT
	QUENCH (Chair: J. Birchley)	
9:10	Update of the QUENCH program	M. Steinbrück, KIT
9:40	Lessons learned from the QUENCH-LOCA experiments	J. Stuckert, KIT
10:10	Pre-Test Benchmarks in the Frame of the NUGENIA QUESA Project: QUENCH-ALISA and CODEX-AIT3	T. Hollands, GRS
10:40	**Coffee break (Group Photo)**	
11:15	First results of the QUENCH-ALISA bundle test	J. Stuckert, KIT
11:45	Aerosol measurements in the QUENCH-ALISA test	J. Kalilainen, PSI
	INVITED TALKS (Chair: M. Steinbrück, KIT)	
12:00	IAEA Activities in the Area of Nuclear Power Reactor Fuel Engineering	M.S. Veshchunov, IAEA
12:30	Dry Storage of Spent Nuclear Fuel and High Active Waste in Germany	G. Spykman, TÜV Nord
13:00	**Lunch**	
	INTERNATIONAL PROGRAMS (Chair: M. Veshchunov, IAEA)	
14:00	UK Research in LOCA-Related Activities and Co-operation with the Karlsruhe Research Centre– A Historical Perspective	S. Walker, ICL pres. by D. Bottomley
14:25	The experimental and computational study of cluster of nuclear dangerous materials inside "Shelter" object	R. Godun, ISPNPP
14:45	JAEA-sponsored projects at the OECD-NEA in support of Fukushima Dai-ichi site activities	D. Bottomley, JAEA
15:10	**Coffee break**	
15:40	CLADS test facilities on BWR core material degradation	Y. Nagae, JAEA
16:00	Main objectives and results of the EU SAFEST project	A. Miassoedov, KIT
16:20	International Severe Accident Research Roadmaps within the SAFEST Project	D. Manara, JRC

Wednesday, 18 Oct 2017

	MODELLING AND CODE APPLICATION I (Chair: C. Allison, ISS)	
9:00	Development of Three-Dimensional Simulation Code for BWR In-vessel Core Degradation	T. Okawa, NRA
9:20	Implementation of QUENCH-10 experiment modelling experience for the modelling of severe accidents in spent fuel pools	K. Kaliatka, LEI
9:40	Novel percolation model in post-transition growth kinetics of heterogeneous zirconium oxide	A. Aryanfar, BIU
10:00	Advanced Analytical Model of Zr-Based Claddings Breakaway Oxidation in Steam-Oxygen-Nitrogen Mixtures	A. Vasiliev, IBRAE
10:20	Validation of the SOCRAT code on the integral experiment CORA-15	A. Tarasov, IBRAE
10:40	**Coffee break**	
	MODELLING AND CODE APPLICATION II (Chair: T. Haste, IRSN)	
11:10	QUENCH-06 Experiment Post-test Calculations and Integrated Uncertainty Analysis with RELAP/SCDAPSIM	C. Allison, ISS
11:30	QUENCH-14, QUENCH-16 and QUENCH-ALISA analysis with RELAP5/SCDAPSIM MOD3.5(KIT) and MOD3.5+(KIT)	O. Dutkiewicz, KIT
11:50	Pre-test Calculations for Test CODEX-AIT3 with ATHLET-CD	C. Bals, GRS
12:10	Integrated uncertainty analysis of CORA BWR experiments using RELAP/SCDAPSIM	C. Allison, ISS
12:30	**Lunch**	
	MODELLING AND CODE APPLICATION III (Chair: T. Hollands, GRS)	
13:30	Study of the influence of the axial discretization in simulations of Quench 06 with SCDAP/RELAP5.mod3.5dp	I. Spasov, TU Sofia
13:50	Assessment of a lower pool analysis model in RELAP/SCDAPSIM with LIVE experiment	H. Madokoro, KIT
14:10	Investigation of thermodynamic phases of Ag-Cd-In system at high temperature and its assessment by CALPHAD	K.V. Gajavalli, IRSN
14:30	Accident Progression Analysis Based on Experiments QUENCH-16 and QUENCH-10	P. Vryashkova, INRNE
15:00	**Bus to Germersheim Fortress and invited dinner**	

Thursday, 19 Oct 2017

	EXPERIMENTS I (Chair: M. Große, KIT)	
9:00	Accident Tolerant Materials - Ideas and Research Directions	B. Sartowska, INCT
9:20	Experimental Behaviour of Chromium Based Coatings	J. Krejci, UHJ
9:40	High-temperature oxidation resistance and self-healing behavior of Cr2AlC MAX phase coating on Zircaloy-4	C. Tang, KIT
10:00	A New Approach to Enhance the Oxidation Resistance of a Zirconium Alloy: High-Temperature Pre-Oxidation	C.M. Lee, UNIST
10:20	New Zircaloy Cladding Behavior Study in LOCA Condition and Annular Fuel R&D Introduction in China	X. He, CIAE
10:40	**Coffee break**	
	EXPERIMENTS II (Chair: J. Stuckert, KIT)	
11:10	Secondary hydriding of zirconium-based fuel claddings at high temperature (LOCA conditions) Part 1: Multi-scale study of hydrogen distribution	J.-C. Brachet, CEA
11:30	Secondary hydriding of zirconium-based fuel claddings at high temperature (LOCA conditions). Part 2: Effect of high hydrogen contents on metallurgical and mechanical properties	T. Le Hong, CEA
11:50	Air Oxidation of Sponge Based E110 Cladding Alloy at High Temperatures	P. Vrbka, CTU
12:10	Variation in time of the high-temperature oxidation of Zircaloy-4 in various atmospheres	P. van Appeldorn, KIT
12:40	**Lunch**	
	EXPERIMENTS III (Chair: J.-C. Brachet, CEA)	
13:45	A quantification of effect of nitrogen during air ingress scenarios	S. Park, HGU/ACT
14:10	Status report on nitriding model	B. Jäckel, PSI
15:00	**Closure of the Workshop**	M. Steinbrück, KIT
16:00	**CoreSOAR Progress Meeting (Room 160; only for project partners)**	T. Haste, IRSN
18:00	**Bus for train station and Hotels**	

**** In Cooperation with the International Atomic Energy Agency (IAEA)***



23. International QUENCH Workshop, 17-19 Oct 2017, KIT, Karlsruhe, Germany

Update of the QUENCH program

Martin Steinbrück, Juri Stuckert, Mirco Große et al.

The main objective of the QUENCH program at KIT is the investigation of the hydrogen source term and materials interactions during LOCA and the early phase of severe accidents including reflood. Bundle experiments as well as separate-effects tests are conducted to provide data for the development of models and the validation of severe fuel damage code systems.

The QUENCH bundle facility is a unique out-of-pile bundle facility with electrically heated fuel rod simulators and extensive instrumentation. So far, 18 experiments with various severe accident (SA) scenarios as well as a series of seven DBA LOCA experiments were conducted. The QUENCH-LOCA series was finished in 2016. One of the main results is the definition of the conditions for secondary hydriding around the burst position and its influence on the mechanical properties of the cladding rods.

The last SA test QUENCH-18 in the framework of the EC-China ALISA project with Ag-In-Cd absorber rods and air ingress phase was conducted end of September 2017. First results were presented by Stuckert.

Separate-effects tests during 2017 were focused on the interaction of zirconium alloys with atmospheres containing nitrogen as well as on various ATF cladding candidates.

QUENCH bundle tests are part of the validation matrices of most SFD code systems which was also reflected during the session “Modelling and code validation”.

Finally, the status of reporting and publishing as well as the numerous national and international cooperations were briefly described and acknowledged.


KIT
arlsruher Institut für Technologie
Martin Steinbrück

Karlsruhe Institute of Technology

Institute for Applied Materials

Update of the QUENCH Programme

M. Steinbrück, J. Stuckert, M. Große et al.

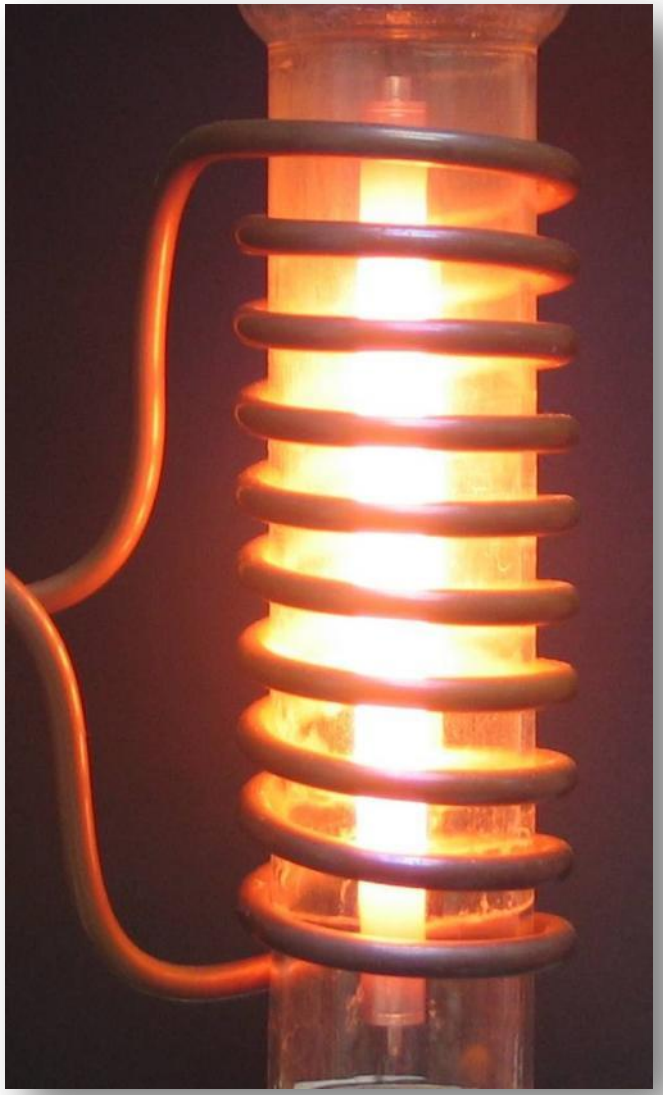
23rd International QUENCH Workshop, Karlsruhe, 17-19 October 2017

Institute for Applied Materials, Programme NUSAFE



Outlook

- Motivation
- Bundle experiments
- Separate-effects tests
- ATF activities
- Modelling / Code validation
- Future planning



Motivation

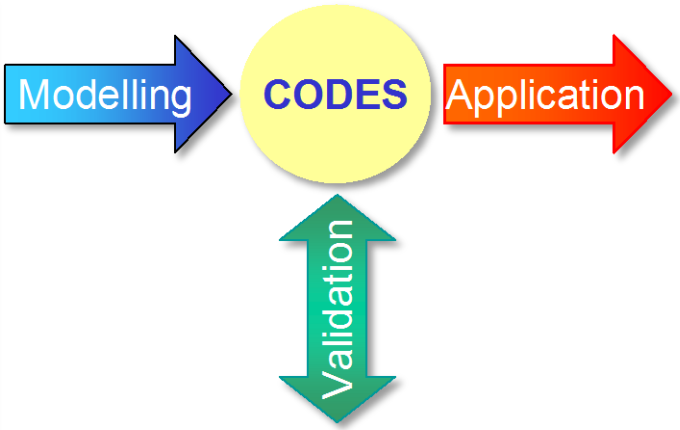
- Reflood is a prime accident management measure to terminate a nuclear accident
- Reflood may cause temperature excursion connected with increased hydrogen and FP release (severe accidents) and embrittlement of cladding and secondary hydriding (LOCA)
- Coolability of a degraded core is a matter of high priority (Fukushima)
- QUENCH experiments (bundle+SET) provide data for development of models and validation of SFD code systems

QUENCH Programme

Investigation of hydrogen source term and materials interactions during LOCA and early phase of severe accidents including reflood



Separate-effects tests





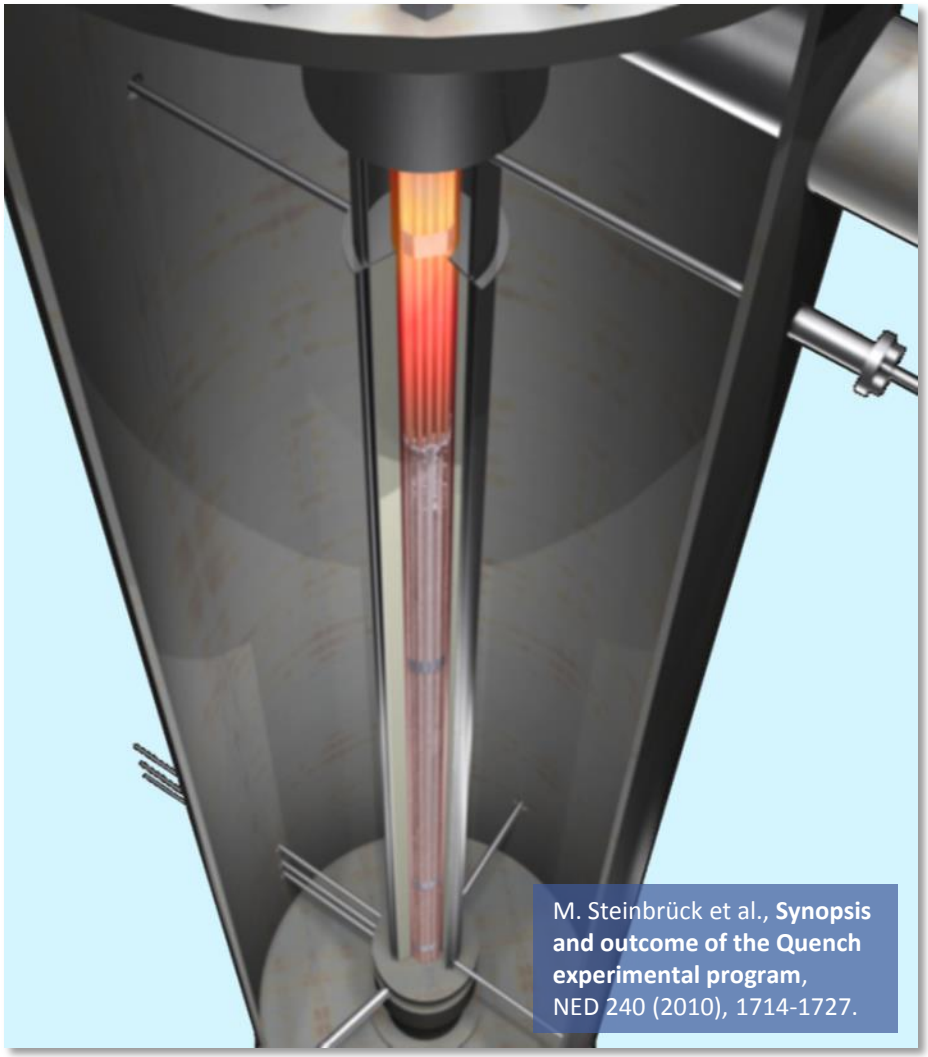
Bundle experiments



PWR fuel element

QUENCH facility

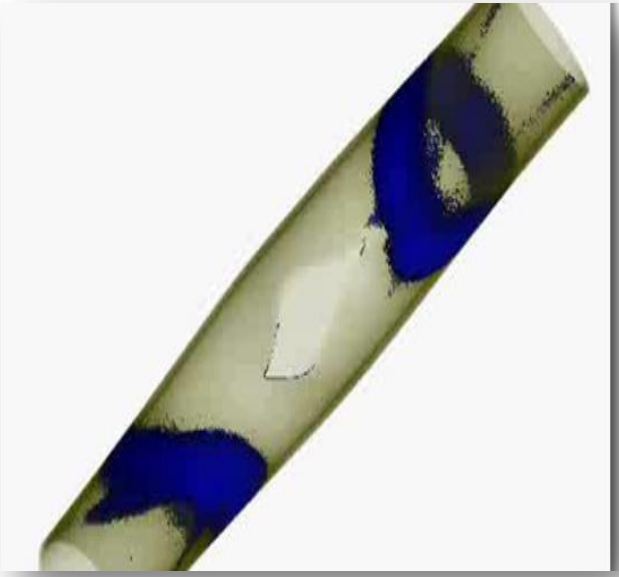
- Unique out-of-pile bundle facility to investigate reflood of an overheated reactor core
- 21-31 electrically heated fuel rod simulators; T up to >2000°C
- Extensive instrumentation for T, p, flow rates, level, etc. + MS
- So far, 18 experiments on SA performed (1996-today)
  - Influence of pre-oxidation, initial temperature, flooding rate
  - B_4C, Ag-In-Cd control rods
  - Air ingress; debris formation
  - Advanced cladding alloys
- 7 DBA LOCA experiments with separately pressurized fuel rods

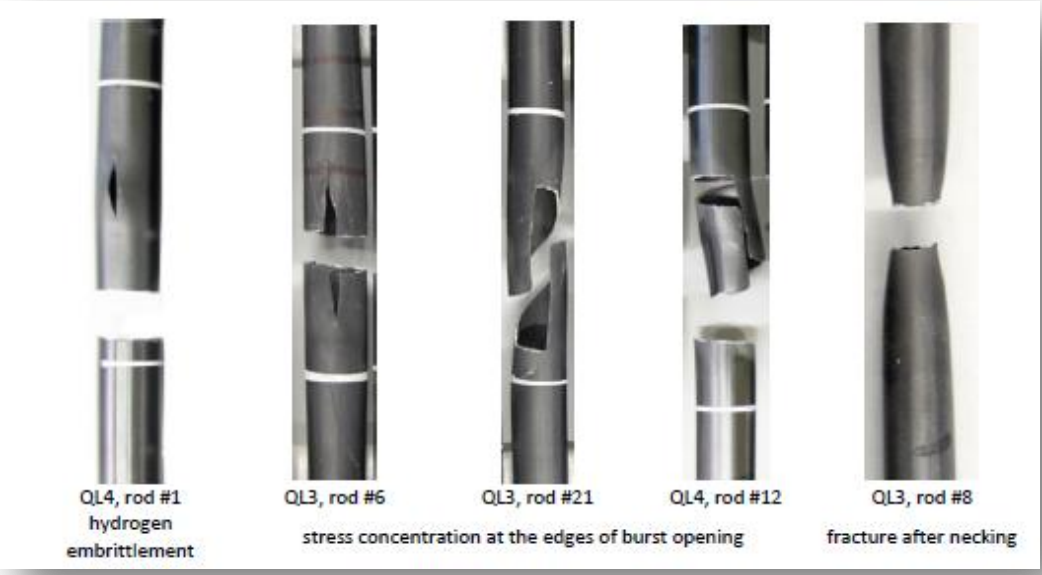


M. Steinbrück et al., **Synopsis and outcome of the Quench experimental program**, NED 240 (2010), 1714-1727.

QUENCH-LOCA series

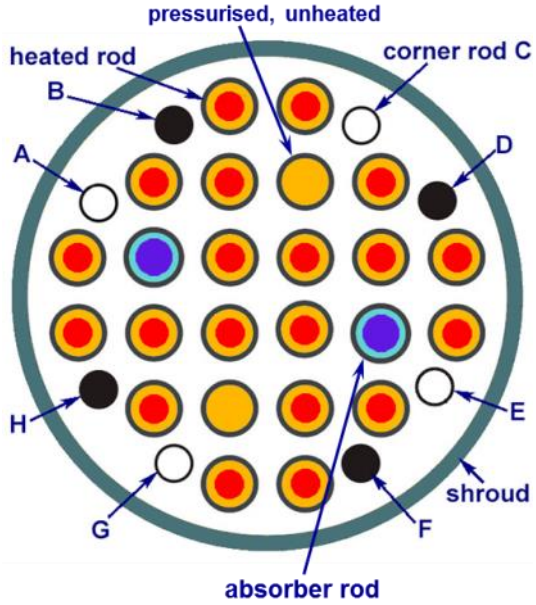
- Seven LB-LOCA experiments supported by German industry (VGB)
- Zircaloy-4, M5® (with and w/o pre-hydriding) and opt. ZIRLO™ claddings (with and w/o pre-hydriding)
- Conditions for secondary hydriding have been established and its influence on mechanical properties
- Final report published (still confidential for project partners)

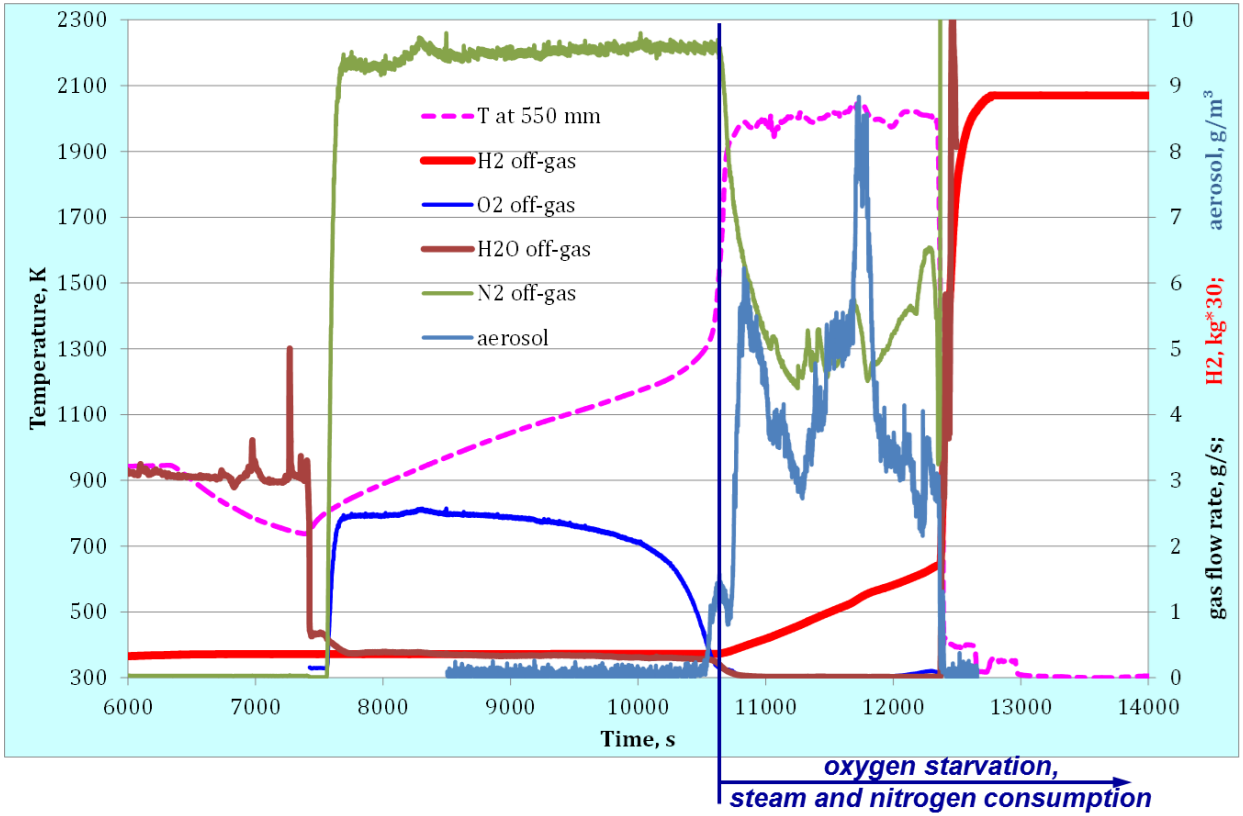




QUENCH-18

- Successfully conducted on 27 Sept 2017
- In the framework of the EC-China ALISA project
- With (i) two pressurized rods, (ii) two Ag-In-Cd absorber rods, and (iii) air ingress

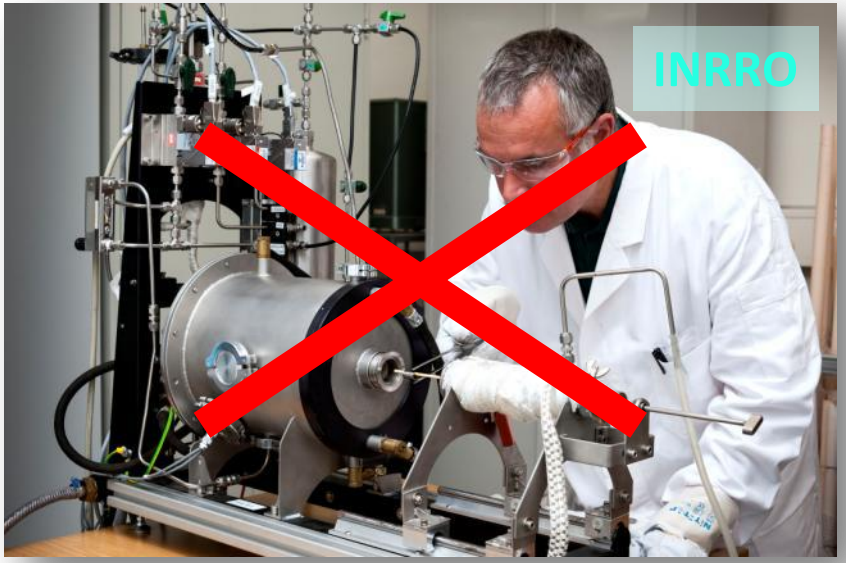




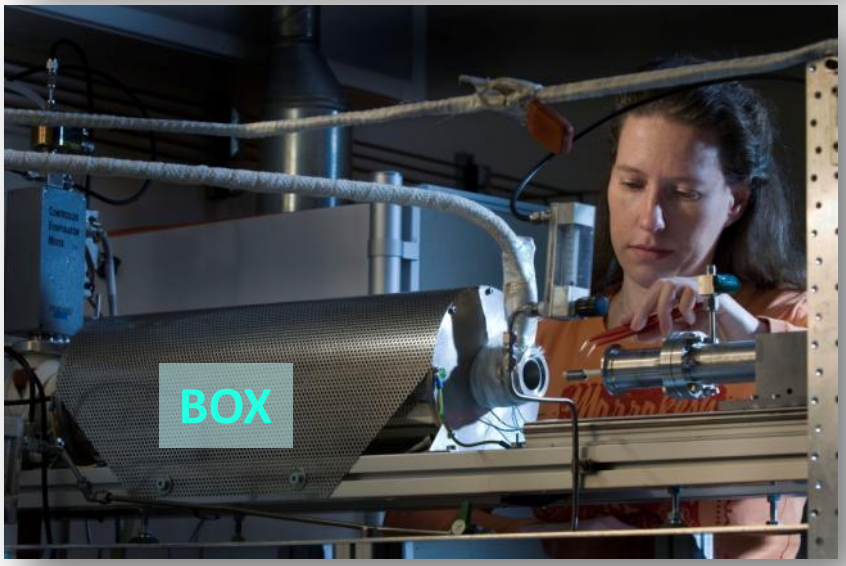
Separate-effects tests in 2017

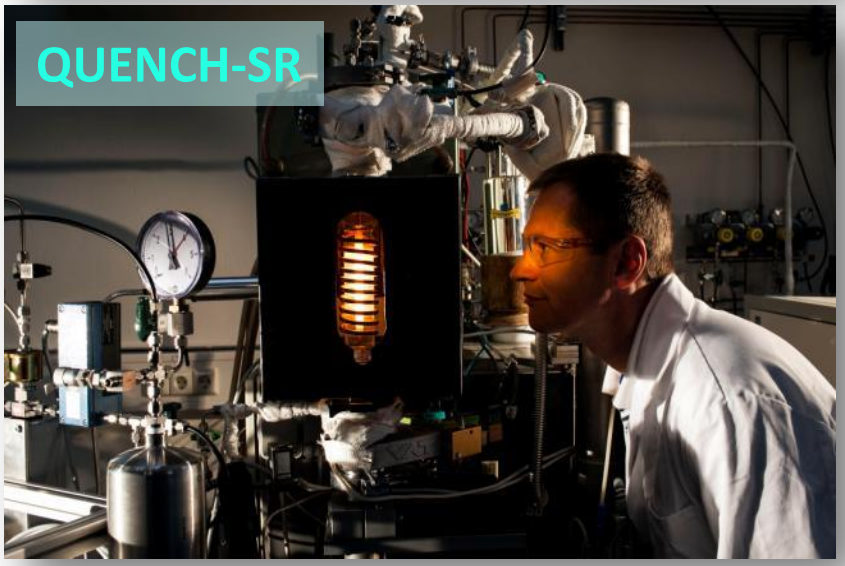
- Experiments on mechanism of air oxidation of Zr alloys
  - Chronology of oxide scale growth in various atmospheres
  - Low and high temperature limits for degrading effect of nitrogen on oxide scales
- Experiments on high-temperature oxidation of silicon carbide ceramic-matrix-composites in various atmospheres for different applications
- Synthesis of MAX phase coatings and high-temperature oxidation of these coatings as well as of bulk FeCrAl and MAX phases
- NR analyses of H_2 permeation through MAX phase coatings
- High-temperature oxidation of high-entropy alloys (HEA)
- Interaction between SiC and Zry

QUENCH Separate-effects tests: Main setups

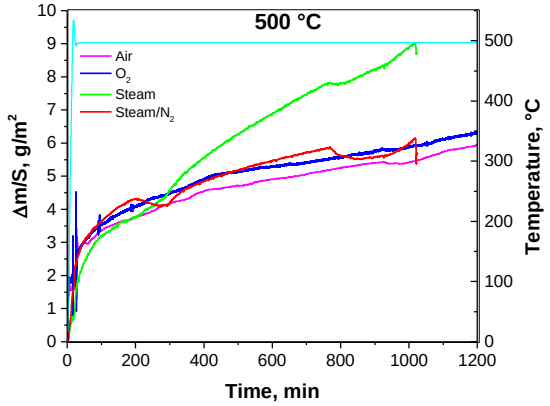


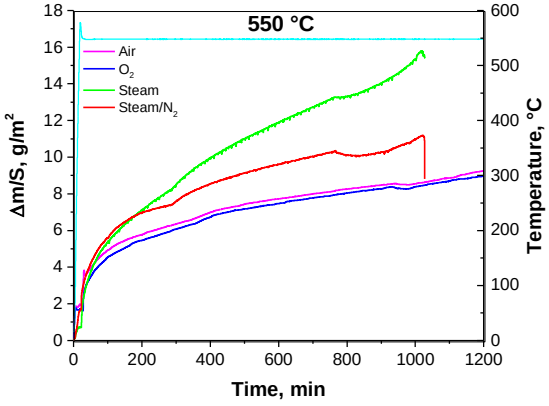


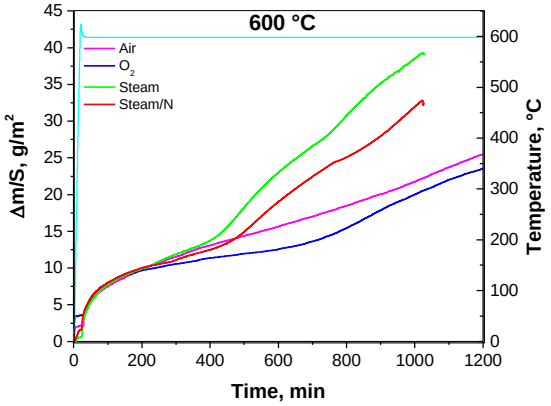


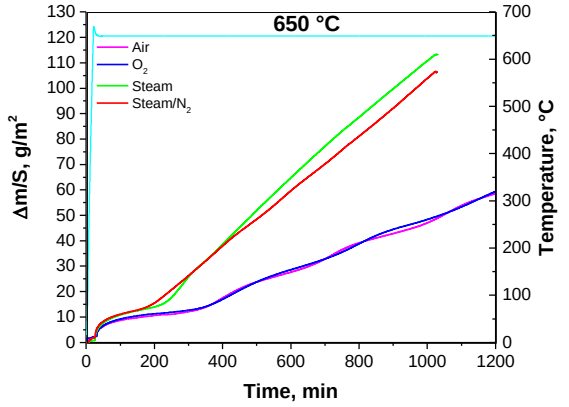


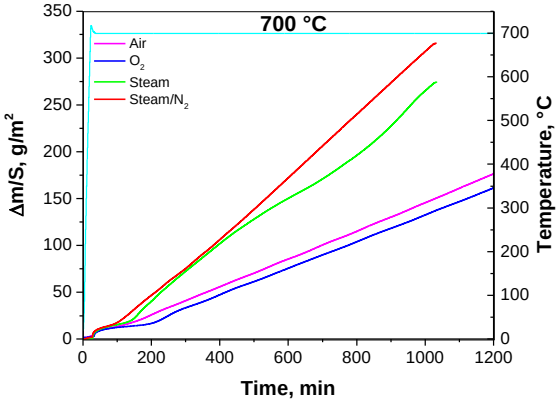
Effect of nitrogen on Zircaloy-4 oxidation at relatively low temperatures

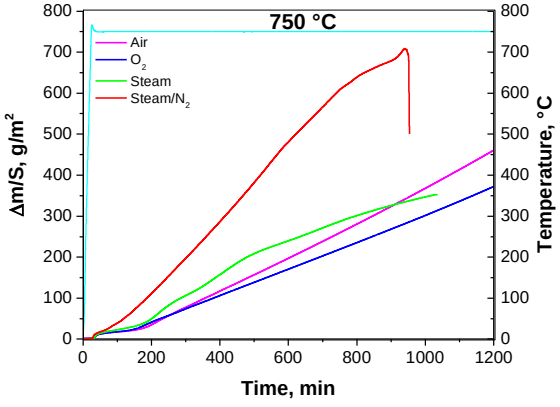










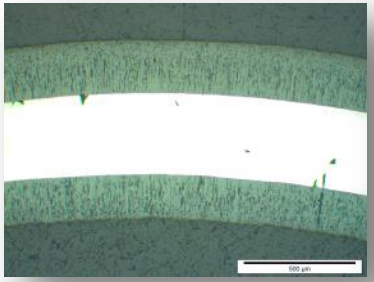


- At temperatures < 700°C nitrogen behaves like an inert gas
- At temperatures > 700°C nitrogen accelerates oxidation kinetics
- Earlier breakaway in steam compared to oxygen

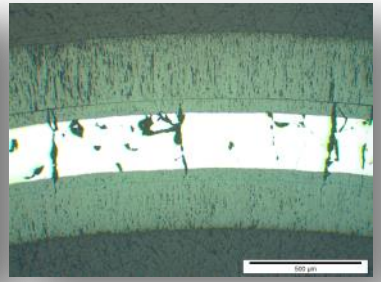
Effect of nitrogen on Zircaloy-4 oxidation at relatively high temperatures

Karlsruhe Institute of Technology

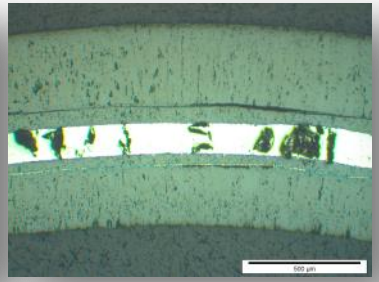
Oxygen



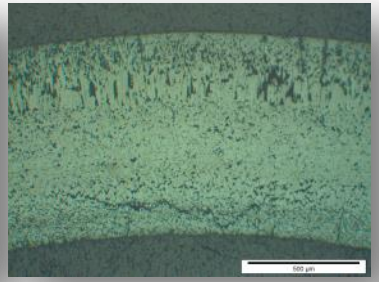
15 min, 1200°C



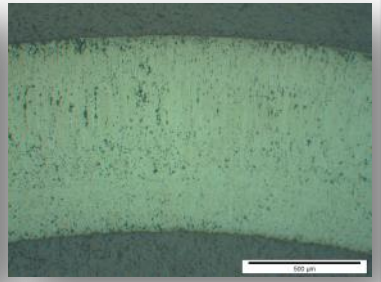
13 min, 1300°C



10 min, 1400°C

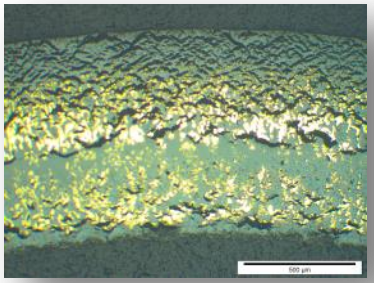


6 min, 1500°C

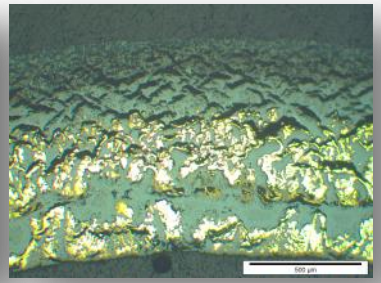


5 min, 1550°C

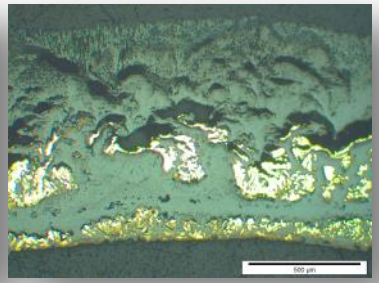
Air



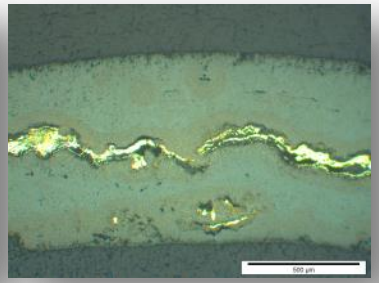
15 min, 1200°C



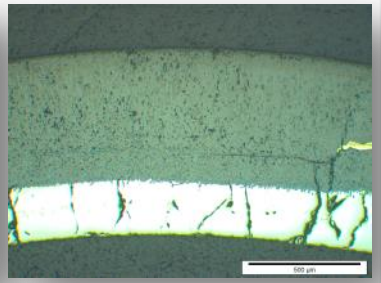
13 min, 1300°C



10 min, 1400°C



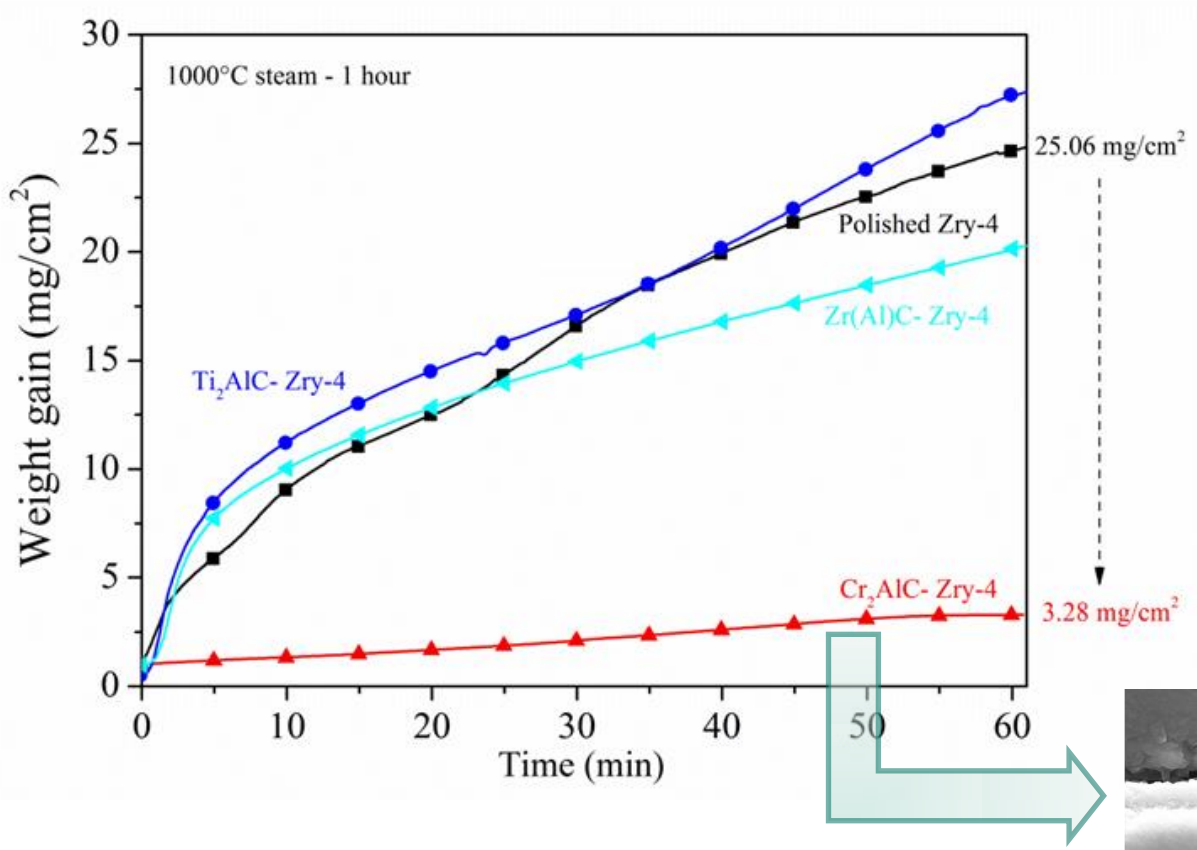
6 min, 1500°C



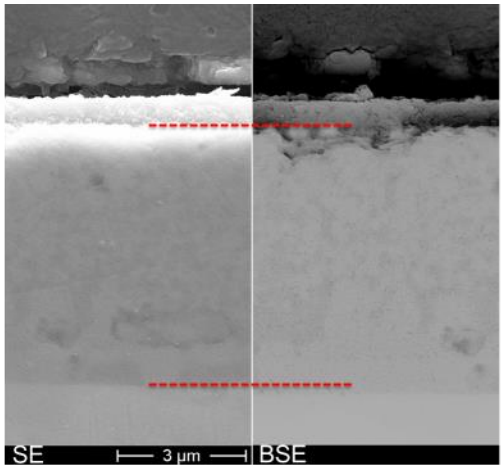
5 min, 1550°C

- Decreasing effect of nitrogen on the oxidation with rising temperatures

MAX phase coatings on Zry-4

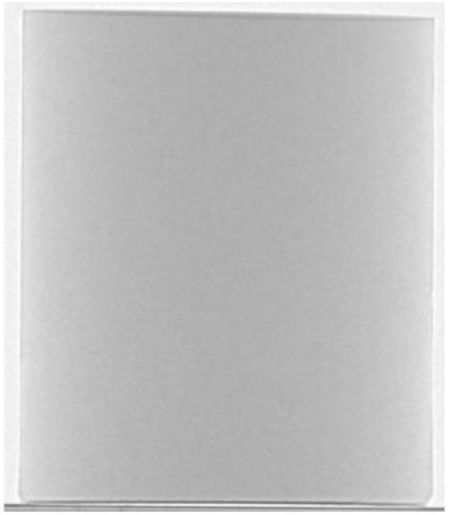


- Ti_2AlC
- Zr-Al-C
- Cr_2AlC

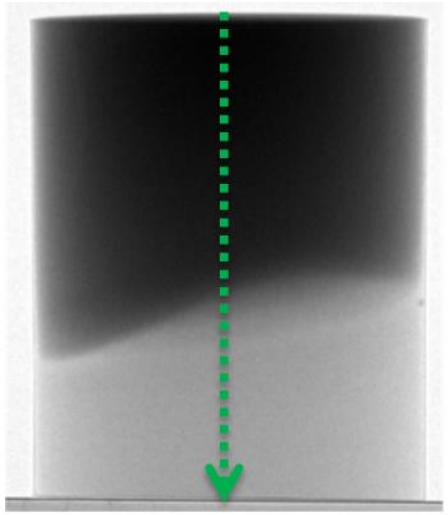


- Promising properties of Cr_2AlC coating

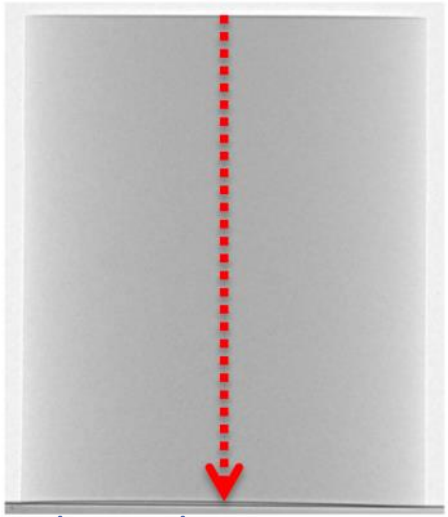
Hydrogen permeability of MAX phase coatings



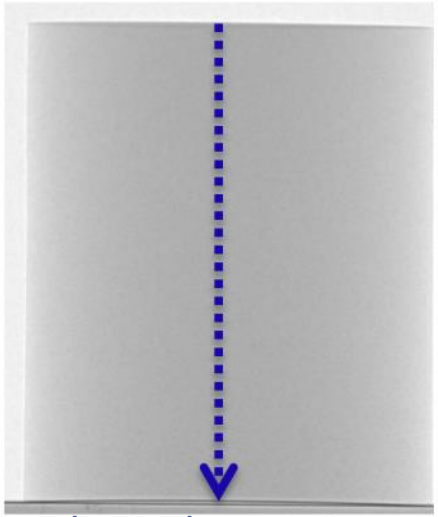
Zry-4 as received



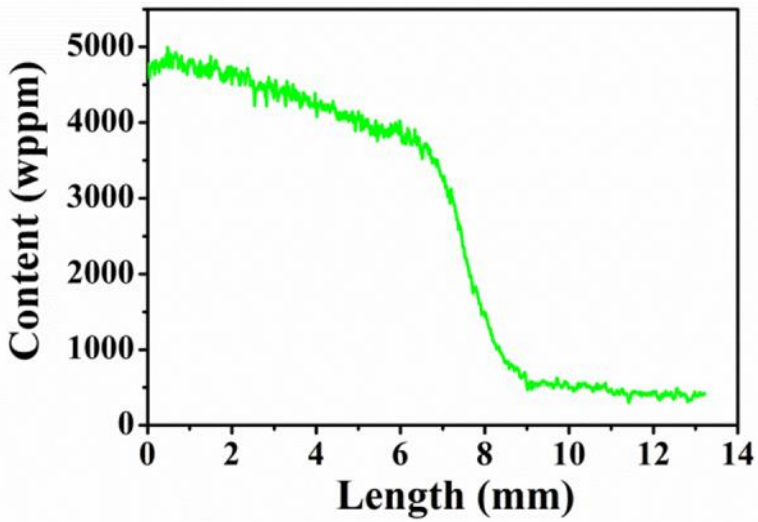
after annealing in Ar/H_2

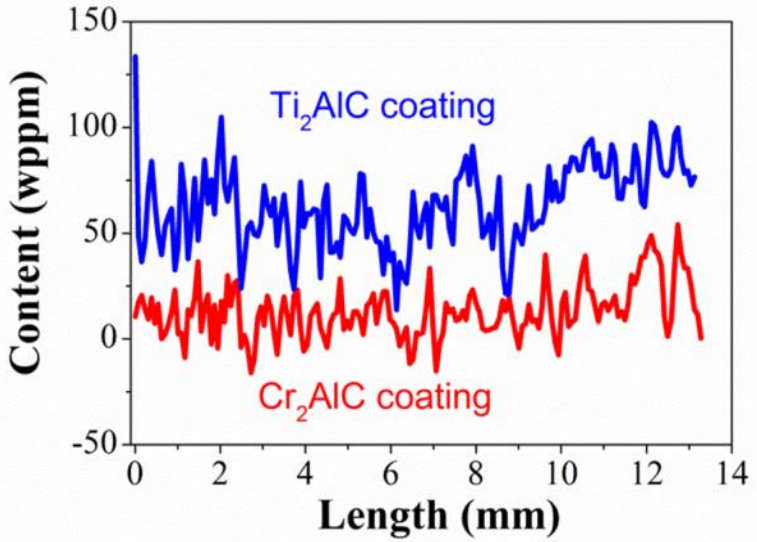


with Cr_2AlC coating
after annealing in Ar/H_2



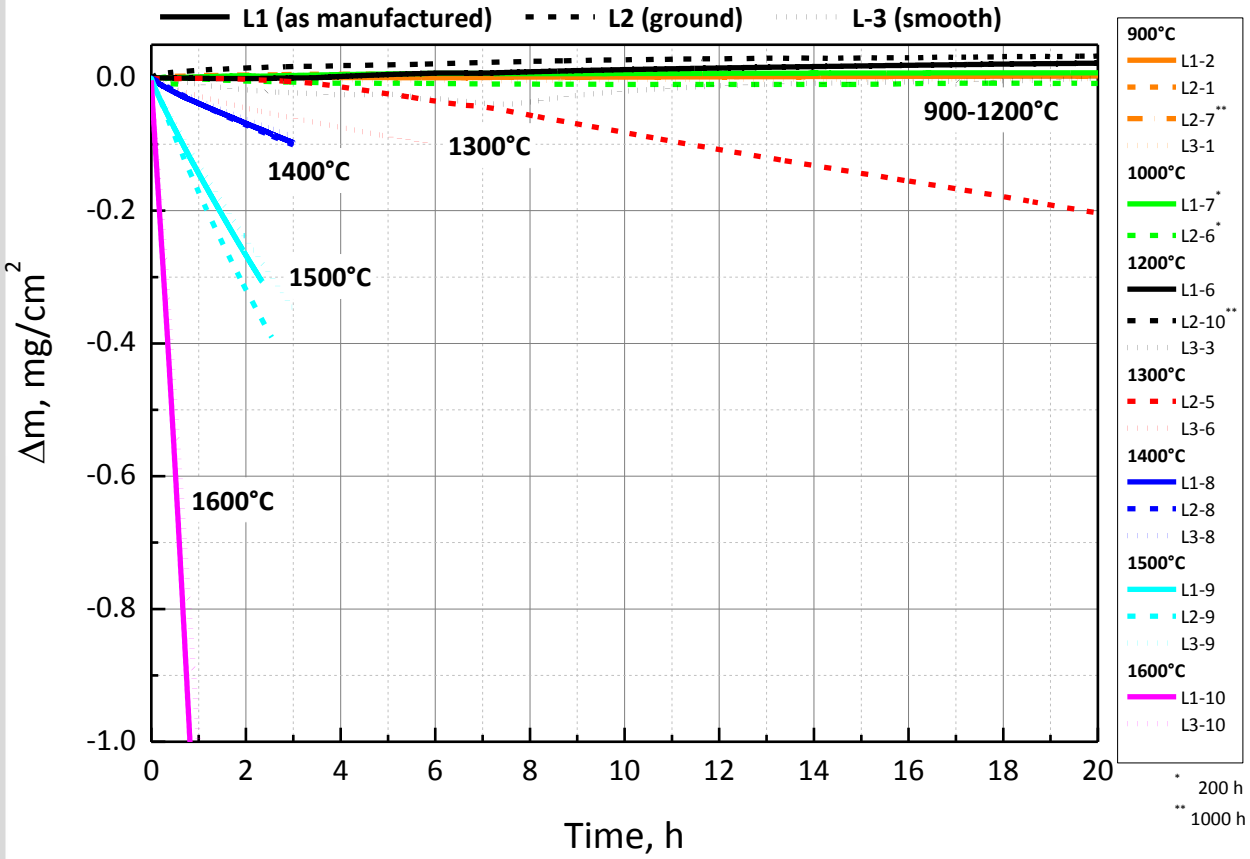
with Ti_2AlC coating
after annealing in Ar/H_2



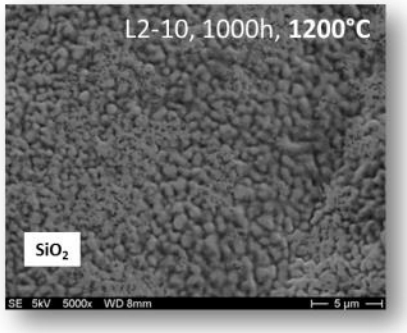


High-Temperature Oxidation of SiC Materials

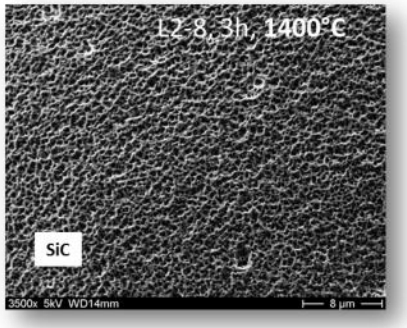
Oxidation of SiC-SiC_f GFR claddings in impure helium atmosphere



900-1200°C: Passive oxidation



1300-1600°C: Active oxidation



QUENCH activities for Accident Tolerant Fuel Claddings

- PhD theses
  - Joining of SiC components by RE alumosilicate glasses (finished)
  - Development of high-temperature resistant coatings for zirconium alloy cladding tubes
- Participation in the OECD-NEA Expert Group on Accident Tolerant Fuels for LWRs (EGATFL, final meeting 01/2018)
- Participation in the IAEA CRP on Accident Tolerant Fuel Concepts for Light Water Reactors (ACTOF)
- Partner of the EC project IL TROVATORE in the framework of HORIZON2020
- Partner in the CARAT project lead by Westinghouse, USA

Modelling and code validation

- QUENCH bundle tests are part of validation matrices of most SFD code systems
- Pre-test calculations for QUENCH-18 in the framework of the NUGENIA QUESA project by GRS, PSI, IBRAE, LEI, EdF
- Participation in the EC CESAM program on ASTEC validation (finished 03/2017)
- QUENCH data are used in the frame of IAEA FUMAC project
- RELAP5/SCDAPSIM analyses of various QUENCH tests
- Mechanistic model for zirconium oxidation in atmospheres containing nitrogen
- Separate-effects test data on air oxidation of Zr alloys are used by PSI, RUB, EdF, ISS and others for model development

Reporting

- QUENCH-LOCA Final report published *(but still restricted to partners)*
- QUENCH-L3/3HT/5: KIT Scientific Reports in preparation
- Papers, book chapters and conference contributions



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journal homepage: www.elsevier.com/locate/jnucmat

Oxidation of Zircaloy-4 in steam-nitrogen mixtures at 600–1200 °C

Martin Steinbrueck*, Fabio Oliveira da Silva, Mirco Grosse

Karlsruhe Institute of Technology, Institute for Applied M

ARTICLE INFO

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Available online 22 April 2017

1. Introduction

Zirconium alloys are worldwide used i
(LWR) as structure materials and fuel clad
cross-section for absorption of thermal neu
properties and high corrosion resistance at
However, oxidation becomes significant at
beyond 800–1200 °C as during accident
degradation of the mechanical properties
the release of hydrogen and heat. Indee
hydrogen causing the explosions during th
the Fukushima Daiichi plants was the zirc
The chemical energy released by this reactic
the residual decay heat a few hours after e
could strongly affect the accident progressi
temperature runaway during reflood of an
[1].

Most investigations of core degradatio
severe nuclear reactor accidents consider
components by steam only. However, there
with air having access to the core. Air in
shutdown conditions when the reactor coo

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Oxidation behavior of Ti_2AlC in the temperature range of 1400 °C–1600 °C in steam

Chongchong Tang*, Martin Steinbrück, Mirco Große, Hans Jürgen Seifert

Institute for Applied Materials (IAM), Karlsruhe Institute of Technology (KIT), D-76021

HIGHLIGHTS

- Breakaway oxidation of Ti_2AlC was observed in steam at 1400 °C after i
- The oxidation kinetics obeyed a linear law, showing non-protective effe
- The maximum tolerant temperature of Ti_2AlC in steam was approximat
- The growth and failure mechanisms of the scale were discussed.

ARTICLE INFO

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Available online 9 March 2017

Keywords:
Ti_3AlC MAX phase
Steam oxidation
High temperature
Microstructure

ABSTRACT

The oxidation behavior of
1400 °C–1600 °C. The ox
then tended to be a linea
exposure up to 24 h at 150
isolated islands with large
layer with a thin discontin
diffusion of Ti and C throu
segregation of TiO_2 at the
The scale adhesion was re
generation of voids at the
the stresses contribute to t
1500 °C. The sample was
accompanied by release o
was approximate 1555 °C.

1. Introduction

Zirconium alloys have been used in current light water reactors (LWRs) as fuel claddings and other structural materials for several decades due to the low thermal neutron capture cross section, outstanding mechanical performance and reasonable corrosion resistance in hot water [1]. Optimized performances of nowadays nuclear grade zirconium alloys have been achieved within decades of research and operation under normal operation conditions. However, the satisfactory performance of zirconium alloys is

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Annals of Nuclear Energy 101 (2017) 347–358

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Annals of Nuclear Energy

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Experiments on silver-indium-cadmium control rod failure during severe nuclear accidents

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ARTICLE INFO

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Keywords:
Silver-indium-cadmium alloy
Control rod
Degradation
Failure
QUENCH

ABSTRACT

A large-scale bundle experiment w
well as a series of small-scale single
A variety of parameters like tempe
possibility of inner oxidation of the
modes from local melt-through to
interaction between Ag-In-Cd mel
temperature of the steel. The cladd
identified various phases formed du
rials. Furthermore, solidus and liqui
by DSC.

1. Introduction

Silver-indium-cadmium alloy with the composition 80 wt% Ag, 15 wt% In, and 5 wt% Cd is used as neutron absorber material in control rods (CR) of many Pressurized Water Reactors (PWR). It is the material with the lowest melting temperature (approx. 1100 K) among all metallic and ceramic materials applied in nuclear reactors, and hence will melt first during design basis and beyond design basis nuclear accidents. In most reactors, the Ag-In-Cd alloy is encased in stainless steel (SS) cladding tubes surrounded by guide tubes made of Zircaloy. Depending on the reactor size 2500–5000 kg of Ag-In-Cd absorber alloy is used in a PWR.

The general failure mechanism of Ag-In-Cd control rods during high-temperature transients has been already described in the overview papers by Petti (1989), Hofmann (1999), Lewis et al. (2008), and De Luze et al. (2013). The absorber rod alloy is thermodynamically stable with its stainless steel cladding, even in the liquid state. All binary phase diagrams between the main stainless steel components (Fe, Cr, Ni) and Ag, In, and Cd show neither formation of compounds nor any eutectic interaction, and only very limited mutual solubility of the elements at higher temperatures (Massalski et al., 1990). However, the absorber rod guide tube made from Zircaloy chemically interacts with the stainless steel (SS) cladding of the absorber rod. A solid-state contact between

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journal homepage: www.elsevier.com/locate/nucengdes

Validation of ASTECV2.1 based on the QUENCH-08 experiment

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[c] Institut de Radioprotection et de Sûreté Nucléaire (IRSN), Nuclear Safety Division/Safety Research/Severe Accident Department, Saint Paul Lez Durance 13115, France

HIGHLIGHTS

- ASTECV2.1 can reproduce QUENCH-08 experimental trends e.g. hydrogen generation.
- Radial temperature gradient and heat transfer through argon gap are underestimated.
- Mesh sizes lower than 55 mm needed to capture the strong axial temperature gradient.
- Minor variations of external electrical resistance strongly affect bundle heat-up.
- Modelling of a bypass and inclusion of currents partially overcome discrepancies.

ARTICLE INFO

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Keywords:
Code validation
Severe accident
ASTEC
QUENCH
Reflooding

ABSTRACT

The Fukushima accidents have shown that further improvements of Severe Accident Management Guidelines (SAMGs) are still necessary. Hence, the enhancement of severe accident codes and their validation based on integral experiments is pursued worldwide. In particular, the capabilities of the European integral severe accident ASTECV2.1 code are being extended within the CESAM project through the improvement of physical models, code numerics and an extensive code validation.

Among the different strategies encompassed in the plant SAMGs, one of the most important ones to prevent core damage is the injection of water into the overheated core (reflooding). However, under certain conditions, reflooding may trigger a sharp hydrogen generation that may jeopardize the containment. Within this work, ASTECV2.1 models describing the early in-vessel phase of the severe accident and its termination by core reflooding are validated against data from the QUENCH test facility. The QUENCH-08, involving the injection of 15 g/s (about 0.6 g/s/rod) of saturated steam at a bundle temperature of 2073 K, has been selected for this comparison.

Results show that ASTECV2.1 is able to reproduce the experimental temperatures and oxide thicknesses at representative bundle locations. The predicted total hydrogen generation (76 g) is similar to the experimental one (84 g). In addition, the choices of an axial mesh size lower than 55 mm and of an external electrical resistance of a 7 mΩ/rod have been justified with parametric analyses. Finally, new modelling options are introduced to overcome some discrepancies present in the reference case and to cancel the effect of the external electrical resistance in the QUENCH model. Results show that the predicted hydrogen generation during quenching (38 g) is closer to the experimental one (37 g) than the reference case, giving a better reproduction of the radial temperature gradient within the hottest zone.

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1. Introduction

In the Nuclear Power Plants (NPPs) operated today in Europe, the Defence-in-Depth concept is applied to ensure that public and environment are protected from a harmful release of radioactive material. Within this safety concept, measures to

Abbreviations: FRS, Fuel Rod Simulator; LWR, Light Water Reactor; NPP, Nuclear Power Plant; SAM, Severe Accident Management; SAMGs, Severe Accident Management Guidelines; Zry, Zircaloy.
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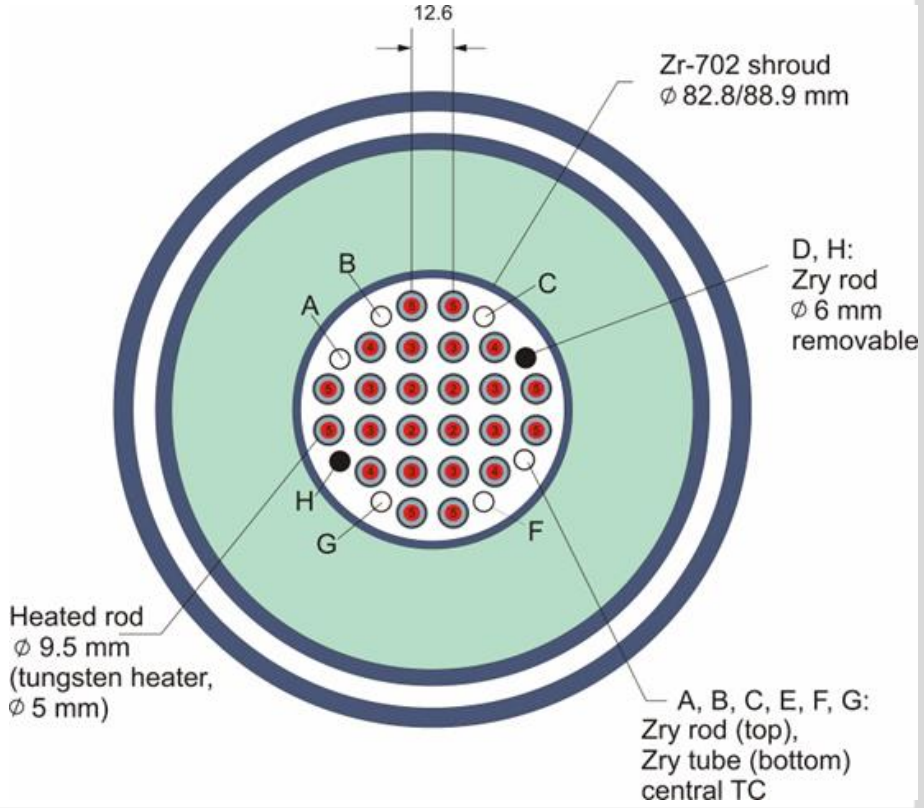
http://dx.doi.org/10.1016/j.nucengdes.2016.12.039
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Outlook 2018-2019

- QUENCH bundle experiment with FeCrAl claddings planned for beginning of 2018 in cooperation between ORNL and KIT
- BWR bundle test with in the framework of the EC SAFEST program in 2018
- CODEX bundle experiment on air ingress in the CODEX facility planed for 02/2018 (proposed by KIT)
- Discussion with various international partners on bundle tests with ATF claddings
- SETs on various further topics (ATF cladding high temperature oxidation, partially in the framework of the H2020 project IL TROVATORE and of the IAEA CRP ACTOF as well as in bilateral cooperation with Westinghouse)
- Activities on safety of long-term dry intermediate storage

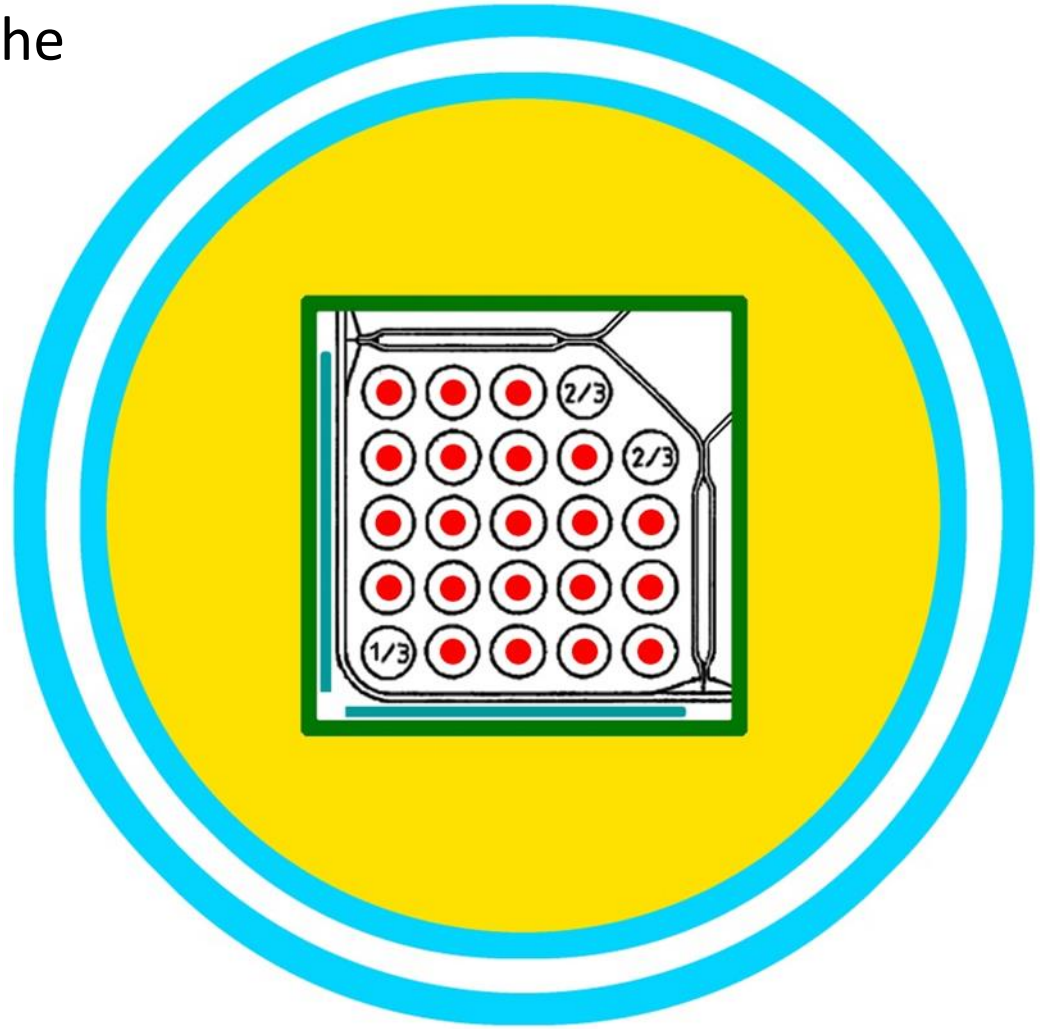
QUENCH bundle experiment with FeCrAl claddings

- In cooperation with ORNL
- Scenario similar to QUENCH-15 (same bundle geometry, same electrical power input)
- To be conducted beginning of 2018
- FeCrAl materials delivered by ORNL to KIT



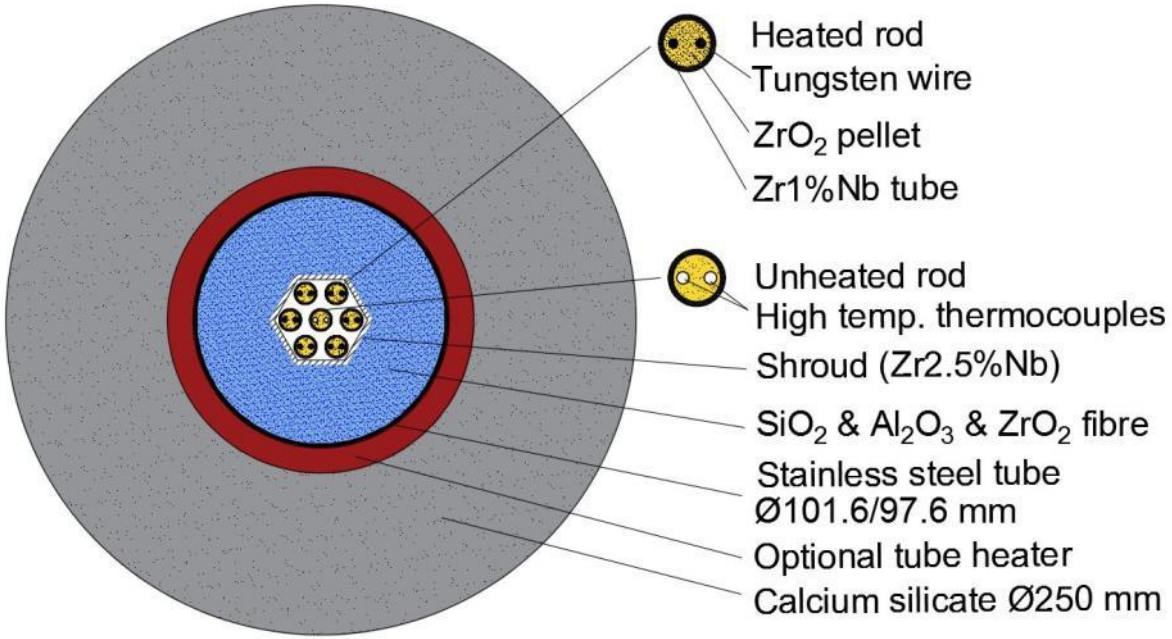
BWR bundle test

- Planned to be conducted in 2018 in the framework of the EC SAFEST program
- Proposed by Swedish regulatory body and supported by Westinghouse
- Square bundle cross section
- Including boron carbide absorber blades and water channels
- Scenario tbd.



CODEX air ingress experiment

- In the framework of the EC supported SAFEST program
- Proposed by KIT and conducted by MTA Budapest (02/2018)
- Smaller, 1-m long 7-rod bundle
- Including steam-air phase
- Including final cool-down phase in argon



Co-operations

Programs

- NUGENIA
- HORIZON 2020
- IAEA
- OECD-NEA

Bilateral

- PSI
- MTA EK
- IRSN, CEA
- RUB-LEE, IKE
- JRC
- GRS
- VGB, AREVA, EdF
- Westinghouse
- USNRC
- UNIST
- NECSA, BAM, HMI
- NRA, JAEA
- ISS
- ORNL
- Various Chinese Organizations





















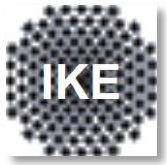


























Acknowledgement

- Helmholtz Association for funding program NUSAFE at KIT
- Program NUSAFE and IAM institute's management for broad support of our activities
- VGB for supporting QUENCH-LOCA test series

- And last but not least the QUENCH team:
  J. Glaser, M. Heck, J. Laier, J. Moch, H. Muscher, U. Peters, A. Pshenichnikov, C. Rössger, U. Stegmaier, C. Tang, P. v. Appeldorn, M. Walter

Lessons learned from the QUENCH-LOCA experiments

J. Stuckert, M. Große, M. Steinbrück, M. Walter, A. Wensauer

Seven QUENCH-LOCA bundle tests (QUENCH-L0…-L5 and -L3HT) with different cladding materials (Zry-4, M5®, Opt. ZIRLO™) were performed according to a temperature/time-scenario typical for a LBLOCA in German PWRs. For two tests (QUENCH-L4 and -L5) pre-hydrogenated claddings (with 100 and 300 wppm H for M5® and Opt. ZIRLO™, correspondingly) were used. Generally, a peak cladding temperature of about 1350 K (1250 K for QUENCH-L5) was reached at the end of the heat-up phase at the bundle elevation of 950 mm. The maximal heat-up rate was 8 K/s (2.6 K/s for commissioning test QUENCH-L0); the cooling phase lasted about 120 s (0 s for QUENCH-L0) and was terminated by 3.3 g/s/effective rod water flooding. The tangential temperature gradient across a rod was between 30 and 70 K on the burst onset.

The detailed profilometry measurements performed over whole length of the claddings showed formation of not only main ballooning area (with burst) but also, for several rods of each bundle, additional two or three ballooning regions. Another parameter calculated based on profilometry measurements is the bundle blockage. The maximal coolant channel blockage was less than 35%. Due to the moderate blockage good bundle coolability was kept for all bundles.

Cladding wall thinning from 725 to 350 µm due to ballooning was observed at the burst side along 50 mm below and above burst opening. The maximal oxide thickness at the outer cladding surfaces was less than 15 µm. Surface cracks, penetrating both layers, were formed in vicinity of burst opening during ballooning. Oxide layer formed after the burst at the inner cladding surface around the burst opening with a thickness of about 15 µm decreasing to 3 µm at a distance of about 20 mm from the burst opening.

The hydrogen, produced during oxidation of the inner cladding surface around the burst opening, can be absorbed by the metal with formation of hydrogen enrichments around the oxidized area (secondary hydrogenation). Such enrichments with a quite complex 3D form were observed for inner rods having been exposed to peak cladding temperatures of more than 1200 K. Except for the QUENCH-L0 test, no hydrogen bands were observed for bundle outer rods, the peak cladding temperature measured for these rods was less than 1200 K. Neutron tomography analyses showed (except the QUENCH-L0 bundle with non-prototypical long duration of the transient stage and the QUENCH-L3HT with not prototypical high temperatures) that the maximal hydrogen concentrations inside the hydrogen bands was less than 750 wppm (averaged through the cladding cross section). EBSD analysis showed that a part of the hydrogen absorbed inside the claddings formed the hydrides with µm-sizes, which are distributed in the matrix intra as well inter granular. Another part of the absorbed hydrogen was probably dissolved in the metallic matrix.

During quenching, following the high-temperature test stages, no fragmentation of claddings was observed meaning that the residual strengths and ductility were sufficient.

Tensile tests at room temperature evidenced fracture at hydrogen bands for several inner rods with local hydrogen concentrations about 1500 wppm and more. Claddings with lower hydrogen concentrations fractured due to stress concentration at burst opening edges. Other tensile tested claddings failed after necking far away from burst region.



Karlsruhe Institute of Technology

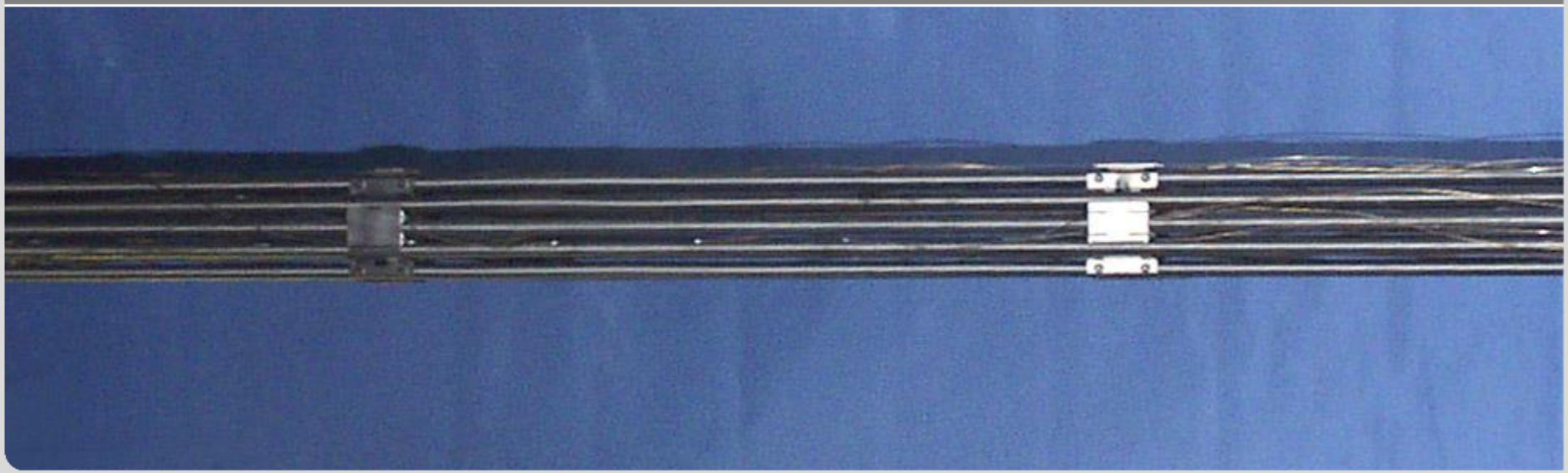
IAM
Institute for Applied Materials

Lessons learned from the QUENCH-LOCA experiments

KIT: J. Stuckert, M. Große, M. Steinbrück, M. Walter,
VGB: A. Wensauer

QWS-23, Karlsruhe 2017

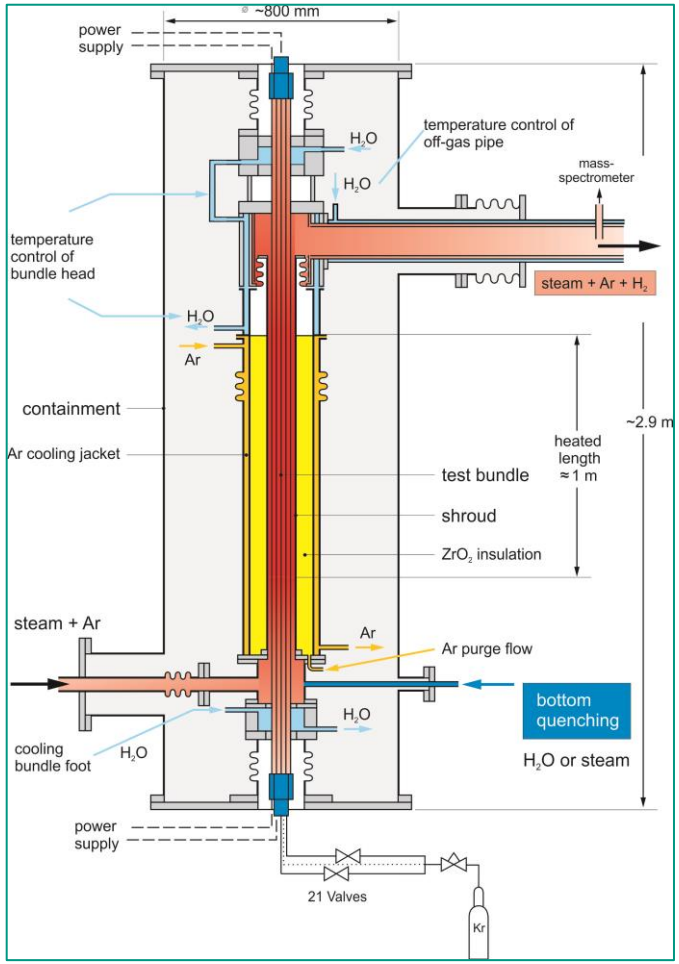
Institute for Applied Materials; Program NUKLEAR



QUENCH-LOCA Test Matrix 2010 – 2016

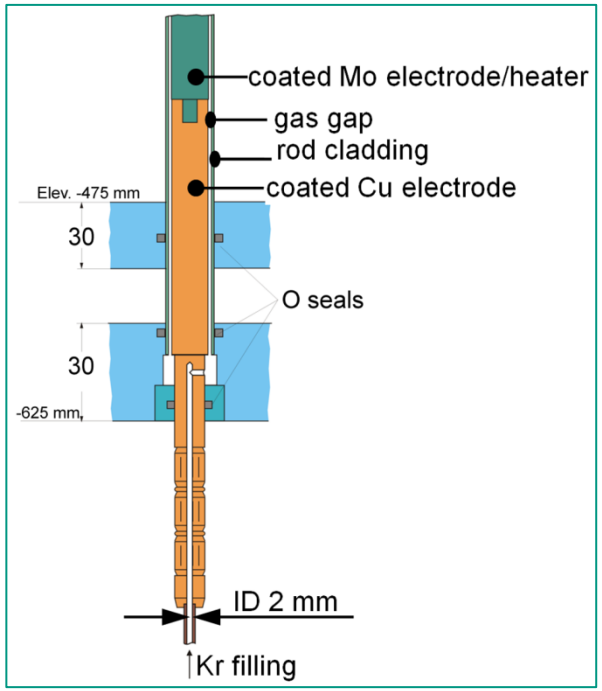
Test	Type of cladding tubes	Max heat-up rate, K/s	Max T on the end of heat-up, K	Duration of inner clad. oxidation at T > 1123 K, s	Secondary cladding hydro-genation	Post-test fracture due to second. hydro-genation
Commissioning QUENCH-L0 July 22, 2010	Zry-4	2.5	1350	110	yes	yes
Reference QUENCH-L1 Feb. 02, 2012	Zry-4	7	1373	105	yes	no
QUENCH-L2 July 30, 2013	M5®	8	1373	88	yes	no
QUENCH-L3HT March 21, 2014	Opt. ZIRLO™	8	1623	252	yes	yes
QUENCH-L4 July 30, 2014	Pre-hydr. M5® (100 wppm H)	8	1385	100	yes	yes
QUENCH-L3 March 17, 2015	Opt. ZIRLO™	8	1346	84	yes	no
QUENCH-L5 Feb. 17, 2016	Pre-hydr. opt. ZIRLO™ (300 wppm H)	8	1257	45	no	-

Hardware elements

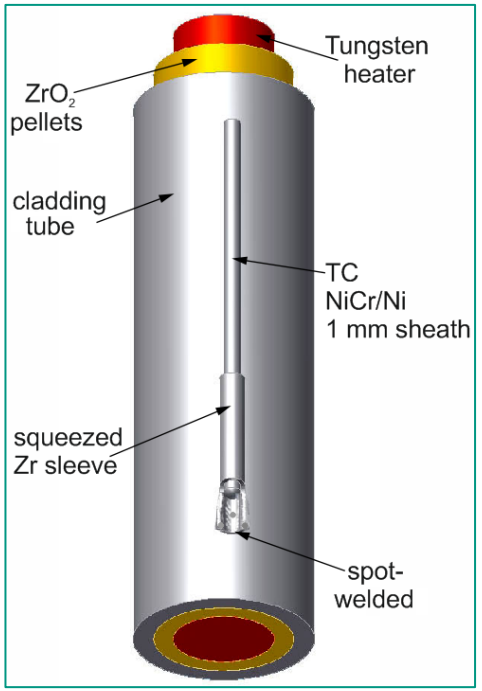


QUENCH facility:

test section with flow lines

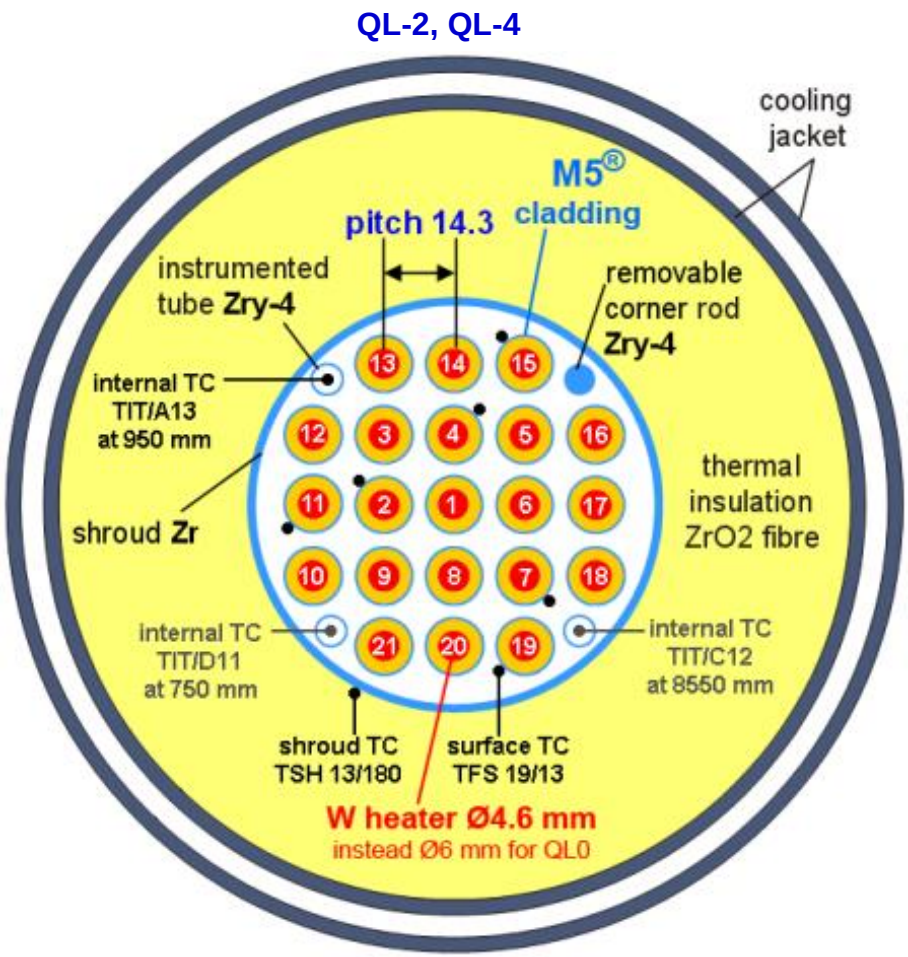


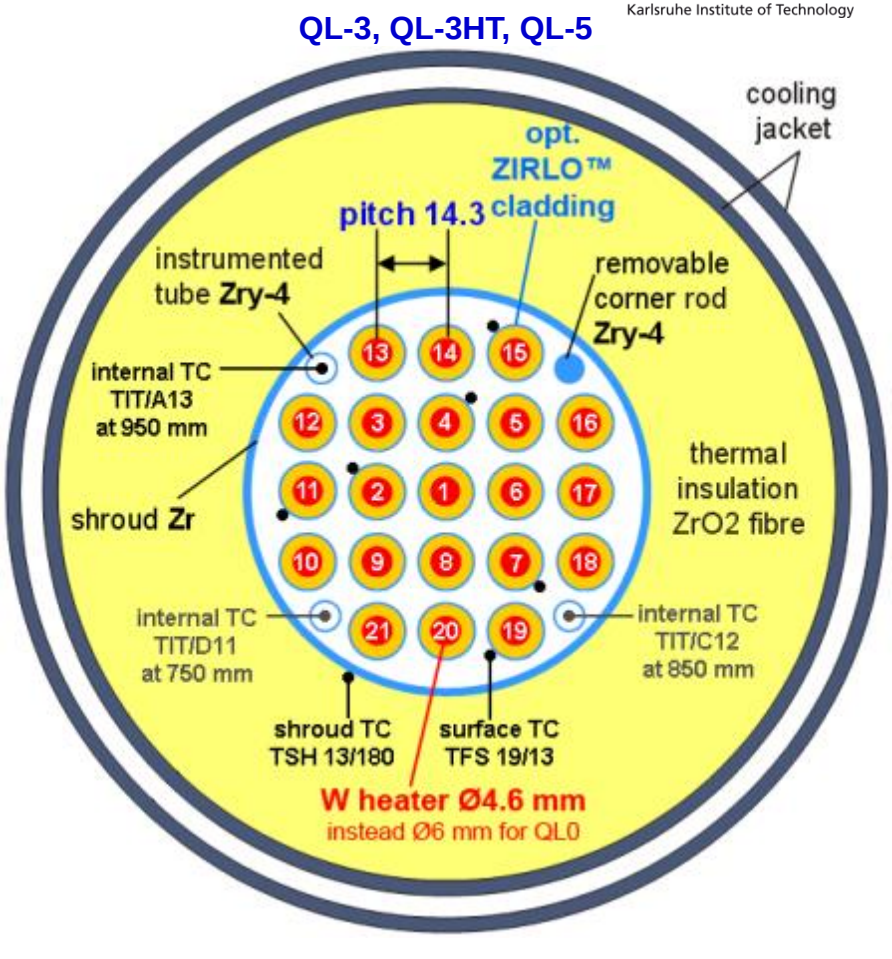
Rod pressurization from bottom of each rod



TC fastening at the test rod

Cross-sections of the QUENCH-L2 … -L5 bundles



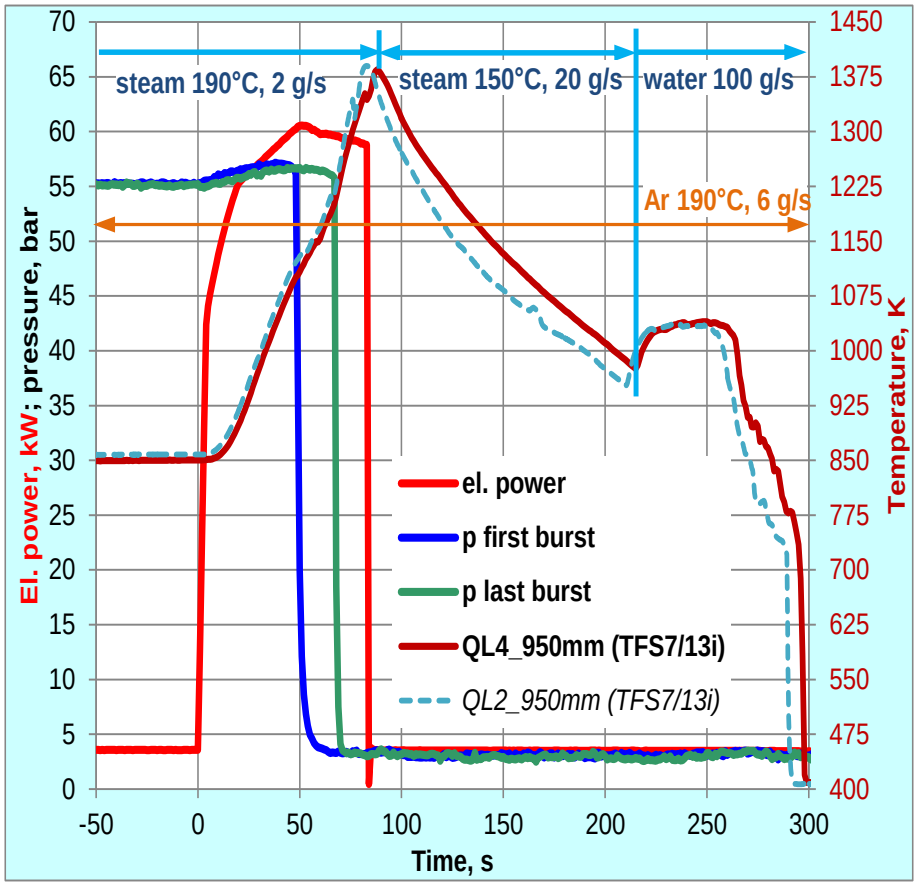


1) The use of ***tungsten*** heaters with smaller diameter (***4.6 mm***) instead tungsten heaters (QUENCH-L0) or tantalum heaters (QUENCH-L1) with diameter of 6 mm has allowed to reach a **higher heat rate**.

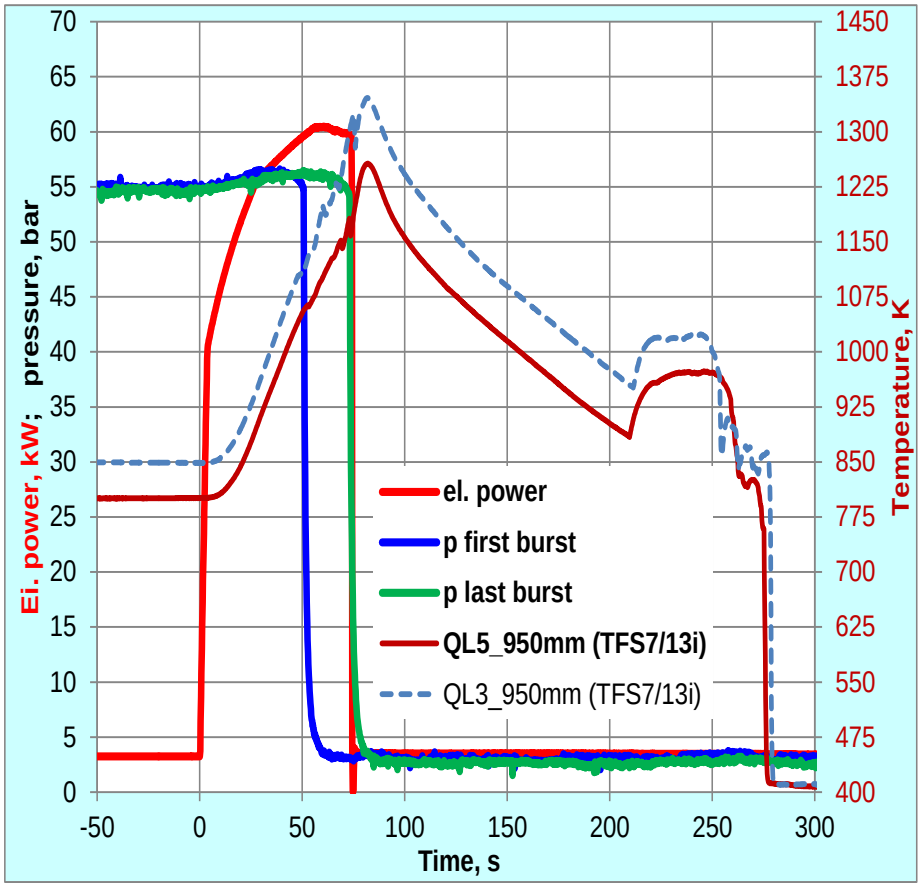
2) All rods are filled with Kr with p=55 bar at T_{pct}=800 K (similar to QUENCH-L1).

Test progresses

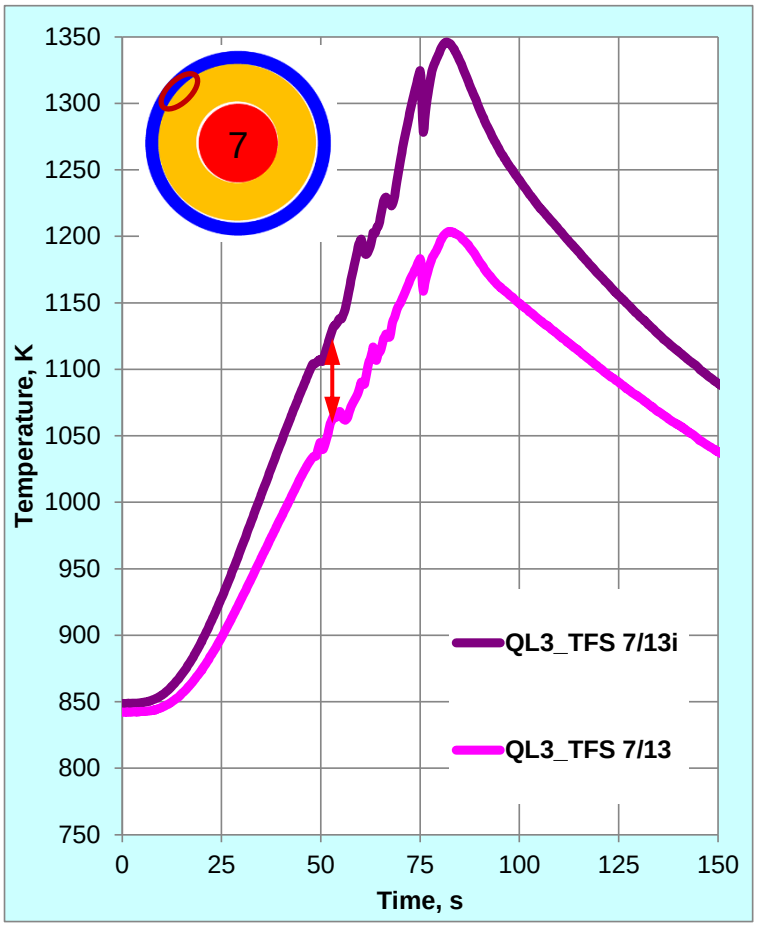
M5: QL-2, QL-4

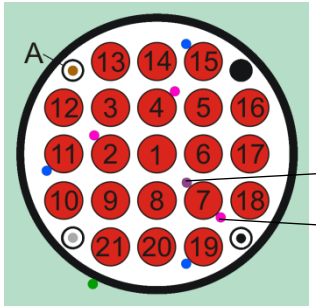


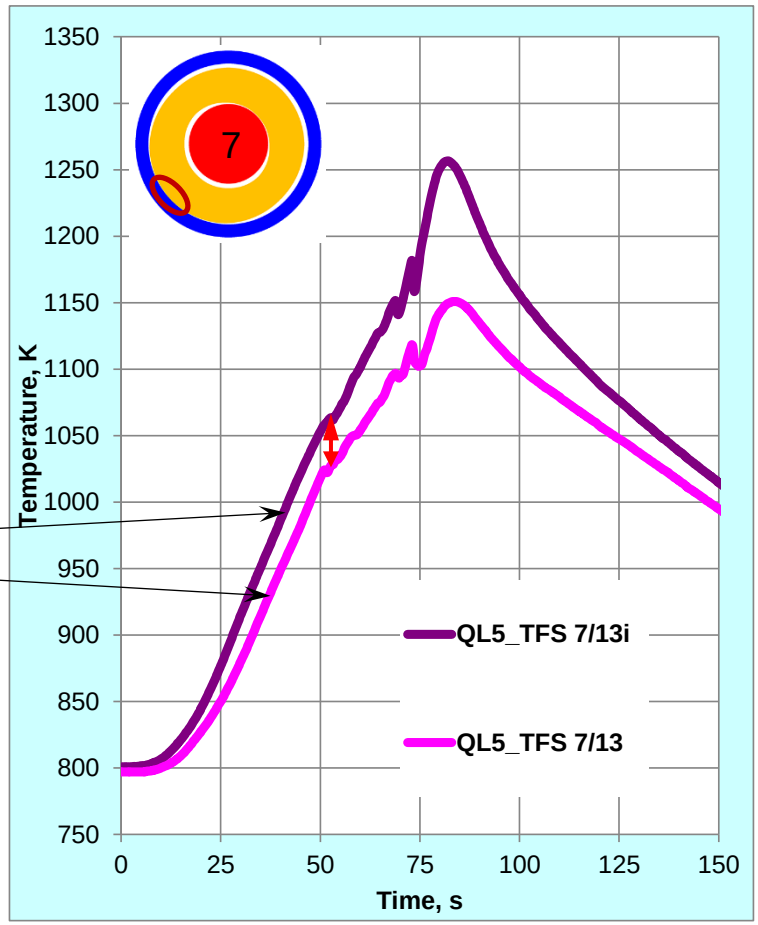
Opt. ZIRLO: QL-3, QL-5



Dependence of radial temperature difference (for rod #7 of QL3 and QL5 bundles) from position of contact between pellet and cladding



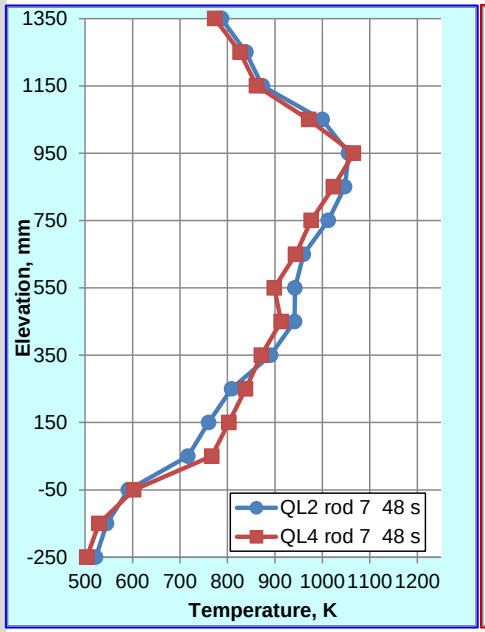


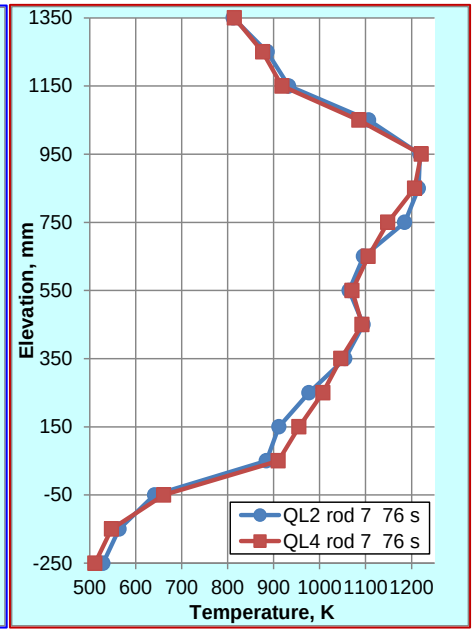


QL-3 **QL-5**

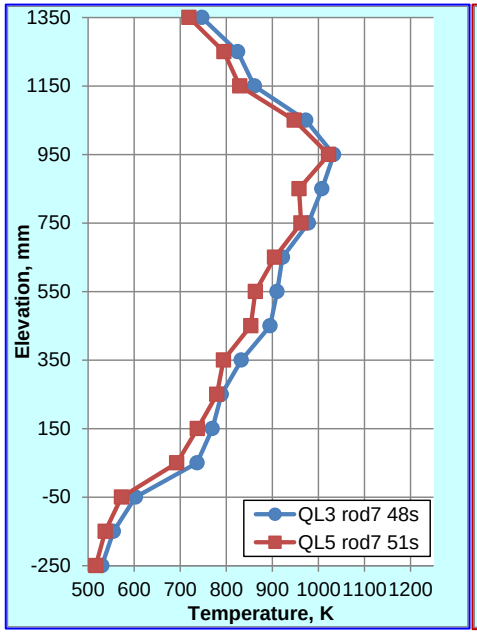
Superposition of two temperatures gradients:
global bundle gradient (from shroud to central rod) and local gradient inside rod

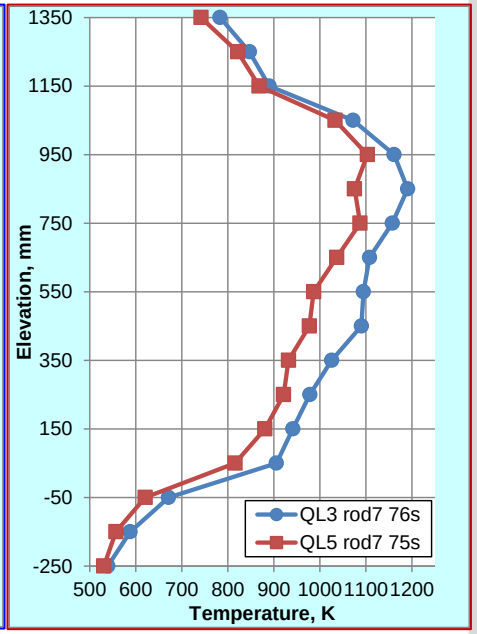
Axial temperature profiles on first cladding burst and end of transient





QL-2 and QL-4

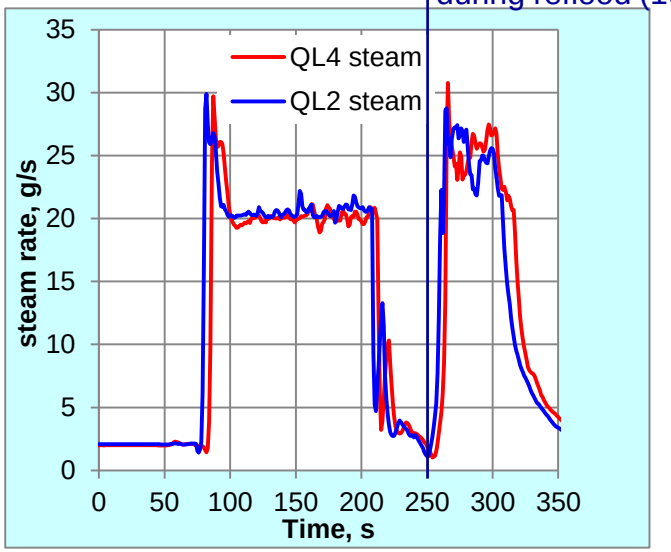


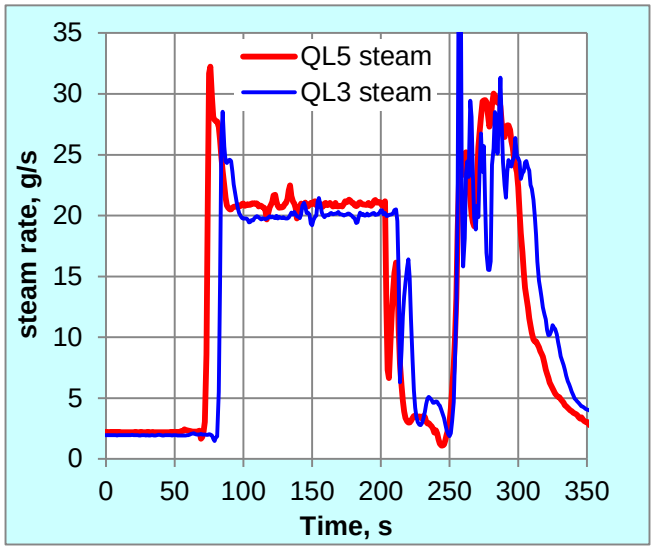


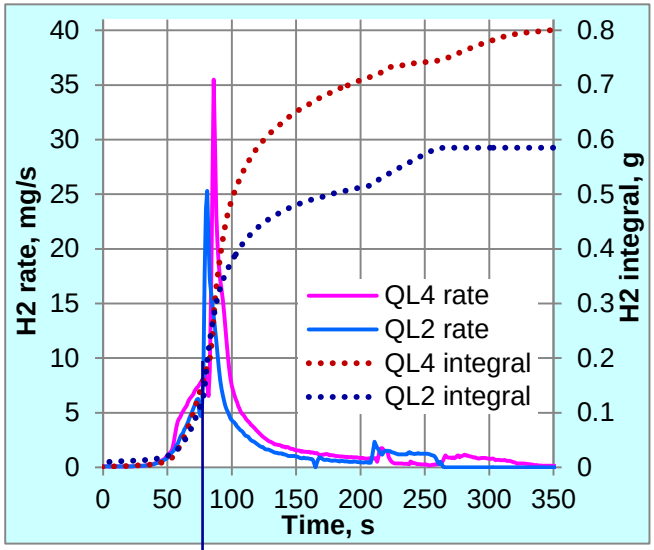
QL-3 and QL-5

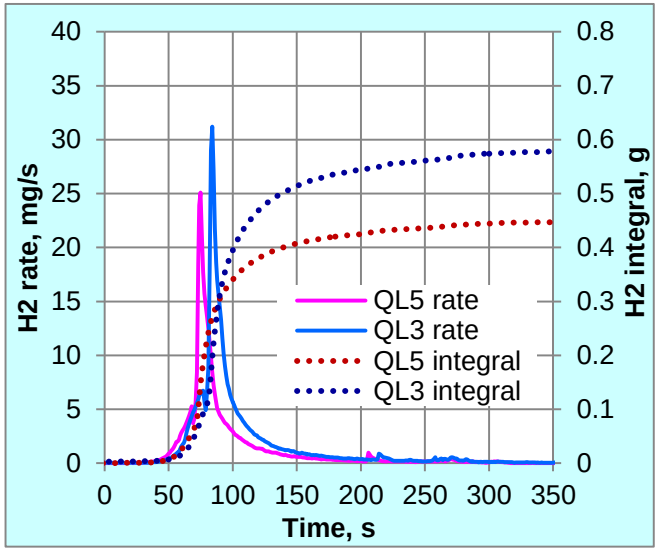
Off-gas mass spectrometer measurements: steam and H_2

steam release (25 g/s)
during reflood (100 g/s water)









H_2 increase during heat-up

H_2 peak at initiation of cool-down in 20 g/s steam

Post-test views of different bundles in the burst region



QL-1, view at 180°



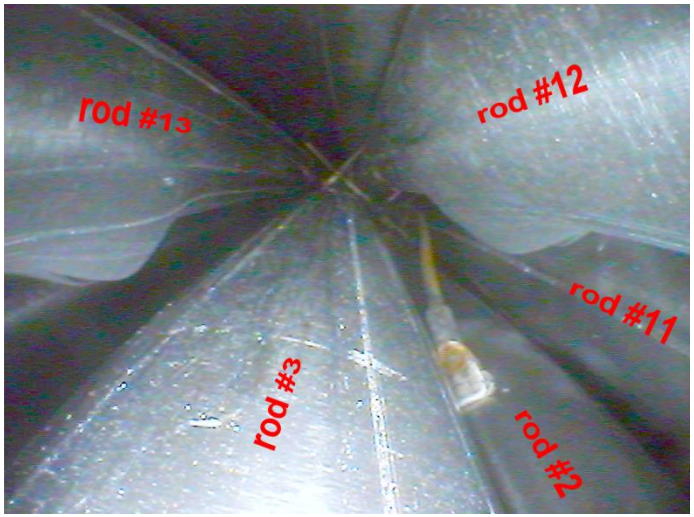
QL-2, view at 270°

Increased bending of QL-0, QL-1, QL-2 and QL-3HT rods due to limited axial heater expansion

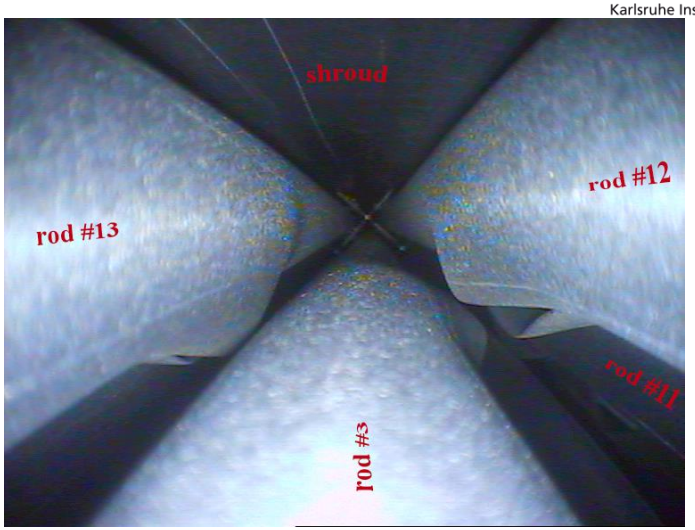


QL4, typical view for all QL-3, -4, -5 bundle positions

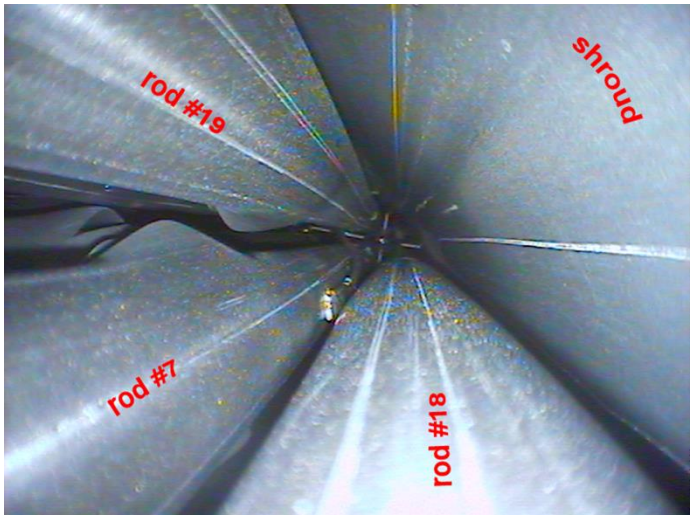
Videoscope observations with camera inserted from the bundle bottom at positions of corner rods



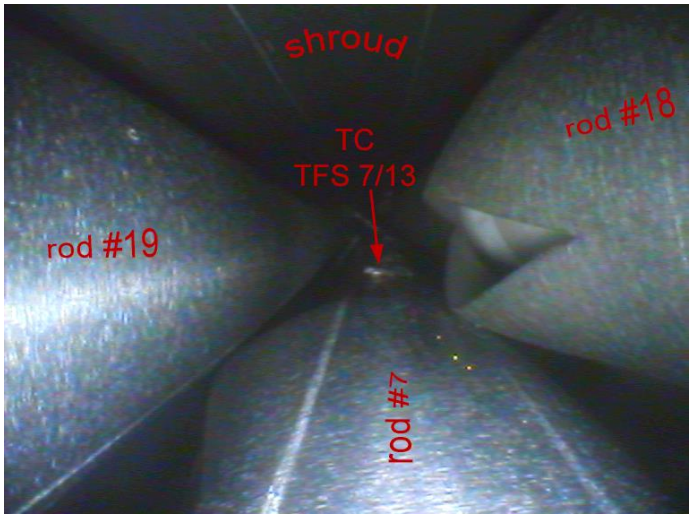
QL-2, at corner rod A



QL-3, at corner rod A

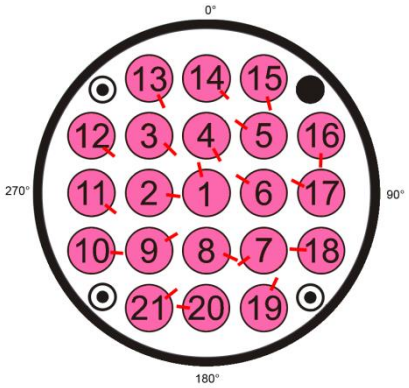


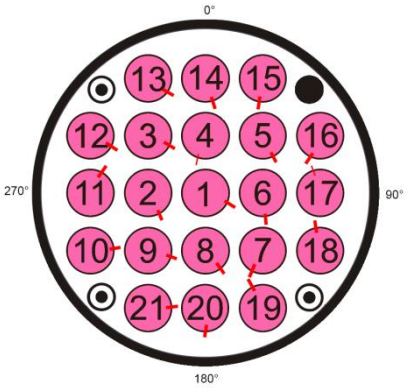
QL-4, at corner rod C

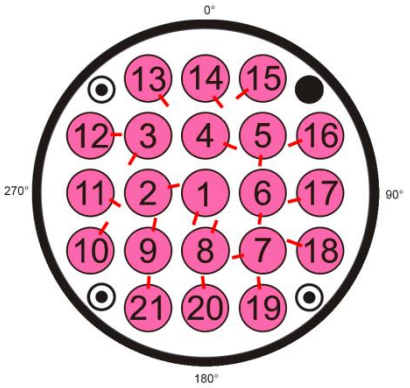


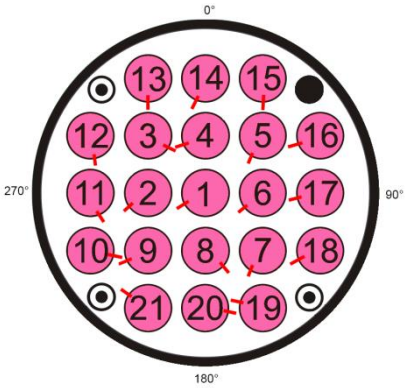
QL-5, at corner rod C

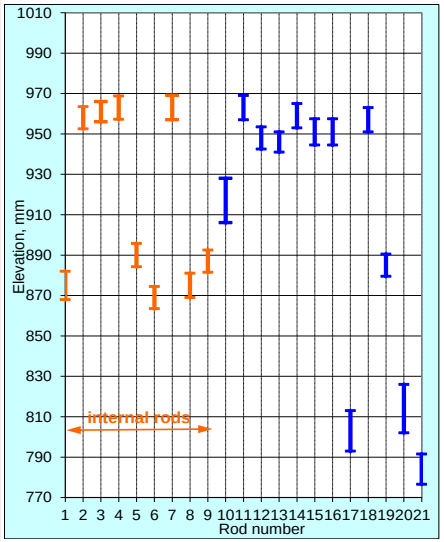
Radial and axial positions of burst openings



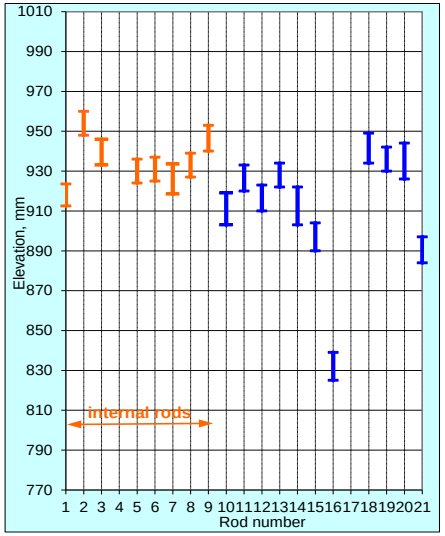




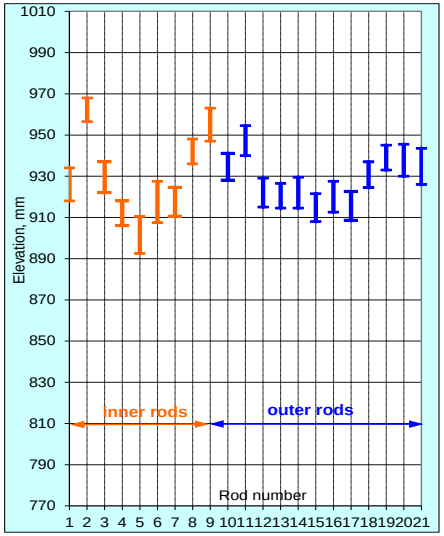




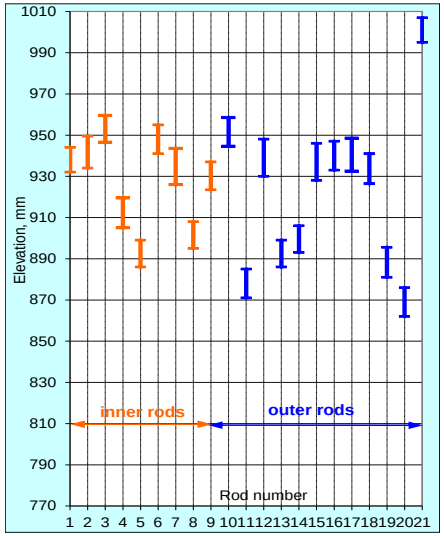
QL-2 (M5®)



QL-4 (hydrogenated M5®)

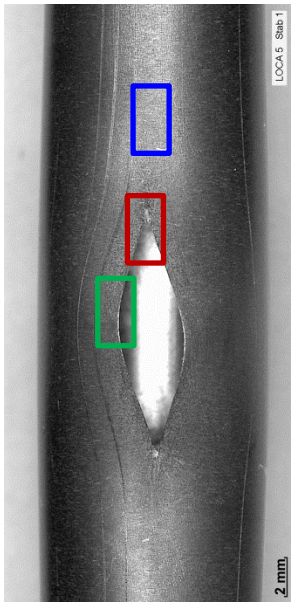


QL-3 (opt. ZIRLO)

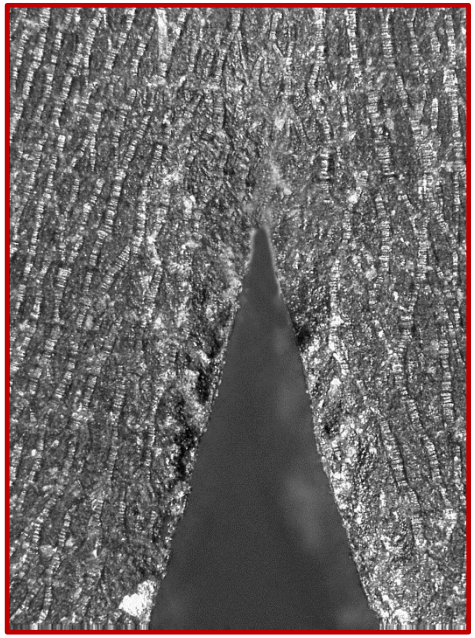


QL-5 (hydrogenated opt. ZIRLO)

Surface cracks around burst opening formed during ballooning at outer surface of cladding



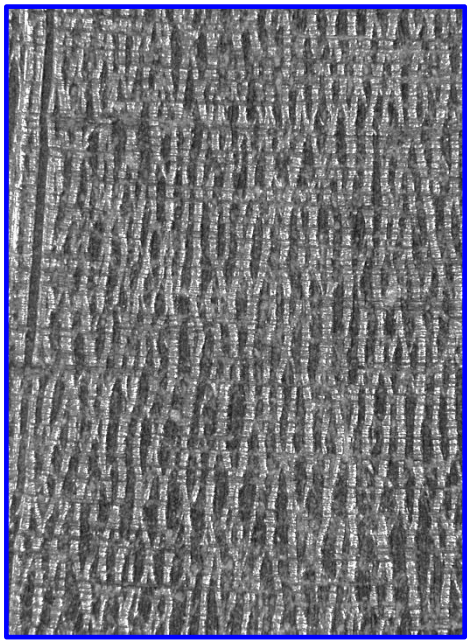
QL-5, rod #1



burst opening top,
≈ 20 cracks/mm



burst opening side,
≈ 20 cracks/mm

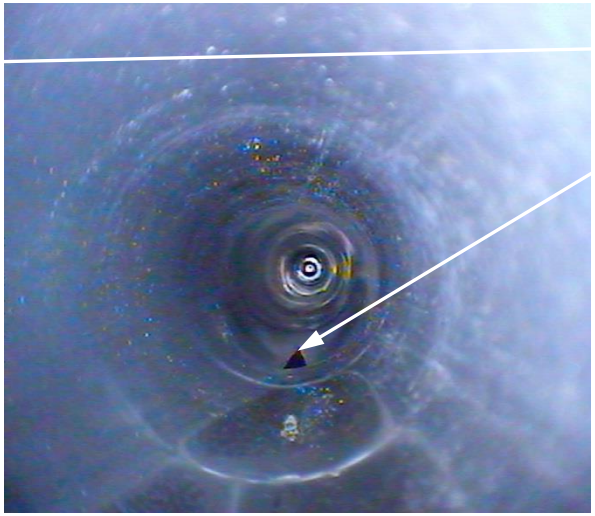


4 mm above opening tip,
≈ 40 cracks/mm

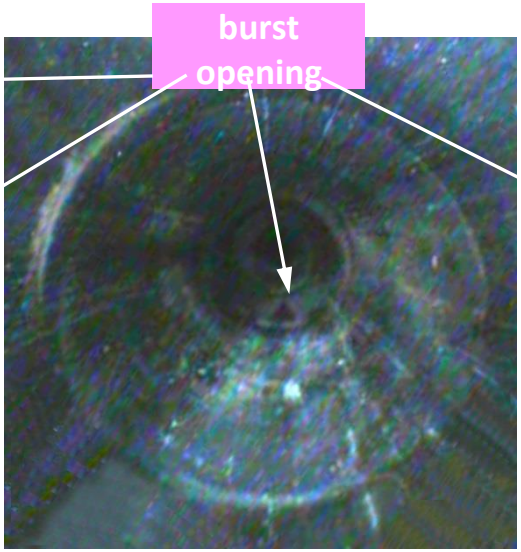
Videoscope observation of contact between pellet and cladding at inner cladding surface below burst opening



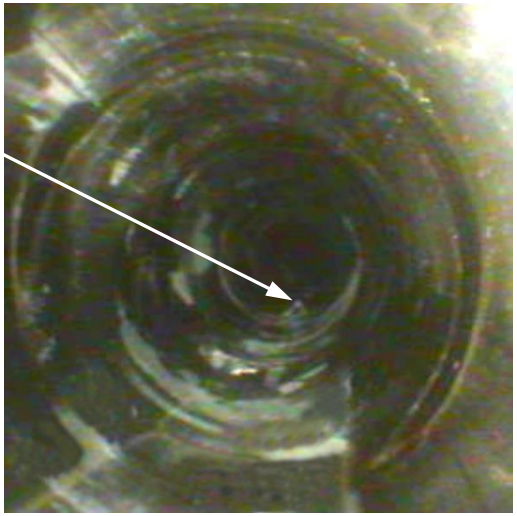
QL2, clad #8



QL4, endoscopy of rod #1



QL3, endoscopy of rod #5



QL5, endoscopy of rod #3

Burst time and burst temperature

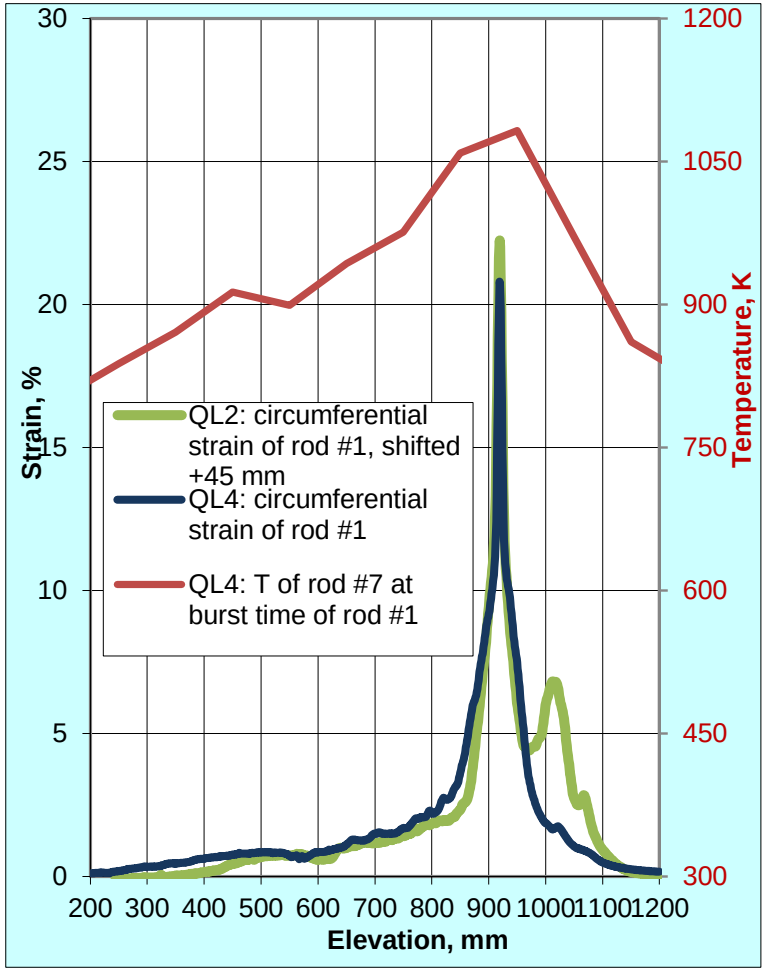
Test	average burst time, inner rods, s	average burst time, outer rods, s	average burst temperature, inner rods, K	average burst temperature, outer rods, K
QL-0 (Zry-4, 2.6 K/s)	124 ± 10	159 ± 9	1176 ± 26	1170 ± 31
QL-1 (Zry-4, 7 K/s)	58 ± 2	75 ± 7	1128 ± 31	1131 ± 30
QL-2 (M5®, 8 K/s)	52 ± 2	65 ± 3	*1148 ± 22*	1133 ± 27
QL-3 (ZIRLO, 8 K/s)	52 ± 2	63 ± 4	*1116 ± 13*	1118 ± 39
QL-3HT (ZIRLO, 7.5 K/s)	54 ± 3	70 ± 7	1127 ± 42	1127 ± 54
QL-4 (hydr. M5®, 8 K/s)	51 ± 3	62 ± 4	***1108 ± 13***	1106 ± 32
QL-5 (hydr. ZIRLO, 8 K/s)	54 ± 2	68 ± 4	***1065 ± 19***	1093 ± 42

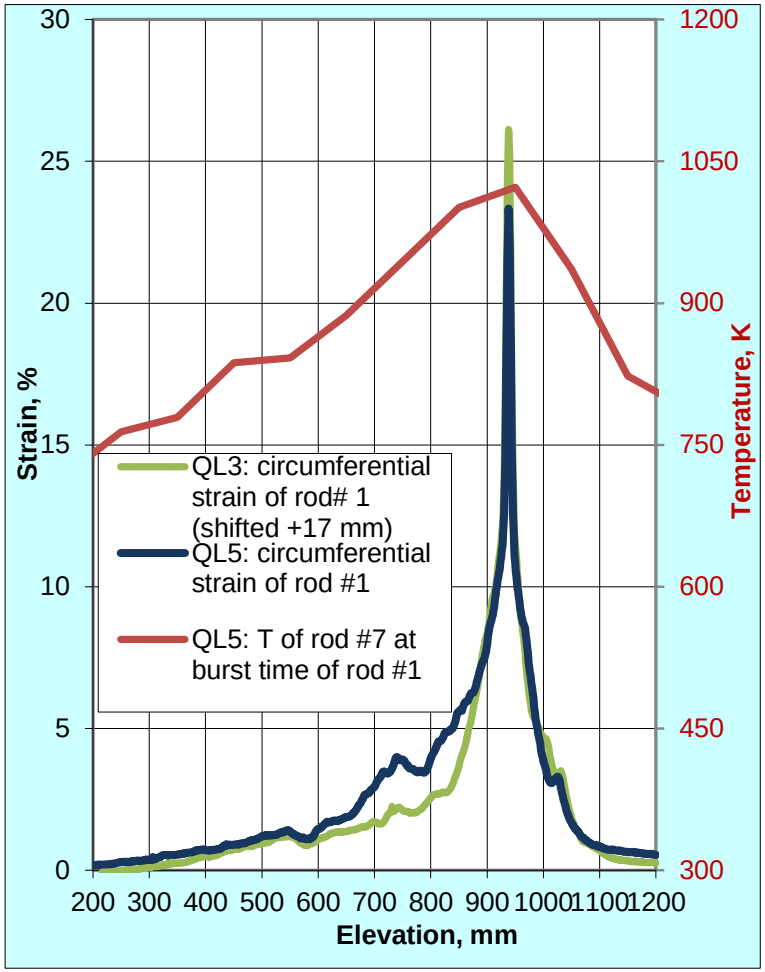
Burst temperature of hydrogenated cladding is about 40 … 50 K lower in comparison to fresh cladding

Average geometrical parameters of burst openings

Test	max burst width, mm	burst length, mm	burst area, mm²
QL0 (Zry-4, 2.6 K/s), bended rods	n. a.	12.7 ± 3.3	33.5 ± 21.1
QL1 (Zry-4, 7 K/s), strong bended rods	3.9 ± 2.6	14.5 ± 6.7	44.7 ± 49.2
QL2 (M5®, 8 K/s), bended rods	3.1 ± 1.2	13.5 ± 4.0	29.0 ± 22.9
QL3 (ZIRLO, 8 K/s)	3.9 ± 0.9	14.4 ± 2.2	31.0 ± 11.8
QL3HT (ZIRLO, 7.5 K/s), strong bended rods	n. a.	19.3 ± 7.3	n. a.
QL4 (hydr. M5®, 8 K/s)	3.3 ± 0.7	13.1 ± 2.0	24.1 ± 7.4
QL5 (hydr. ZIRLO, 8 K/s)	3.8 ± 0.6	14.3 ± 1.8	29.9 ± 8.8

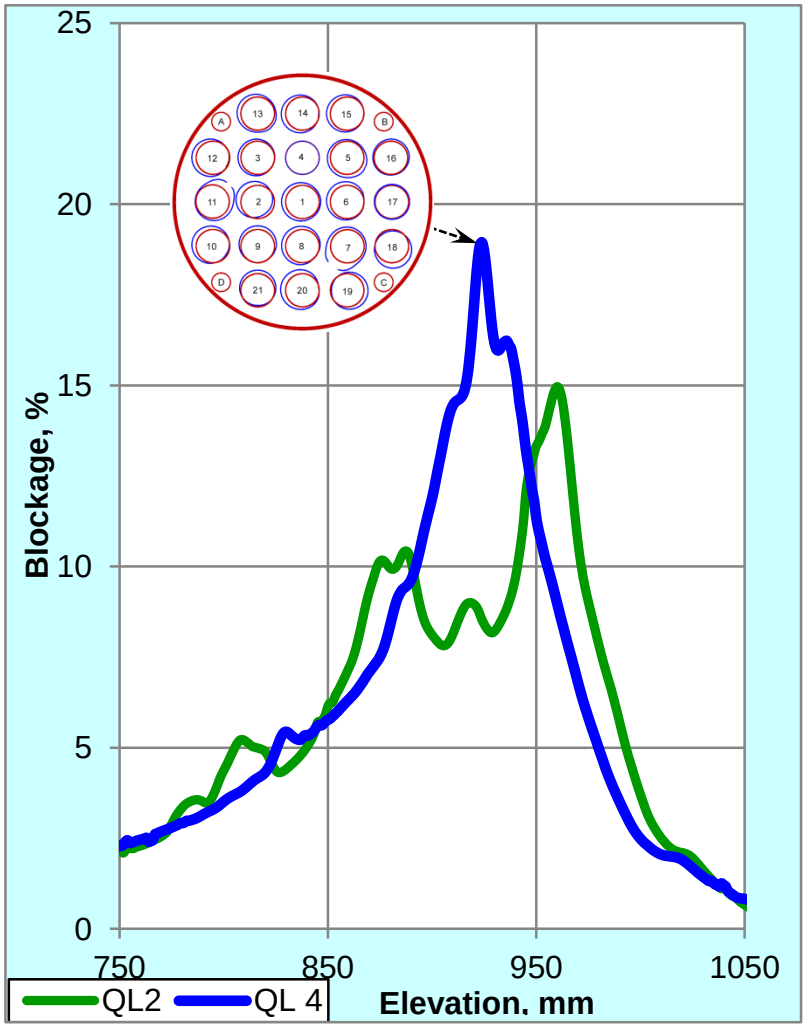
Results of laser profilometry: typical axial distributions of the circumferential strain of claddings

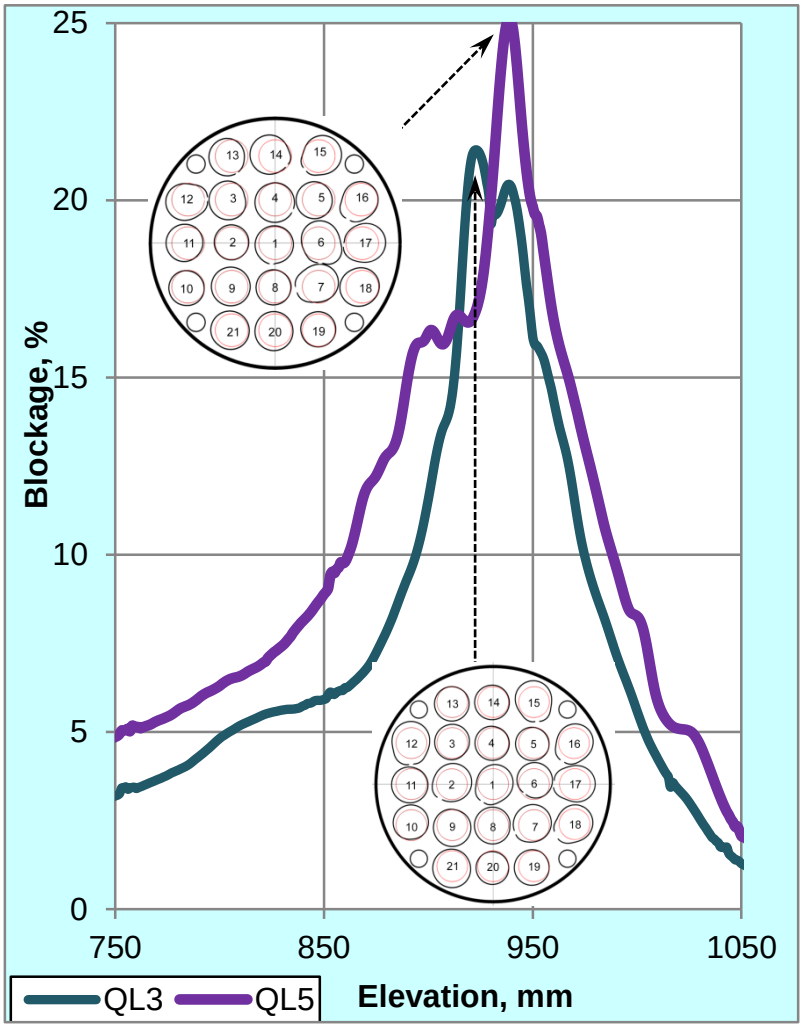




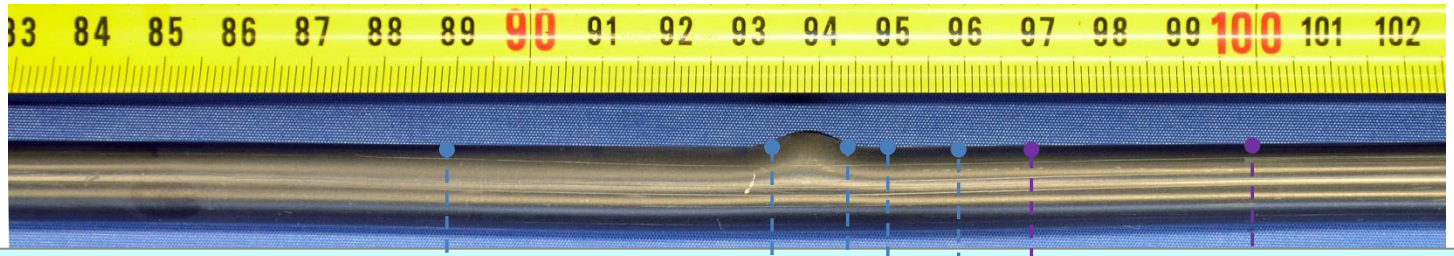
- additional ballooning peaks (alongside the main burst peak) formed during heat-up
- circumferential strain values in ballooning regions outside of burst opening are noticeably higher for the pre-hydrogenated claddings: cladding starts to creep earlier due to α→β transformation around dissolving hydrides

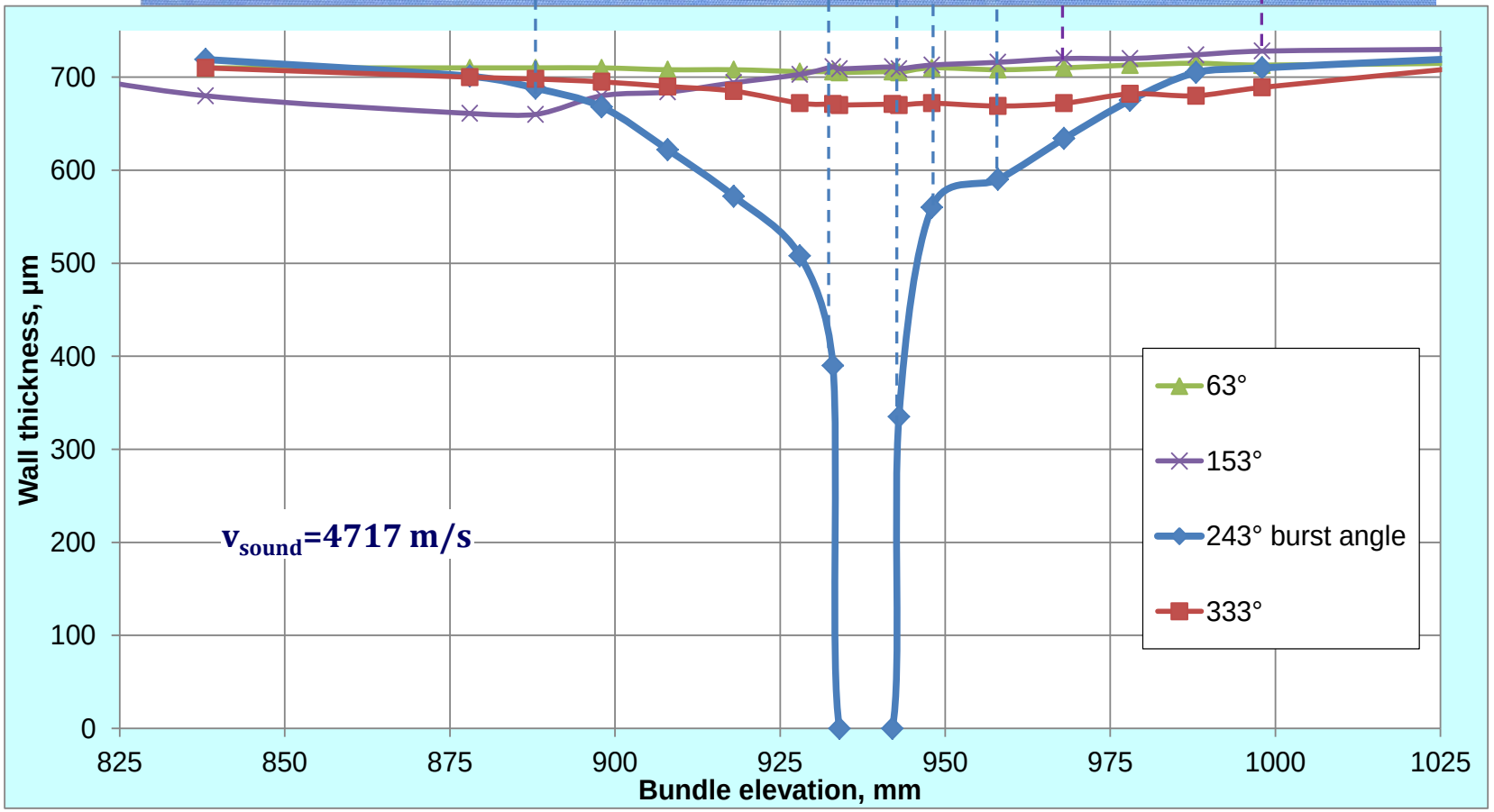
Results of laser profilometry: axial distribution of coolant channel blockages



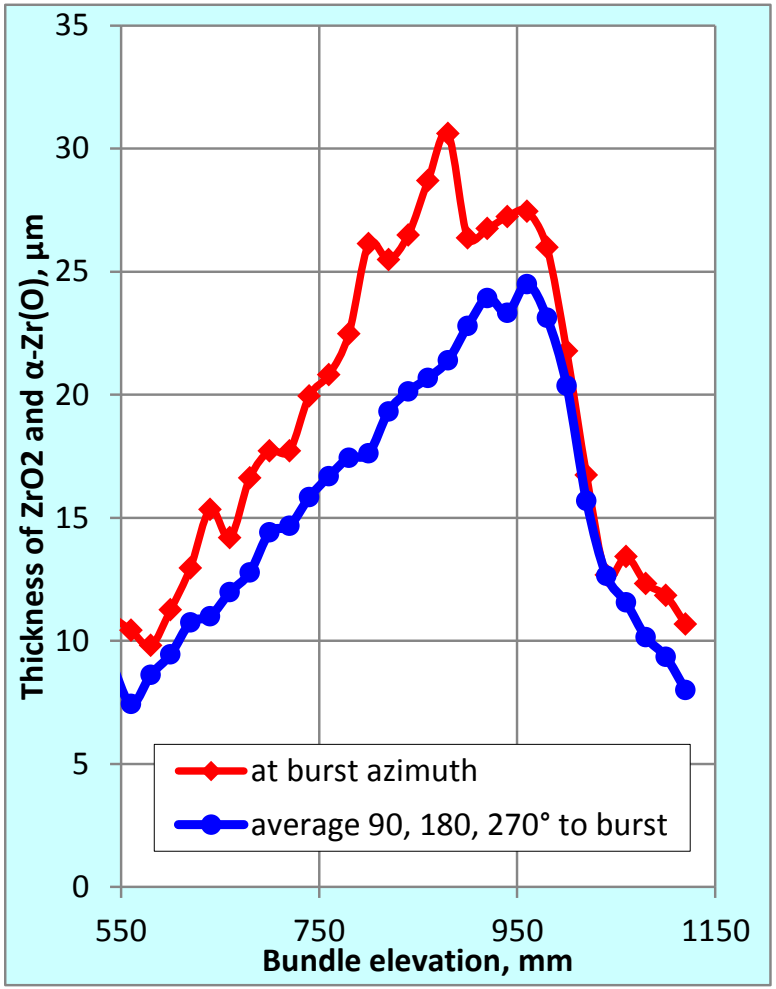


Ultrasound measurements: typical cladding wall thinning below and above the burst opening (rod #1, QUENCH-L5)

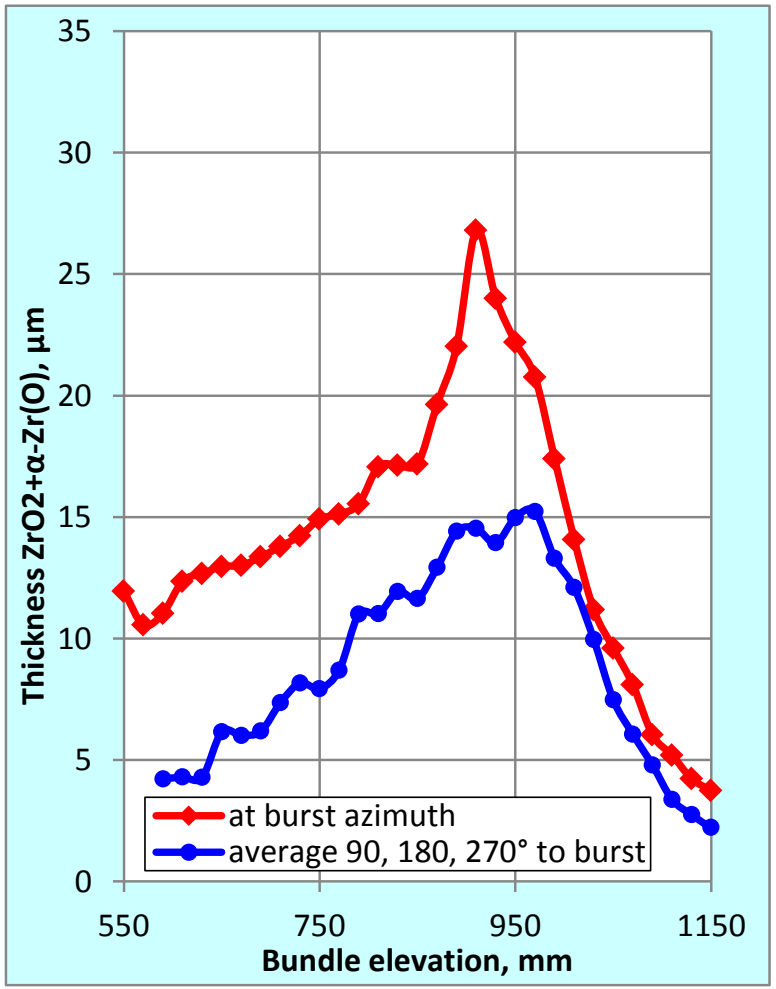




Eddy current measurements: axial distribution of oxidation rate (ZrO_2+α-Zr(O) outer layers)

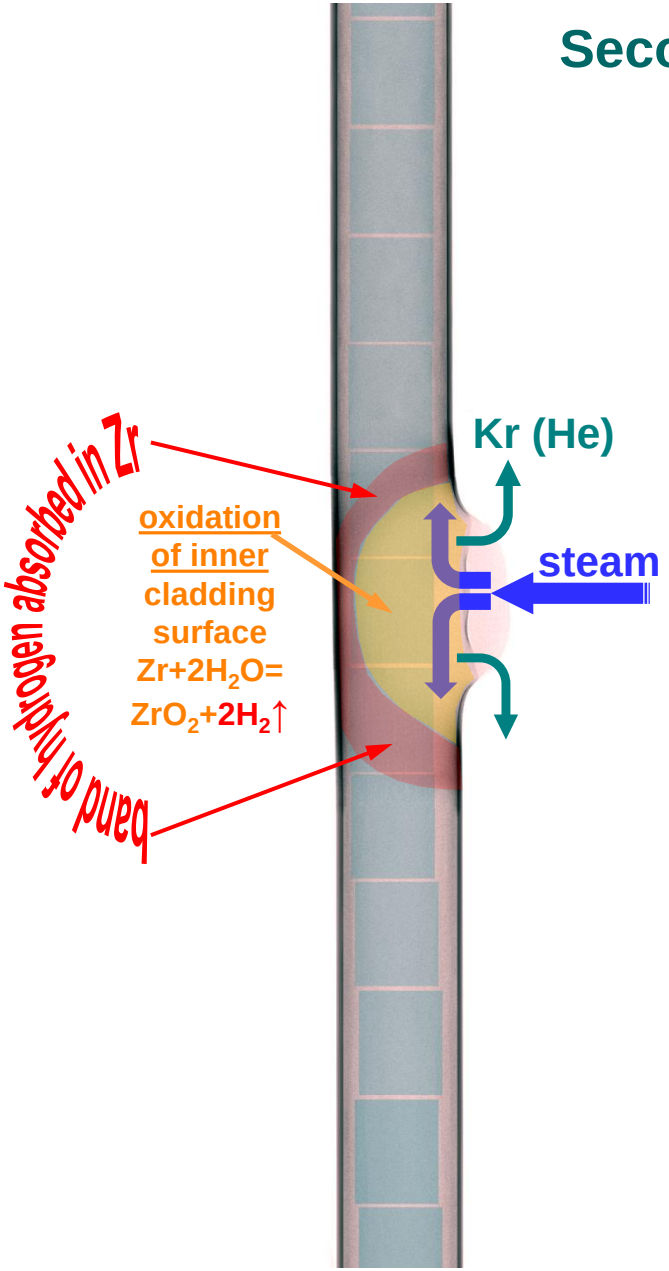


QL-4, central rod



QL-5, central rod

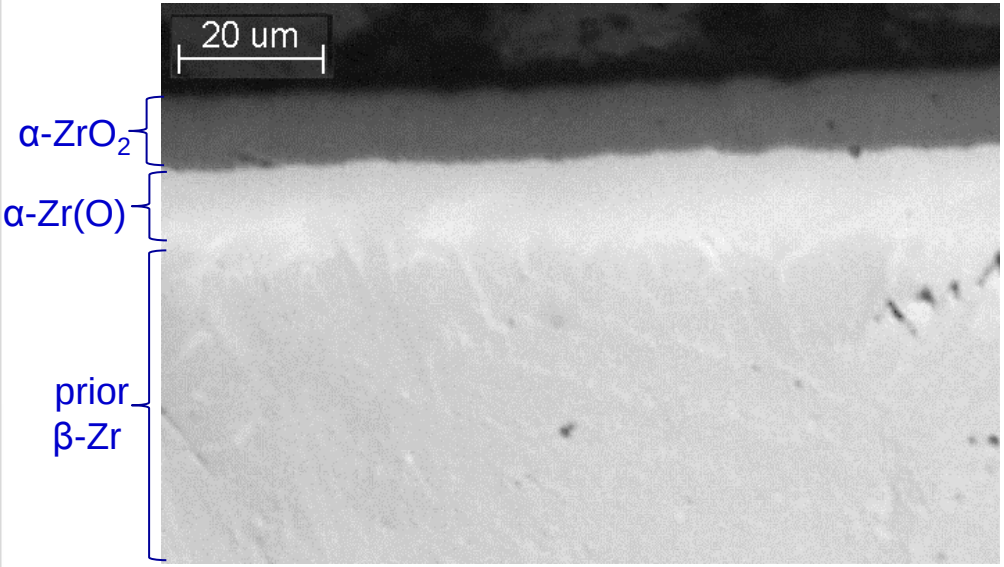
Secondary hydrogenation of cladding



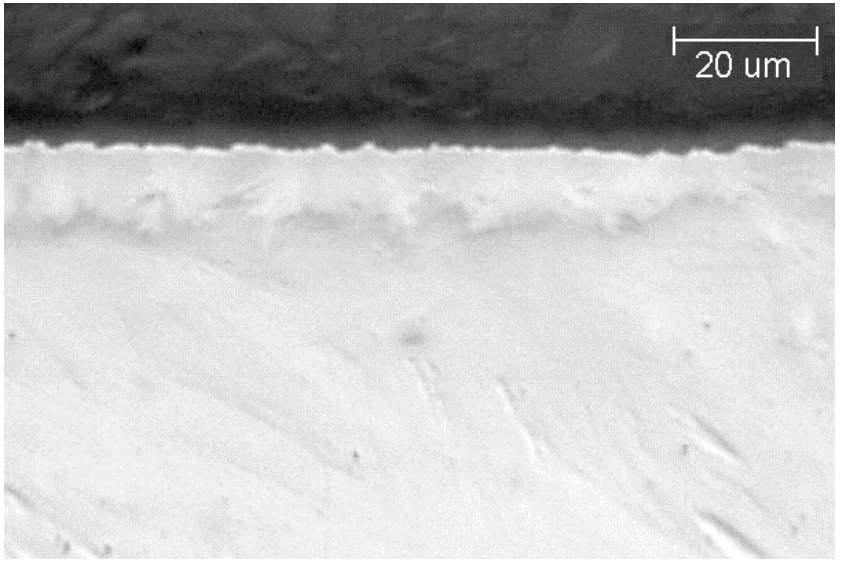
Sequence of phenomena during LOCA:

- cladding ballooning and burst, relief of inner rod pressure
- steam penetration through the burst opening, steam propagation inside the gap between cladding and pellet
- oxidation of inner cladding surface with hydrogen release
- absorption of hydrogen by cladding at the boundary of inner oxidised area at temperatures above the phase transition $\alpha \rightarrow (\alpha+\beta)$ in Zr alloy
- local embrittlement of cladding near to burst opening

Structures of inner cladding surface: thinning of inner oxide layer



QL-4, clad #1: inner oxidation of cladding at the burst elevation

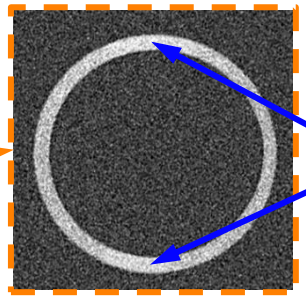


QL-4, clad #1: decreased inner oxidation of cladding at the elevation of hydrogen band

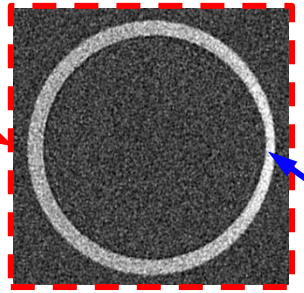
Neutron tomography: reconstruction of hydrogen distribution and determination of hydrogen concentration



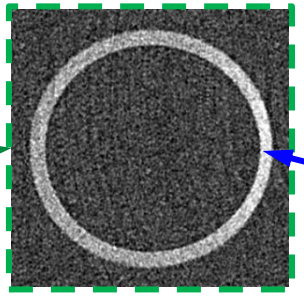
QL-3, clad #6, n^0 3D image



light spots max C_H



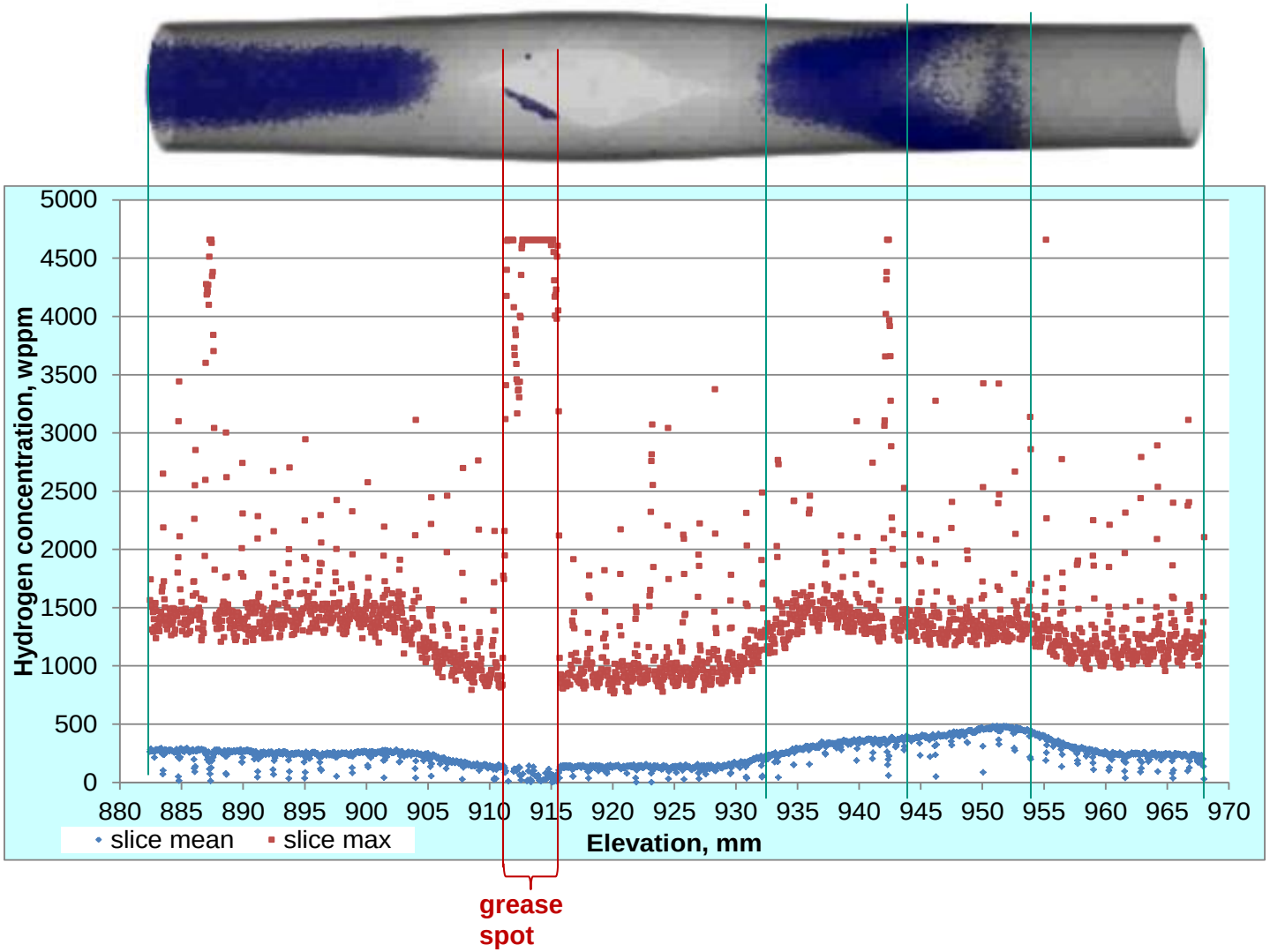
mean C_H = 475 ± 5 wppm,
local max C_H = 1430 ± 70 wppm



mean C_H = 278 ± 15 wppm,
local max C_H = 1450 ± 100 wppm

n^0 3D cross section,
axial thickness 40 µm
(voxel size)

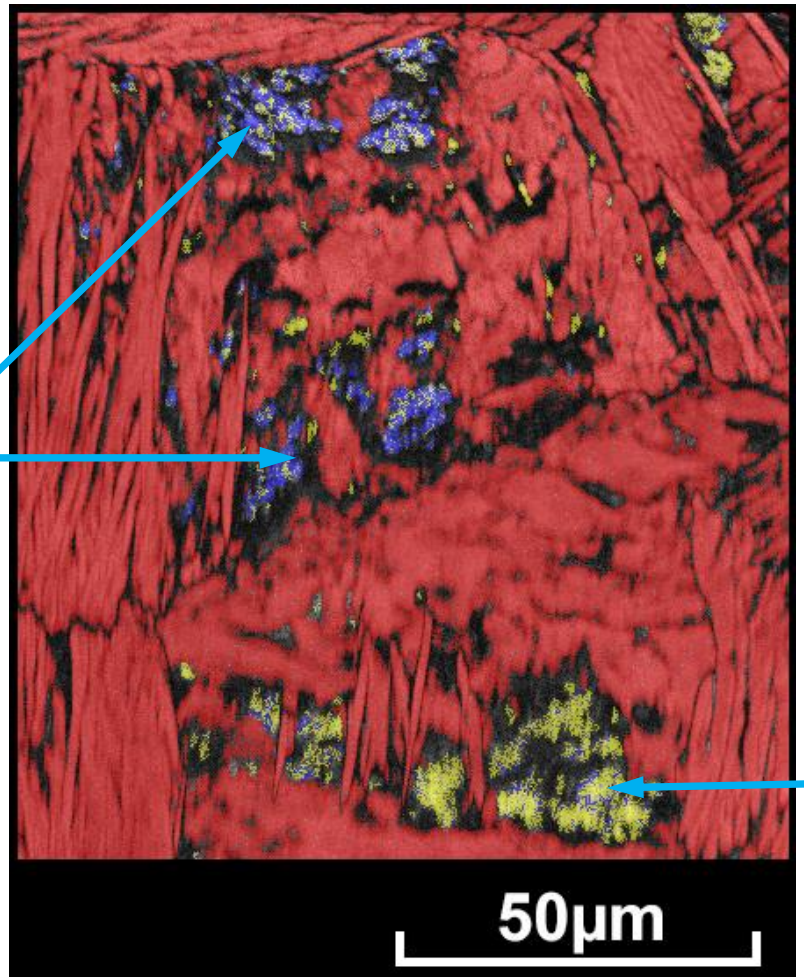
Neutron tomography: axial distribution of hydrogen (mean and max values for each elevation)



Axial maxima of the mean value of the hydrogen concentration (wppm) (results of n^0 tomography)

rod #	QL-0	QL-1	QL-2	QL-3	QL-3HT	QL-4 (H 100)	QL-5 (H 300)
1	1393		120			640	390
2		715	235	240			
3	845		240	180	2000	165	345
4				505	1940		
5				366			238
6			220	475		273	
7			170	363			327
8			440	307			
9			220	190		371	285
10				165		236	
11							
12			90				
13			150		940		
14			215			230	
16							
17							
18			150				
19							
20			160				
21			90				293

EBSD analysis of cladding region inside hydrogen band
(QL-4, central rod)

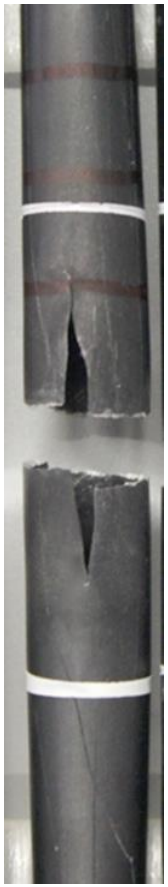


hydride clusters near to boundaries of the region of coalesced grains with the same spatial orientation

Tensile test results: typical cladding fracture types



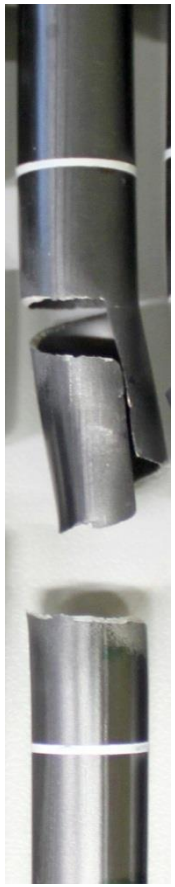
QL-4, clad #1
hydrogen
embrittlement



QL-3, clad #6



QL-3, clad #21



QL-4, clad #12

stress concentration at the edges of burst opening



QL-3, clad #12
necking
far away from
burst region

Tensile test results: typical parameters

Para-meter	QL-2		QL-4 100 wppm H		QL3		QL5 300 wppm H	
	stress conc.	necking	stress conc.	necking	stress conc.	necking	stress conc.	necking
ultimate tensile strength, MPa	496	507	512	520	514	521	542	554
fracture stress, MPa	453	384	511	402	501	385	541	448
elon-gation at fracture, %	7.8	11.2	6.1	9.7	9.2	11.0	5.5	9.0

Summary and conclusions

- Seven QUENCH-LOCA bundle tests with different cladding materials (Zry-4, M5®, Opt. ZIRLO™) were performed according to a temperature/time-scenario typical for a LBLOCA in a German PWR. For two tests (QUENCH-L4 and -L5) pre-hydrogenated claddings (with 100 and 300 wppm H for M5® and Opt. ZIRLO™, correspondingly) were used.
- Generally, a peak cladding temperature of about 1350 K (1250 K for QUENCH-L5) was reached at the end of the heat-up phase at the bundle elevation of 950 mm. The maximal heat-up rate was 8 K/s (2.6 K/s for commissioning test QUENCH-L0); the cooling phase lasted about 120 s (0 s for QUENCH-L0) and was terminated by 3.3 g/s/effective rod water flooding. The tangential temperature gradient across a rod was between 30 and 70 K on the burst onset (important parameter for ballooning size, accordingly to the REBEKA criterion).
- The detailed profilometry of the whole length of the rods showed formation of not only main ballooning area (with burst) but also, for several rods of each bundle, additional two or three ballooning regions. The reason should be the relatively long duration of the ballooning process at the main position and reaching of ballooning conditions (T>1000 K) at other bundle elevations.
- The maximal coolant channel blockage was less than 35% even for conservatively assuming coplanar ballooning. Due to this only moderate blockage, good bundle coolability was kept for all bundles.

Summary and conclusions (cont.)

- Cladding wall thinning from 725 to 350 µm due to ballooning was observed at the burst side along 50 mm below and above burst opening. The maximal oxide thickness at the outer cladding surfaces was less than 15 µm. Surface cracks, penetrating both layers, were formed in vicinity of burst opening during ballooning.
- Oxide layer formed after the burst at the inner cladding surface around the burst opening with a thickness of about 15 µm decreasing to 3 µm at a distance of about 20 mm from the burst opening.
- Hydrogen enrichments of a quite complex 3D form were observed for inner rods having seen peak cladding temperatures of more than 1200 K.
- Neutron tomography analysis showed the maximal hydrogen concentrations inside the hydrogen bands less of 750 wppm (averaged through the cladding cross section).
- EBSD analysis showed that the hydrides with µm-sizes are distributed in the matrix intra as well inter granular
- During quenching, following the high-temperature test stages, no fragmentation of claddings was observed (residual strengths and ductility were sufficient).
- Tensile tests at room temperature evidenced fracture at hydrogen bands for several inner rods with local hydrogen concentrations about 1500 wppm and more. Claddings with lower hydrogen concentrations fractured due to stress concentration at burst opening edges. Other tensile tested claddings failed after necking far away from burst region.

Acknowledgment

The QUENCH-LOCA experiments are supported and partly sponsored by the association of the German utilities (VGB). The M5® claddings and Zircaloy-4 spacer material were provided by AREVA. The opt. ZIRLO™ claddings and spacer material were provided by WESTINGHOUSE.

The authors would like to thank Mr. C. Rössger, Mr. J. Moch, Mrs. J. Laier and Mrs. U. Peters for intensive work during test preparation and post-test investigations, Dr. A. Pshenichnikov for the EBSD analysis, Dr. H. Leiste for the X-ray diffractometry measurements.

Thank you for your attention

https://www.iam.kit.edu/wpt/loca/
http://www.iam.kit.edu/awp/666.php
http://quench.forschung.kit.edu/

Pre-Test Benchmarks in the Frame of the NUGENIA QUESA Project: QUENCH-ALISA and CODEX-AIT3

T. Hollands
Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH, Germany

During a severe accident in an NPP, air ingress into the vessel will lead to a mixed atmosphere inside the core region that is mainly composed of steam and air. As air is a highly oxidizing this can lead to an enhanced core oxidation and degradation affecting the release of Fission Products (FP), especially increasing that of ruthenium. This FP is of particular importance because of its high radio-toxicity and its ability to form highly volatile oxides. Oxygen affinity is decreasing from Zircalloy cladding, to fuel and to ruthenium inclusions in the fuel. Therefore, understanding the phenomena governing cladding oxidation by air is a prerequisite for realistic source term predictions. Based on the SAFEST (CODEX) and ALISA (QUENCH) calls for proposal, the idea is to define a counterpart of QUENCH-16 but with a mixture of air and steam instead of pure air in order to be more representative of a reactor case.

In the frame of the NUGENIA QUESA project, pre-test benchmarks were organised by GRS (and previously also EdF) for both air ingress tests to predict the assumed test scenario and to identify suitable boundary conditions for achieve the test objectives. Both tests were conducted following basically the same procedures, but with different geometry and defined temperature limits: pre-oxidation, intermediate cooldown, air/steam ingress phase, cooldown by water (QUENCH) or argon (CODEX). For the QUENCH-ALISA test at KIT (Germany) five participants performed the simulations with four codes (ATHLET-CD of AC^2, MAAP, RELAP/SCDAPSIM, SOCRAT). Most codes predict a similar behaviour until reflood with the boundary conditions defined based on parameter studies. One notable difference to QUENCH-16 was that during pre-oxidation the power in the core was reduced in the codes in order to avoid premature strong oxidation and temperature escalation, keeping the temperatures below the control rod melting temperature. A parameter study of the flow rates during the air ingress phase was performed to identify the flow rates which allow at least partial starvation of oxygen, steam and/or nitrogen, a main objective of the experiment. The ATHLET-CD results show that the way oxidation of the molten control rod is considered can influence the overall thermal behaviour of the bundle. For CODEX-AIT3 at MTA EK (Hungary) three participants performed the calculations with three codes (ASTEC, ATHLET-CD of AC^2, SOCRAT). An intensive parameter study was performed with ATHLET-CD to identify suitable boundary conditions to achieving test objectives. These boundary conditions were used for the benchmark. Even with this additional information, in the SOCRAT simulation with benchmark boundary conditions the power had to be modified (factor 1.5). In the ASTEC simulation the air ingress phase before the cooldown criterion was reached was much longer compared to the other codes. Apart from this, the ASTEC results show good agreement on temperature behaviour for elevations above 250 mm. Overall, the three simulations show reasonable agreement, but there are larger differences than found for the QUENCH-ALISA test although very similar model options were used.


Thorsten Hollands
23rd Int

Pre-Test Benchmarks in the Frame of the NUGENIA QUESA Project: QUENCH-ALISA and CODEX-AIT3

Thorsten Hollands

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH, Germany

23rd International QUENCH Workshop

Karlsruhe Institute of Technology

17. – 19. October 2017

Content

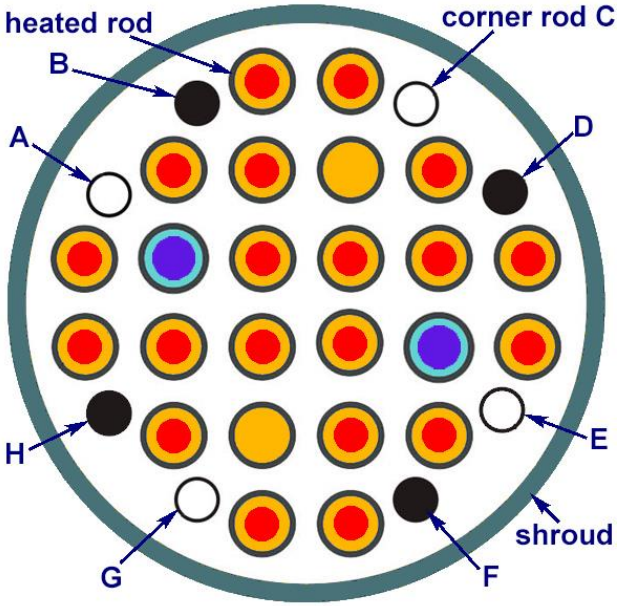
- QUENCH-ALISA pre-test benchmark
  - QUENCH facility
  - Test scopes and transient
  - Participants and codes
  - Experimental parameters
  - Results of pre-test simulations
  - Main findings

- CODEX-AIT3 pre-test benchmark
  - CODEX facility
  - Test scopes and transient
  - Participants and codes
  - Experimental parameters
  - Results of pre-test simulations
  - Main findings

QUENCH-ALISA pre-test benchmark

QUENCH-ALISA test

Component	No.	Material properties	Outer diameter (mm)	Inner diameter (mm)
Rod Cladding	22	ZIRLO (for the benchmark)	9.50	8.36
SIC Absorber Rod	2	Ag 80%, In 15%, Cd 5% density: 10.17 g/cm³	7.65	
Absorber Cladding	2	Steel AISI 316	9.68	7.72
Absorber Guide Tube	2	Zr alloy	12.45	11.25
Corner Rod	8	Zry-4	6.00	
Shroud	1	Zry	89.00	83.00



- 5 ZIRLO grid spacers with pitch 12.6 mm (for the benchmark)

QUENCH-ALISA test – Scopes and transient

- Scope of the test:
  - First integral test on oxidation with air/steam mixture.
  - Starvation of oxygen and/or steam (global or local) is very important, because data on nitride formation are expected.
- Test transient:
  - Pre-oxidation
  - Intermediate cooldown
  - Air/steam ingress
  - Quenching
- The electrical power for QUENCH-ALISA is the one for QUENCH-16 minus 2 kW during pre-oxidation.

Participants and codes

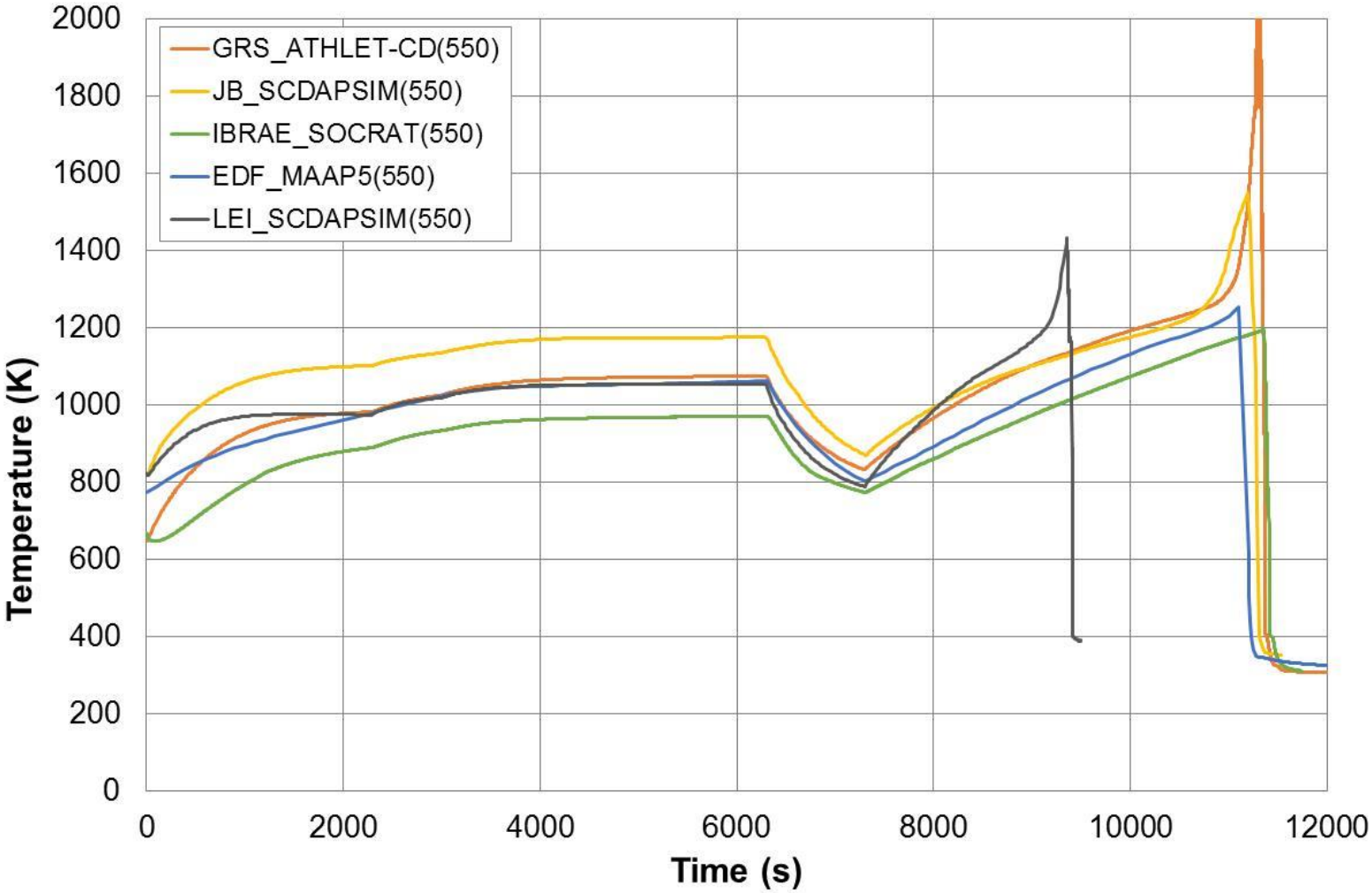
- Participants:
  - GRS ATHLET-CD
  - EDF MAAP5
  - Jon Birchley SCDAP-SIM
  - LEI RELAP/SCDAPSIM
  - IBRAE SOCRAT

Participants and codes
- Air oxidation model options -

Code Version	ATHLET-CD 31a	MAAP5.02a2	SCDAPSIM35 (JB)	RELAP/SCDAPSIM (LEI)	SOCRAT v2
Air Oxidation (or Oxygen) Correlation	Steinbrück	Powers	Utsuega-Hofmann/Urnaic Heidrick	Utsuega-Hofmann/Urnaic Heidrick	Oxygen diffusion enhancement factor depending on ZrN molar fraction. Pure oxygen is treated as vapour except chemical heat.
Nitriding Correlation	Hollands	KIT - EDF	No nitriding. Birchley model for accelerated oxidation.	no	Empirical model depending on nitrogen content in gas mixture.
Possibility of nitrides reoxidation	Reoxidation not modelled, indirect consideration with simple porosity model which has an effect on the following steam oxidation.	Indirect consideration with same law as for oxidation.	no	no	no

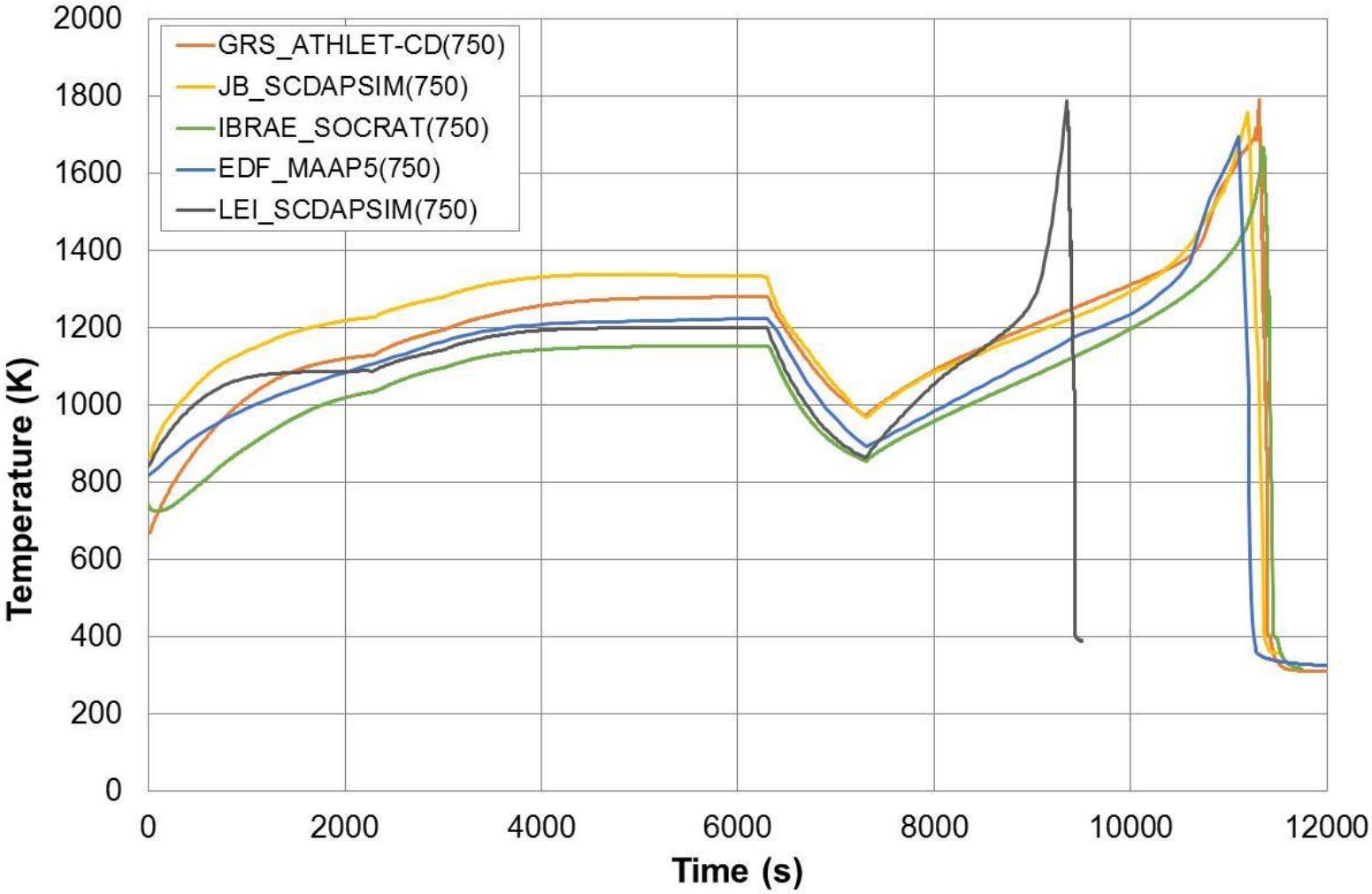
Results of pre-test simulations
- Temperatures (I) -

- Temperatures at 550 mm



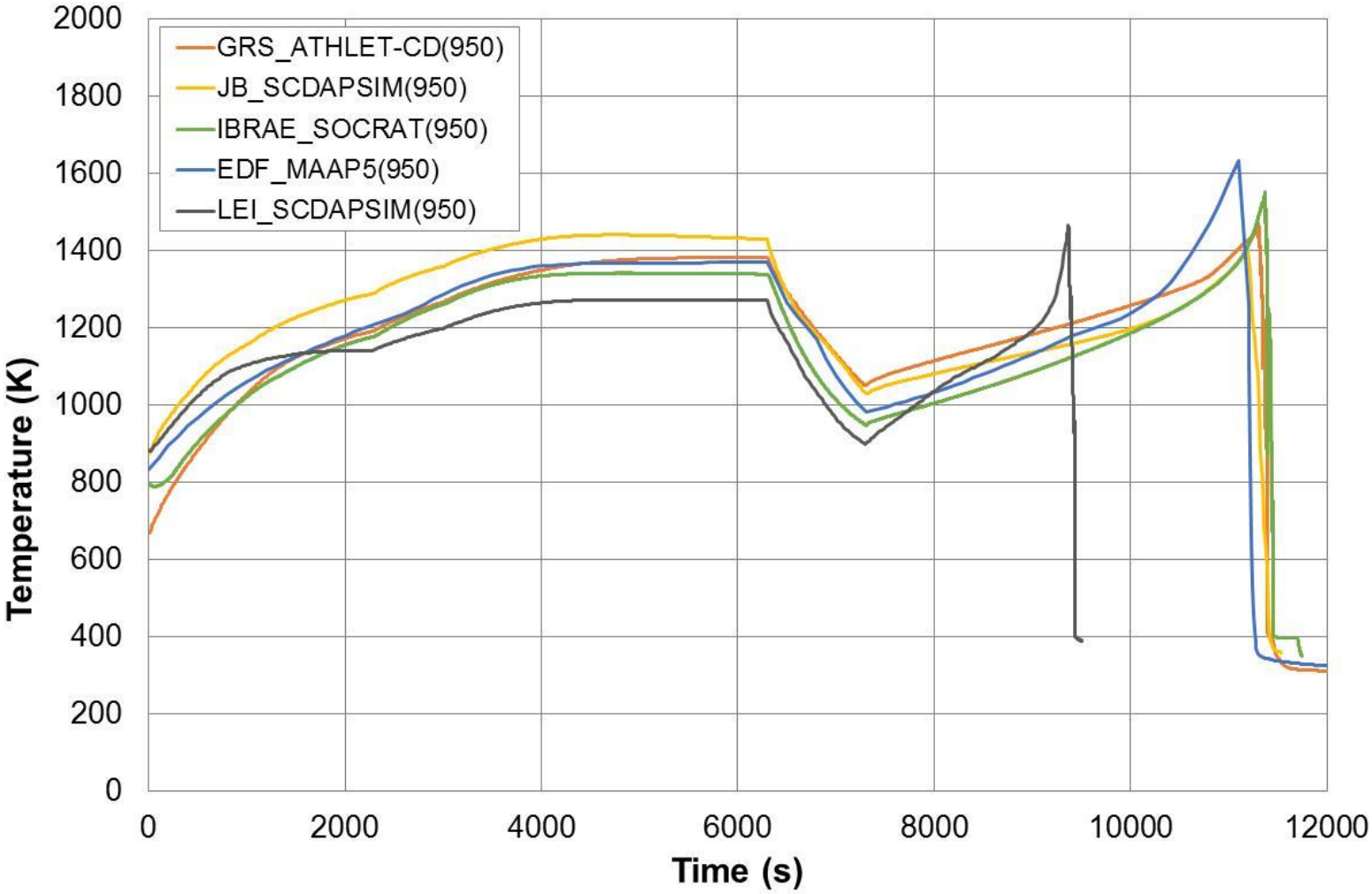
Results of pre-test simulations
- Temperatures (II) -

- Temperatures at 750 mm

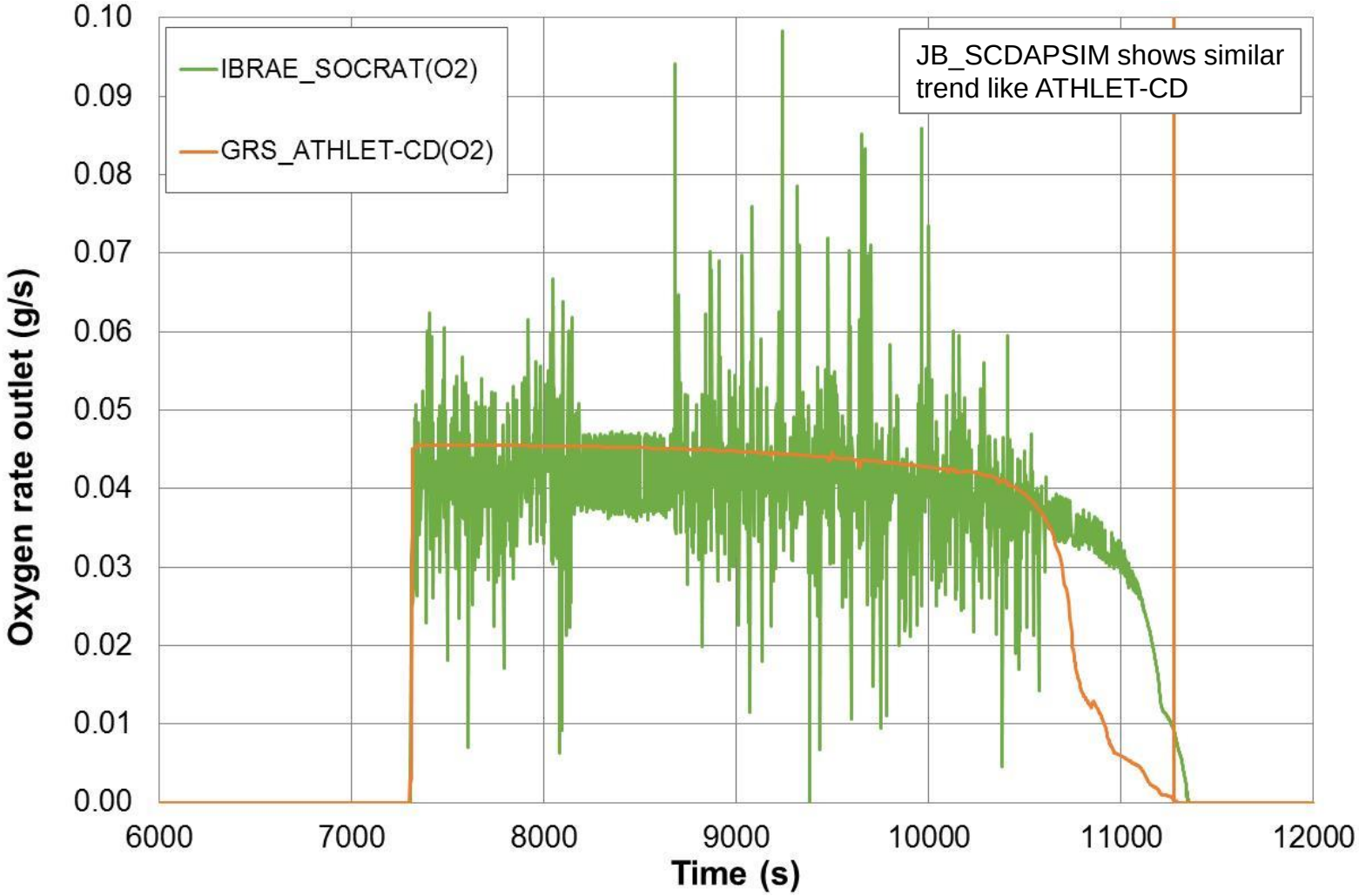


Results of pre-test simulations
- Temperatures (III) -

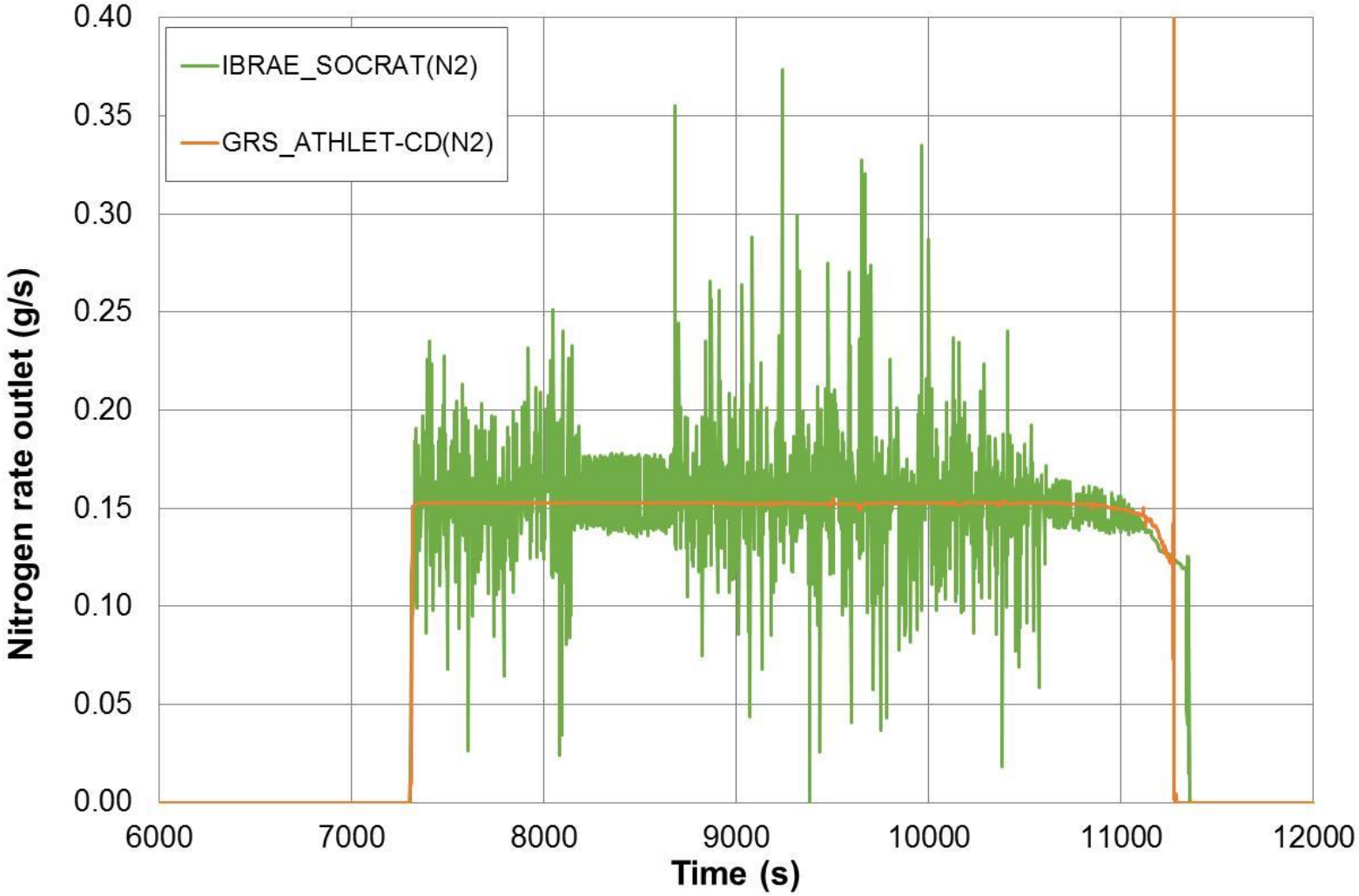
- Temperatures at 950 mm



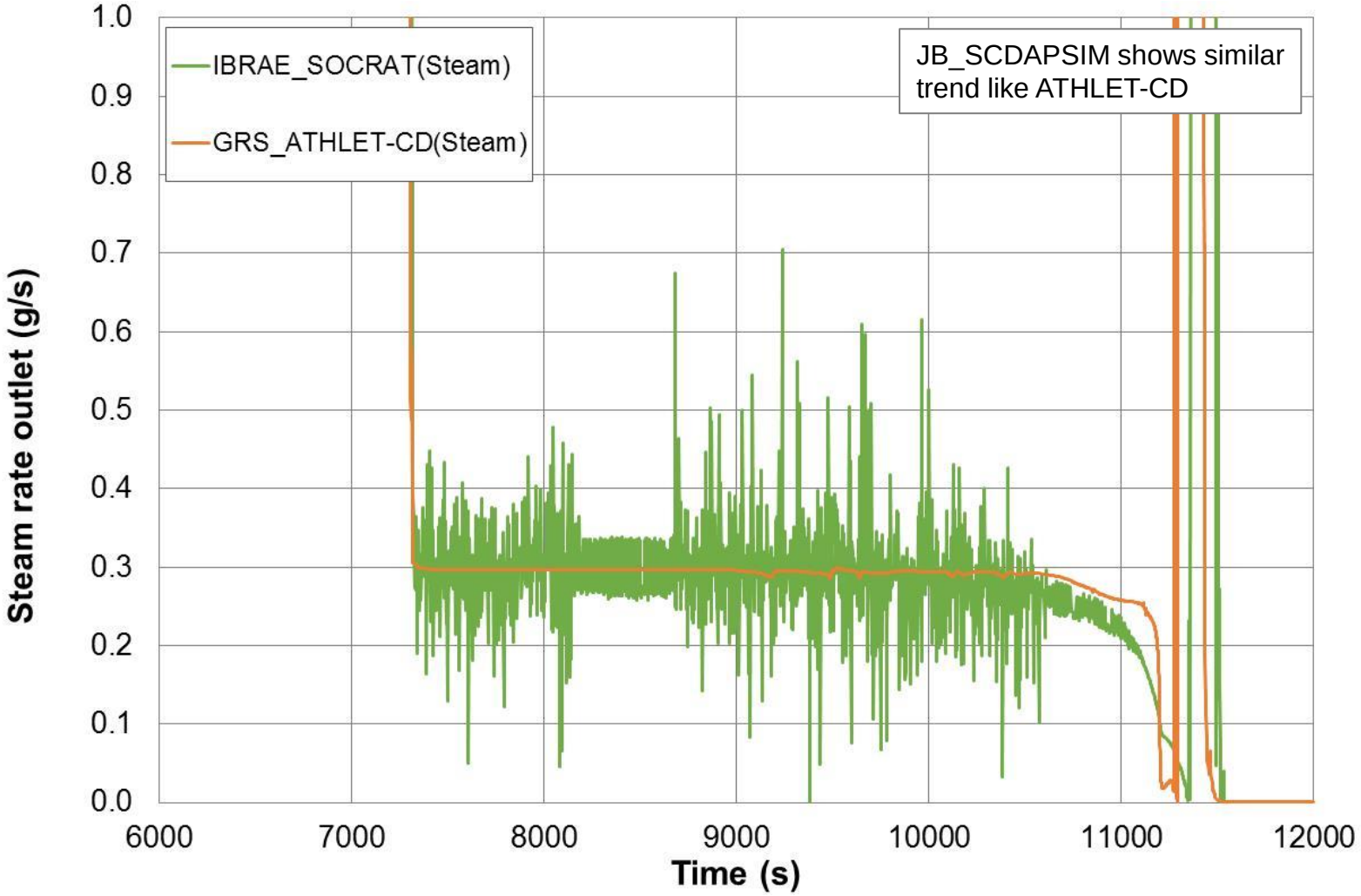
Results of pre-test simulations
- Oxygen flow rate at outlet -



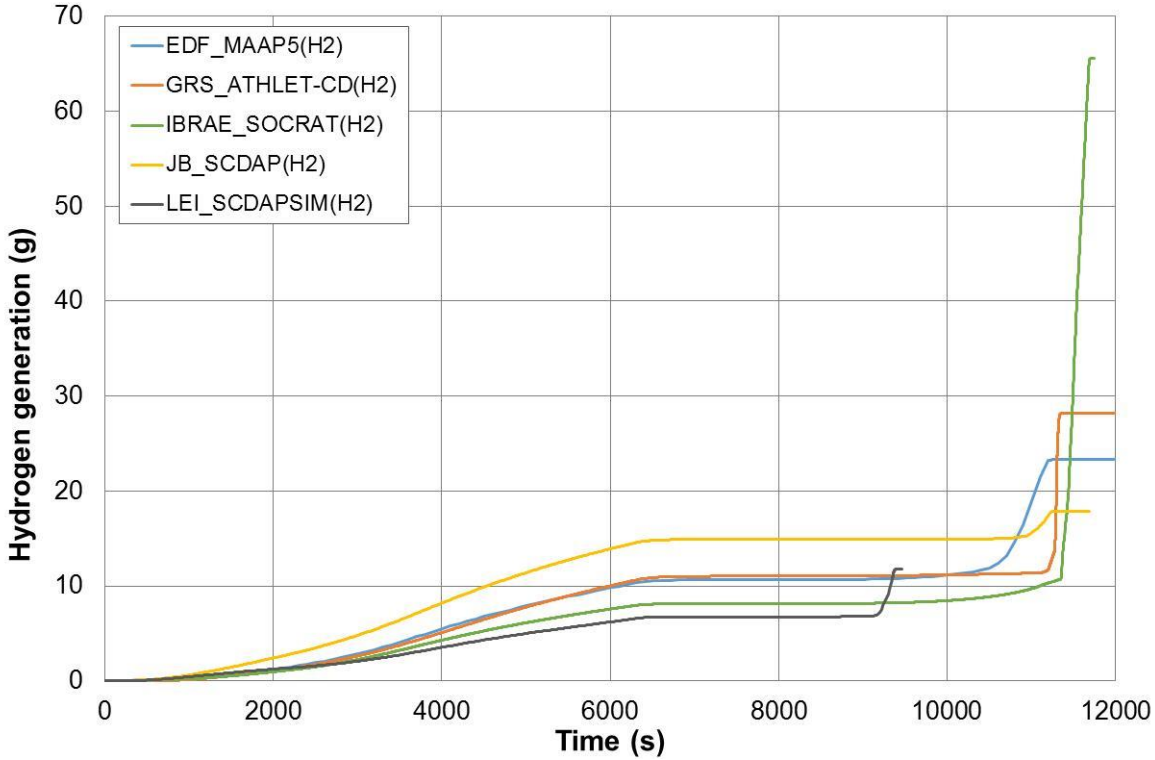
Results of pre-test simulations
- Nitrogen flow rate at outlet -



Results of pre-test simulations
- Steam flow rate at outlet -



Results of pre-test simulations
- Hydrogen Generation -



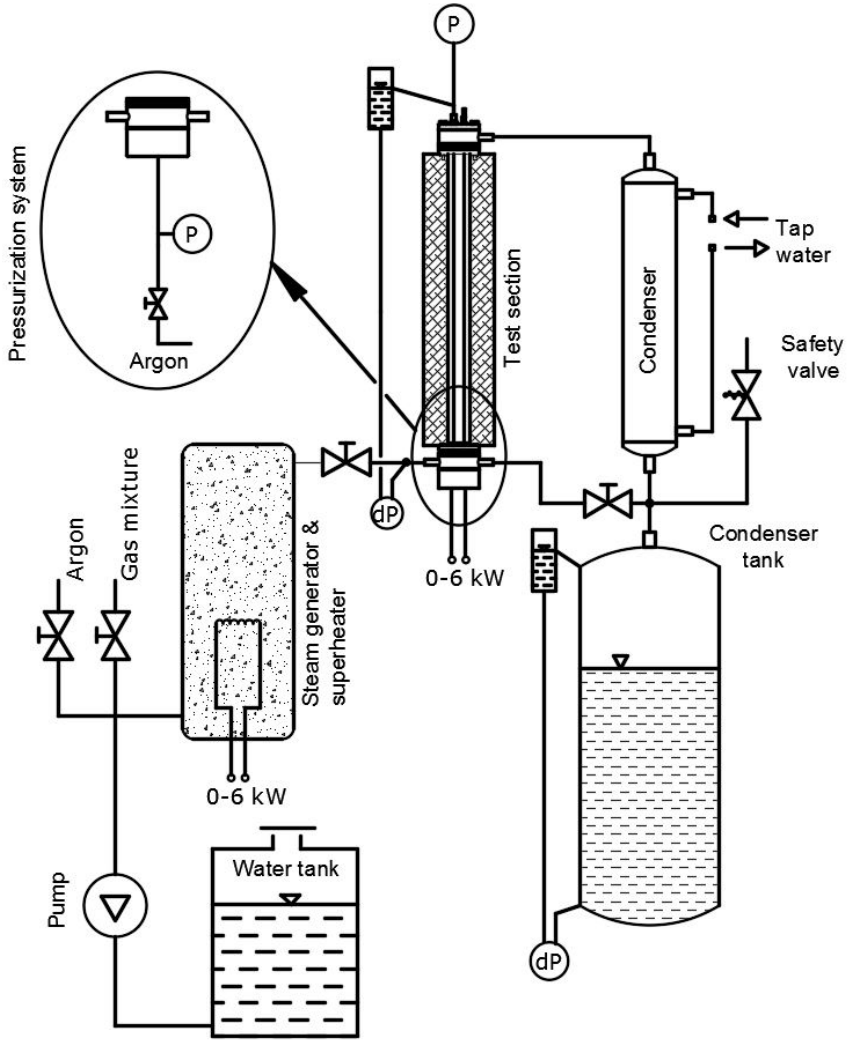
		ATHLET-CD	MAAP5	SCDAPSim (JB)	RELAP/ SCDAPSIM (LEI)	SOCRAT
Total H_2 generation (g)	end of pre-oxidation	11.0 g	10.7 g	14.9 g	6.8 g	8.1 g
	end of reflood	28.2 g	23.5 g	17.9 g	11.8 g	65.5 g

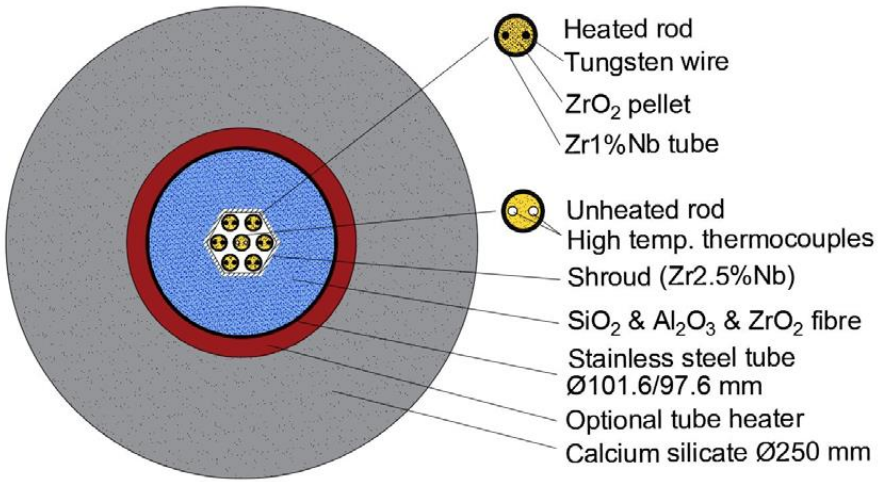
Main findings

- Most codes predict a similar behaviour until reflood.
- The reduction of power during pre-oxidation was necessary to avoid strong oxidation and temperature escalation and to keep the temperature below the melting temperature of the control rod.
- A variation of the flow rates during the air ingress phase was necessary to identify the flow rates which allows at least partial starvation of oxygen, steam and/or nitrogen.
- The external resistance can significantly influence the heat-up.
- The way how to consider the oxidation of the molten control rod can influence the total thermal behaviour of the bundle (ATHLET-CD).

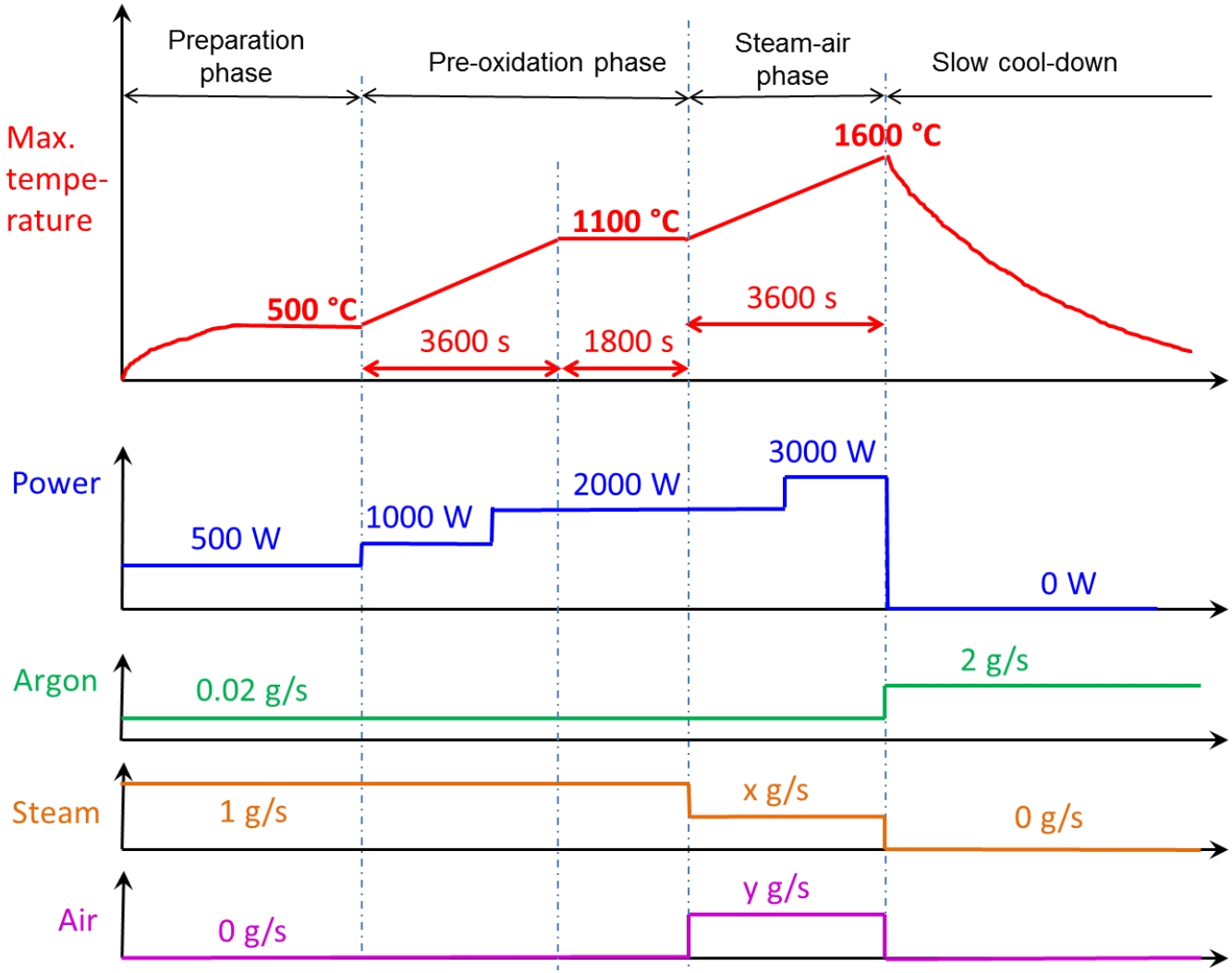
CODEX-AIT3 pre-test benchmark

CODEX facility





CODEX-AIT3 – Proposed test conduct



CODEX-AIT3 test – Scopes and transient

- Scope of the test:
  - 1100 °C maximum temperature during pre-oxidation.
  - Maximum oxide scale should be close to 100 µm at the end of pre-oxidation.
  - Steam and oxygen starvation during air ingress.
  - The duration of air ingress phase should be about 1 hour.
  - Maximum temperature (during air ingress) should be 1600 °C.
- Test transient:
  - Pre-oxidation
  - Intermediate cooldown (as one result of the pre-test simulations to achieve the test targets)
  - Air/steam ingress
  - Cooldown with argon

Participants and codes
- Air oxidation model options -

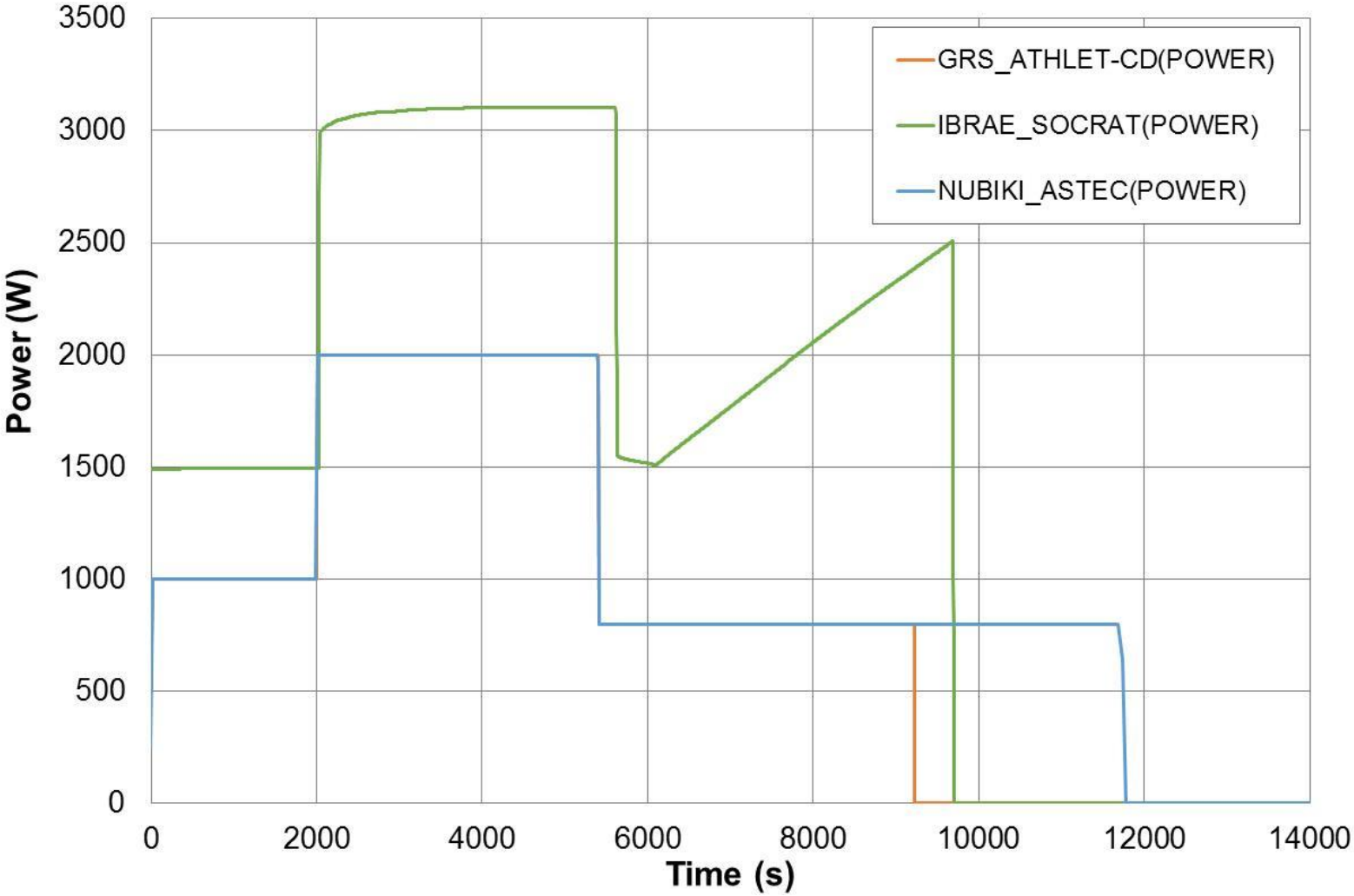
- Participants:
  - GRS ATHLET-CD
  - IBRAE SOCRAT
  - NUBIKI ASTEC

Code Version	ATHLET-CD 31a	SOCRAT v2	ASTEC V2.1.1
Air Oxidation (or Oxygen) Correlation	Steinbrück	Oxygen diffusion enhancement factor depending on ZrN molar fraction. Pure oxygen is treated as vapour except chemical heat.	AEKI
Nitriding Correlation	Hollands	Empirical model depending on nitrogen content in gas mixture.	no
Possibility of nitrides reoxidation (yes or no)	Reoxidation not modelled, indirect consideration with simple porosity model which effects steam oxidation.	no	no

Modelling with Severe Accident Codes
- Boundary Conditions: Mass flow rates -

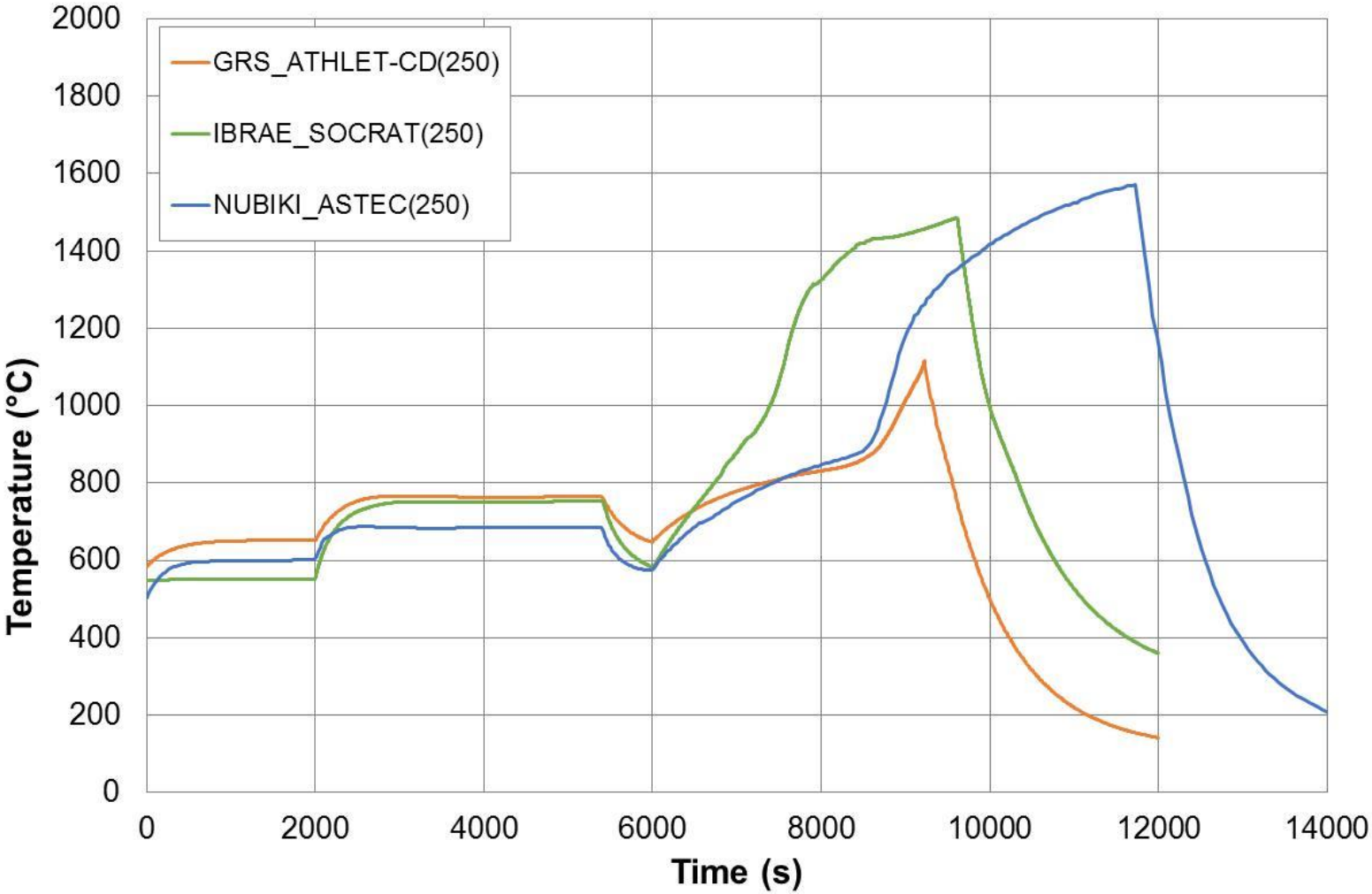
Code Version	ATHLET-CD 31a	SOCRAT v2	ASTEC V2.1.1
steam flow rate during pre-ox (g/s)	1.0	1.0	1.0
argon flow rate during pre-ox (g/s)	1.0	1.0	1.0
steam flow rate during air phase (g/s)	0.02	0.02	0.02
air flow rate during air phase (g/s)	0.05	0.05	0.05
argon flow rate during air phase (g/s)	0.1	0.1	0.1
argon flow rate during cooldown (g/s)	2.0	2.0	2.0

Modelling with Severe Accident Codes
- Boundary Conditions: Electrical power -



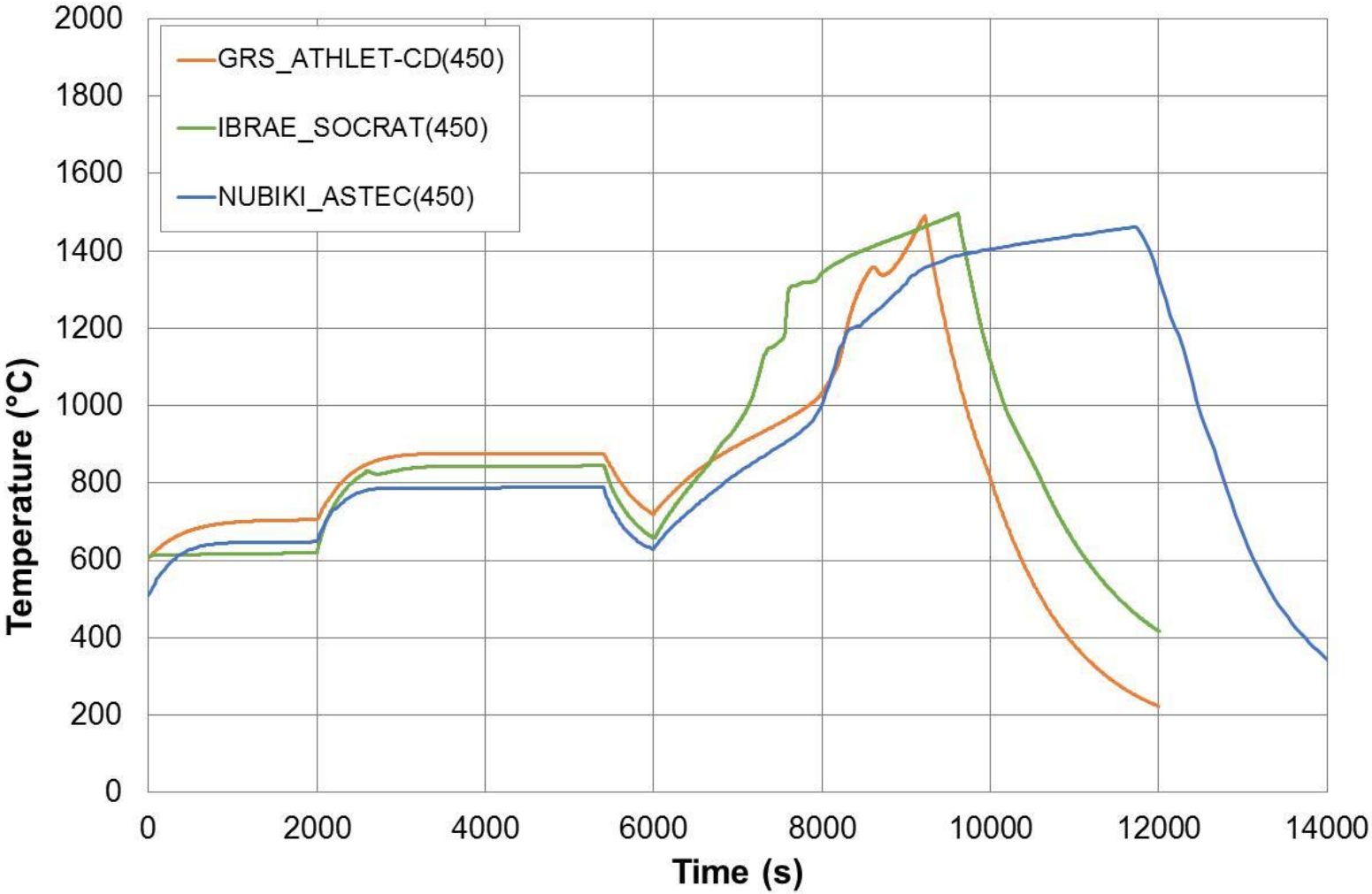
Results of pre-test simulations
- Temperatures (I) -

- Temperatures at 250 mm



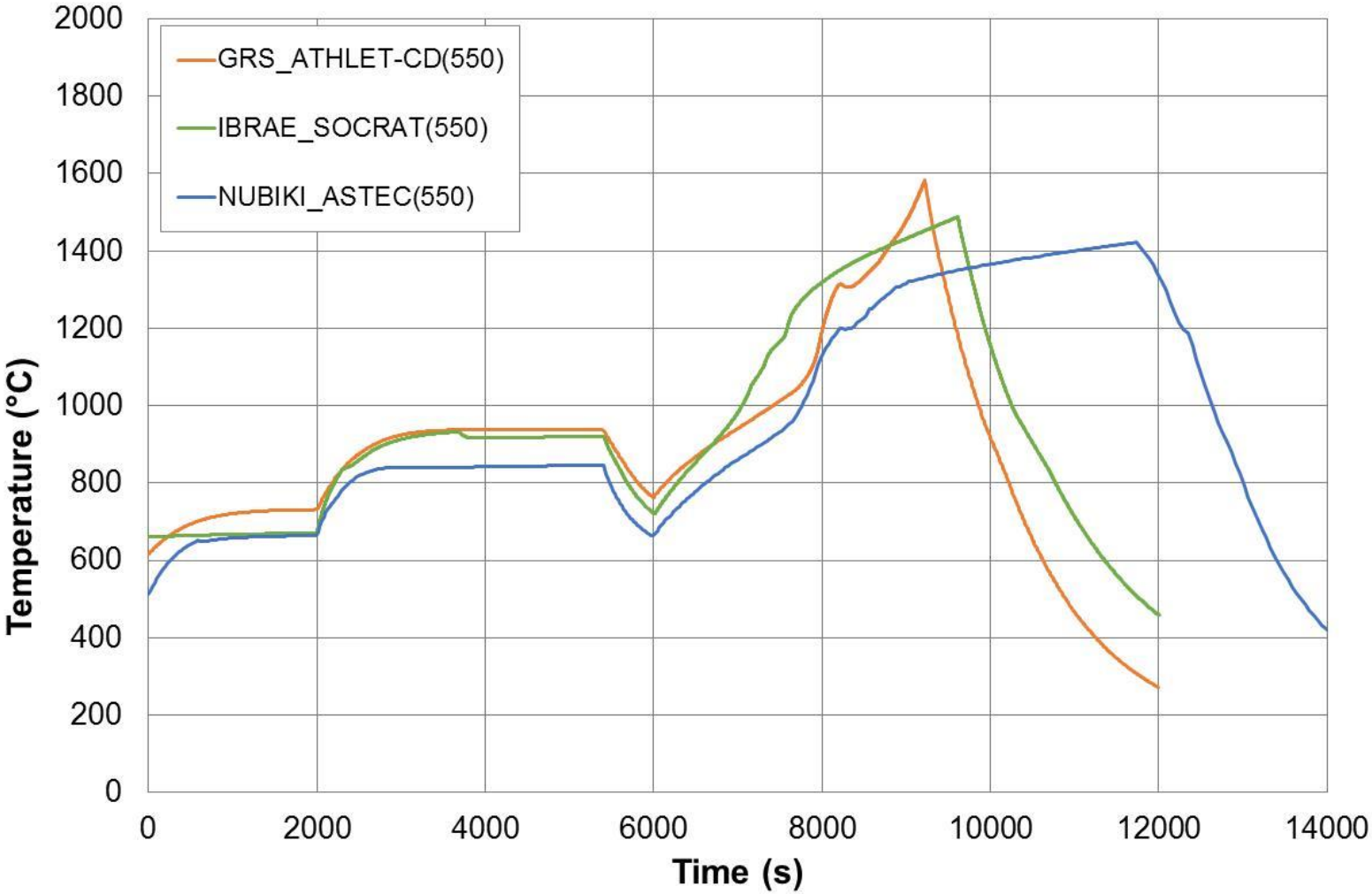
Results of pre-test simulations
- Temperatures (II) -

- Temperatures at 450 mm



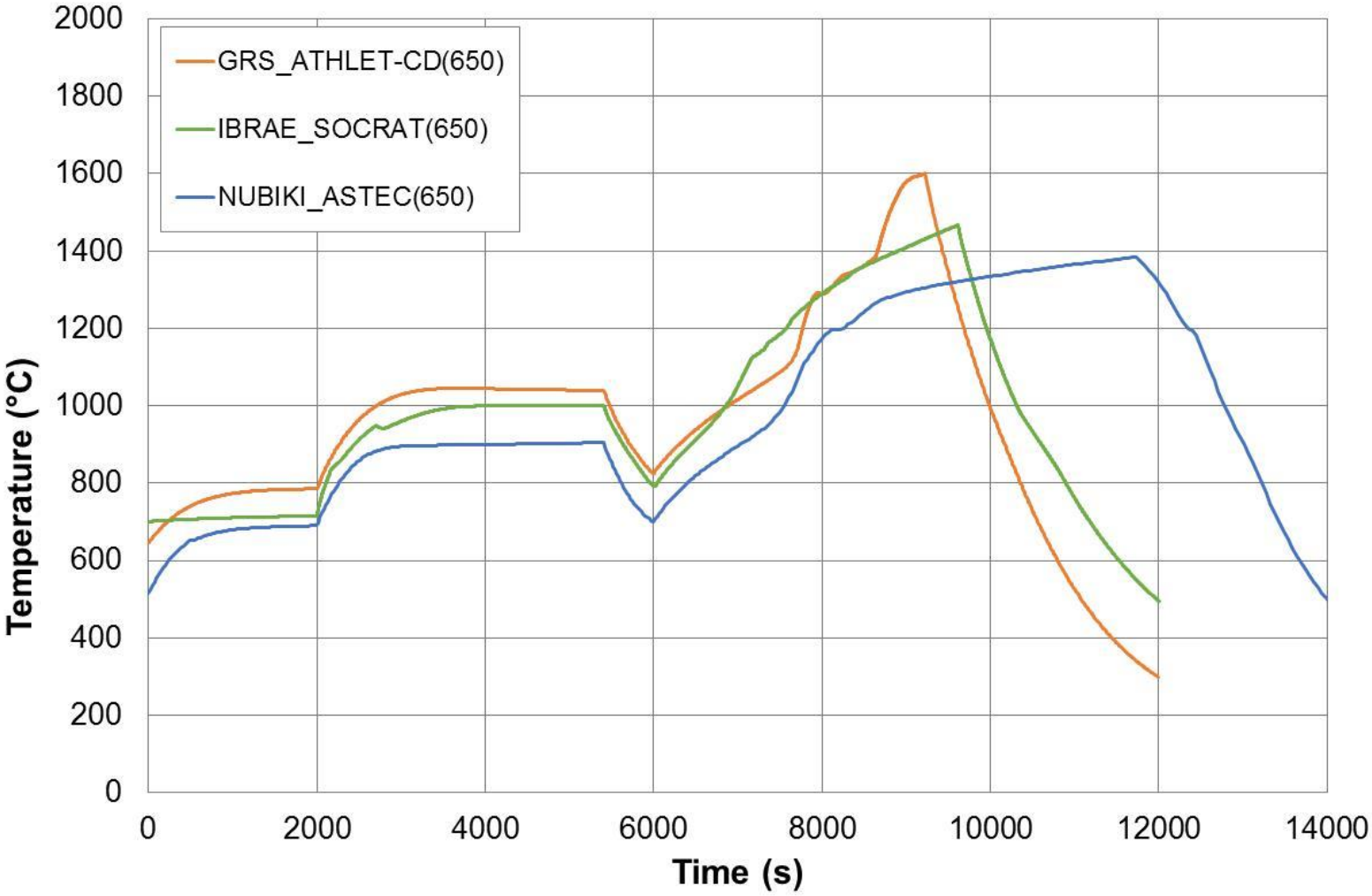
Results of pre-test simulations
- Temperatures (III) -

- Temperatures at 550 mm



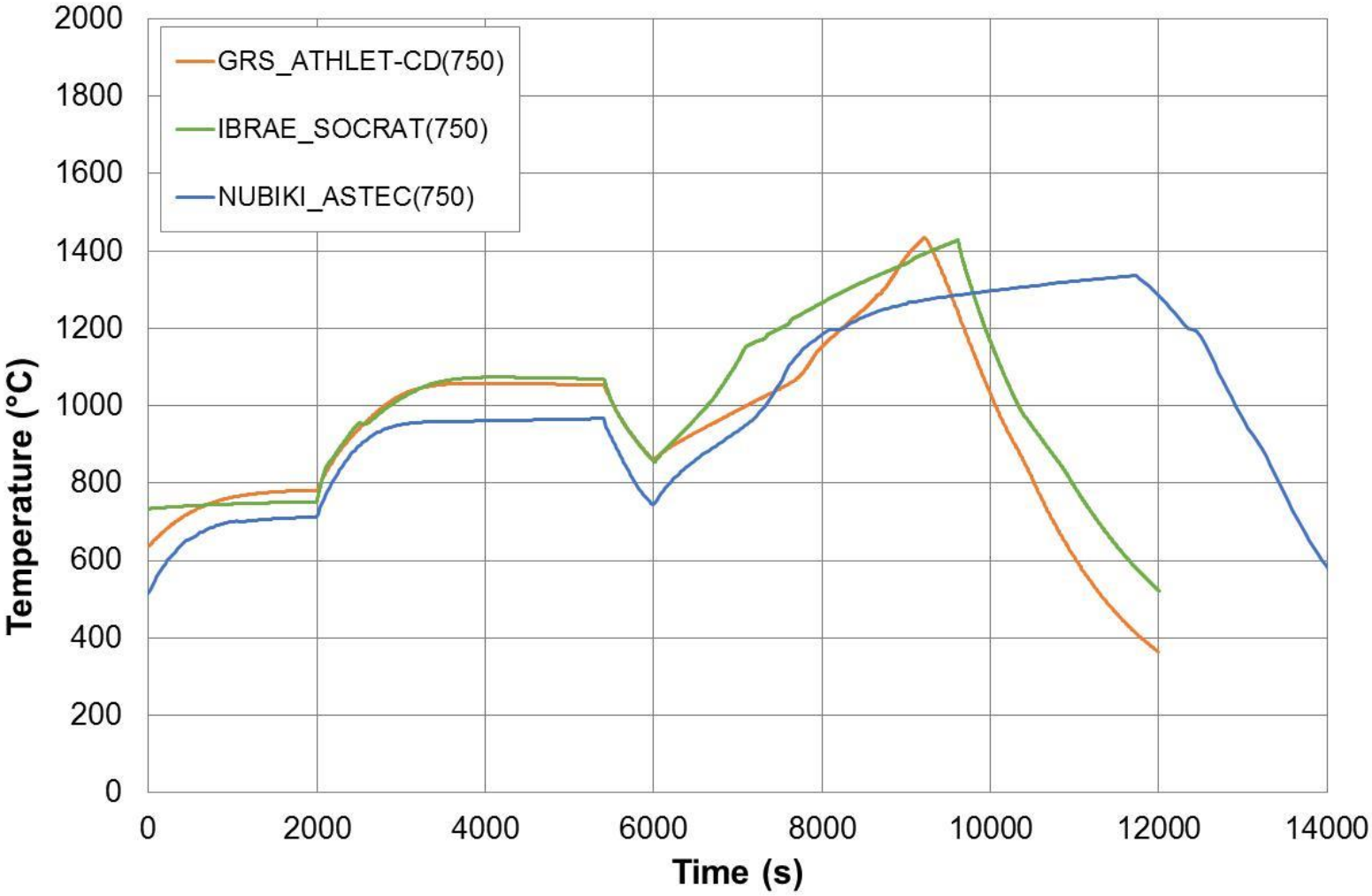
Results of pre-test simulations
- Temperatures (IV) -

- Temperatures at 650 mm

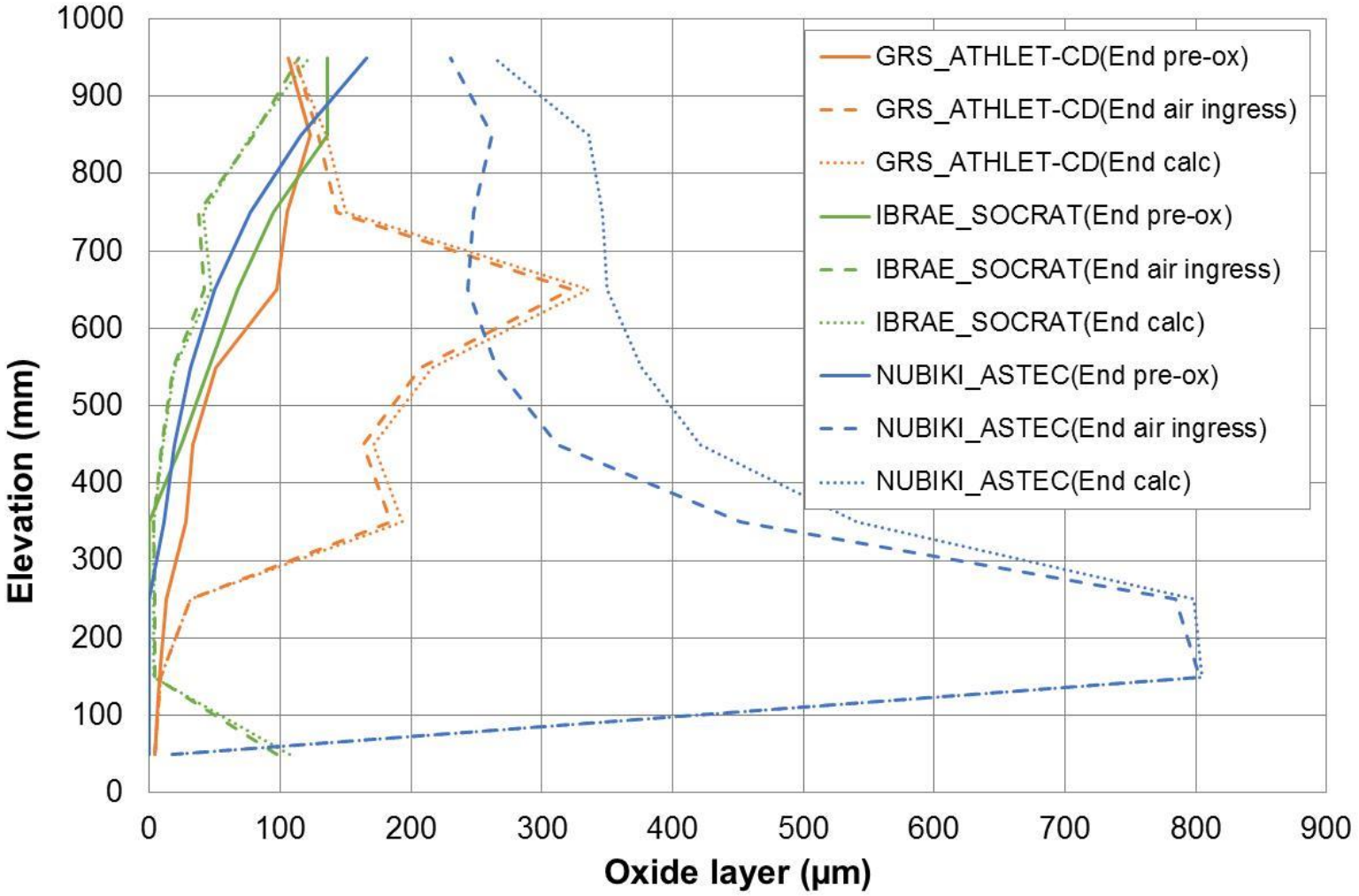


Results of pre-test simulations
- Temperatures (V) -

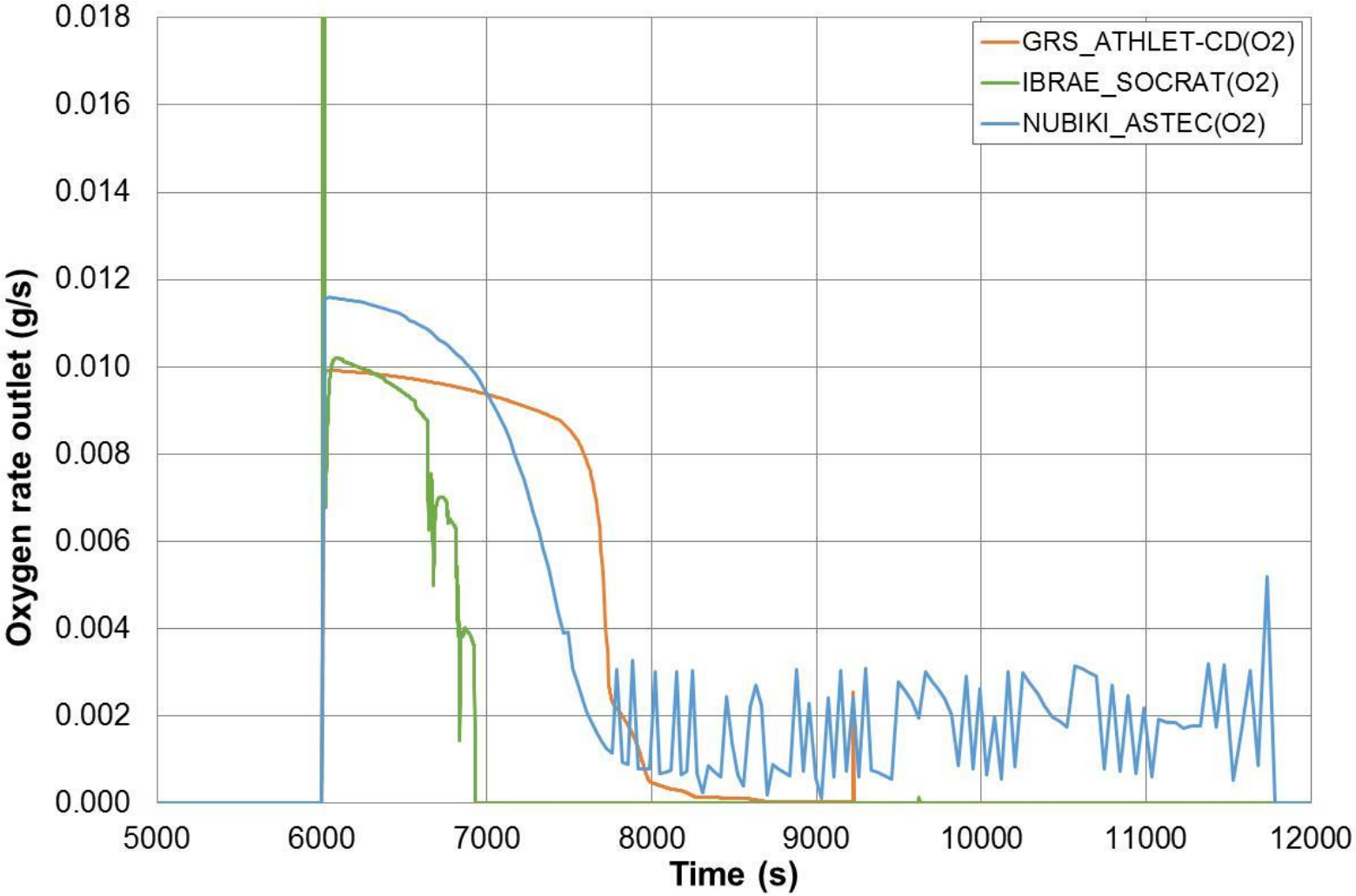
- Temperatures at 750 mm



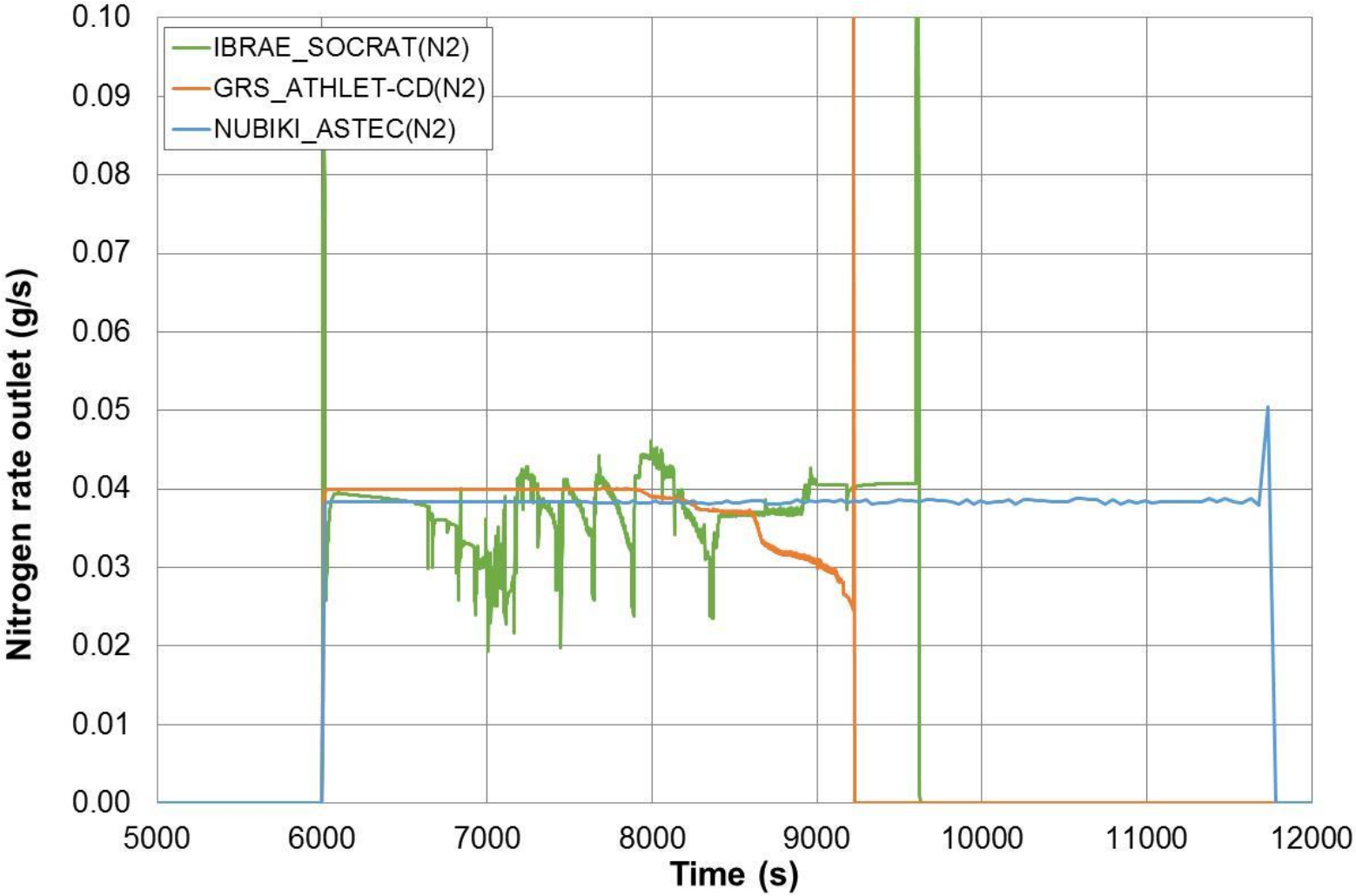
Results of pre-test simulations
- Oxide layer -



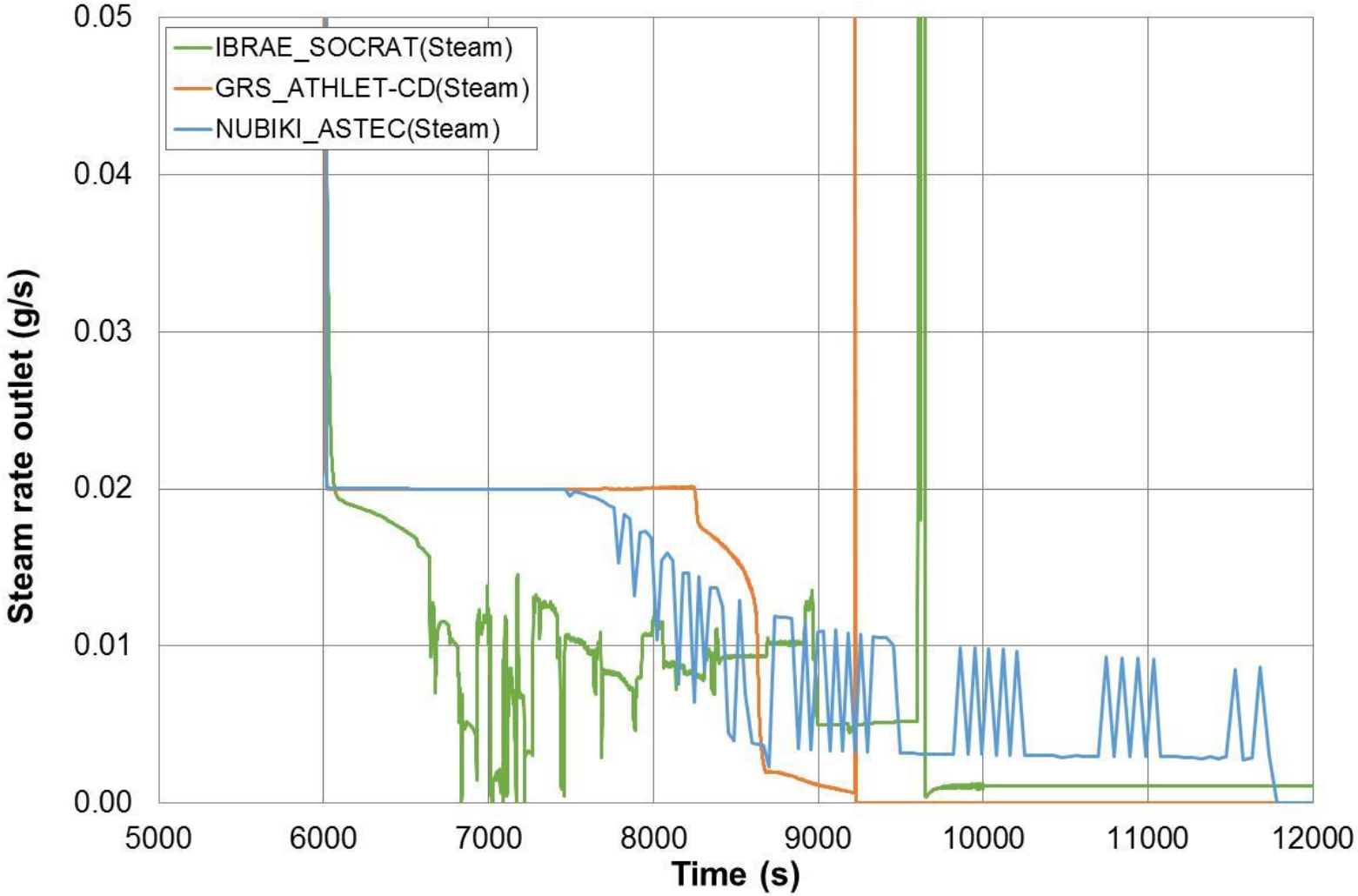
Results of pre-test simulations
- Oxygen flow rate at outlet -



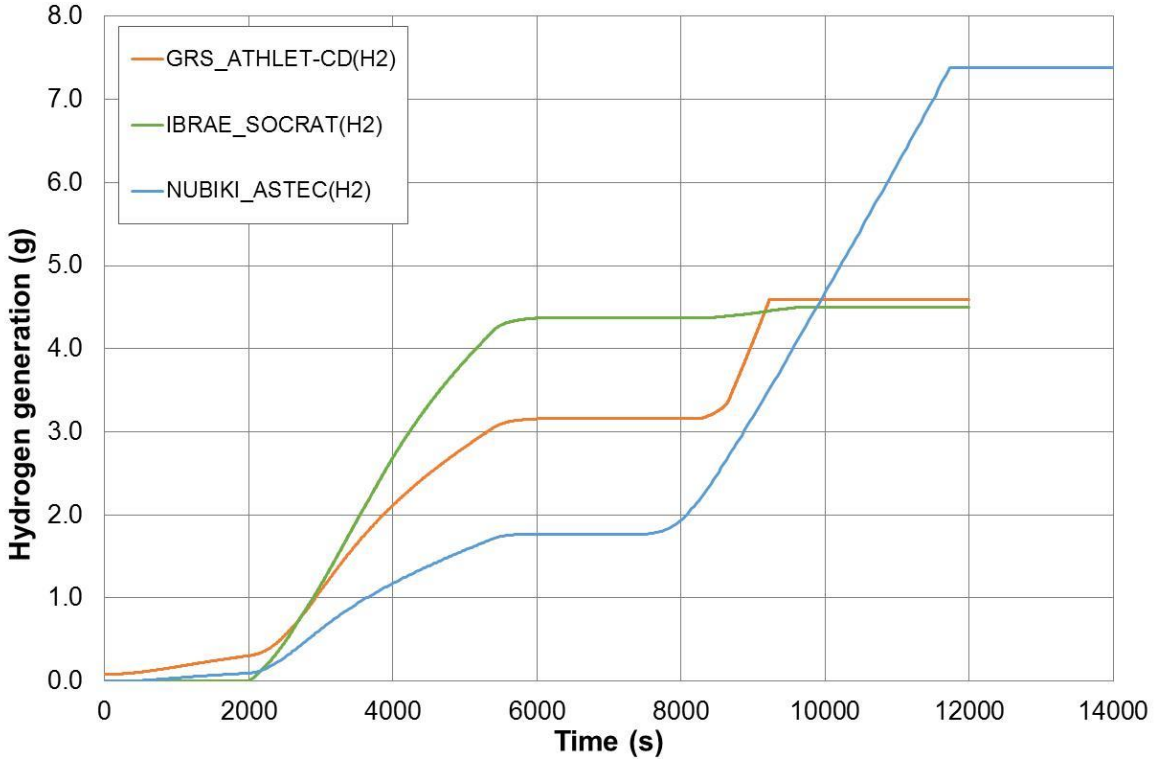
Results of pre-test simulations
- Nitrogen flow rate at outlet -



Results of pre-test simulations
- Steam flow rate at outlet during air ingress -



Results of pre-test simulations
- Hydrogen Generation -



		ATHLET-CD	SOCRAT	ASTEC
Total H_2 generation (g)	end of pre-oxidation	3.2 g	4.2 g	1.72 g
	end of air ingress	4.6 g	4.5 g	7.38 g
	end of simulation	4.6 g	4.5 g	7.38 g

Main findings

- With the new boundary conditions the targets of the experiments were achieved.
- But for SOCRAT simulation the power had to be modified (factor 1.5).
- For the ASTEC simulation the air ingress phase was much longer before cooldown criterion was reached.
- Except the longer time of the ASTEC results the temperature behaviour is quite similar for elevations above 250 mm.
- Oxide layer thicknesses varied strongly between different simulations.

First results of the QUENCH-ALISA bundle test

J. Stuckert, J. Birchley, M. Große, T. Hollands, J. Kalilainen,
J. Laier, T. Lind, J. Moch, U. Peters, M. Steinbrück, Y. Zhang

Experiment QUENCH-18 on air ingress and aerosol release was successfully conducted at KIT on 27 September 2017. This test was performed in the frame of the EC supported ALISA programme. It was proposed by XJTU Xi'an (China) and supported by PSI (Switzerland) and GRS (Germany). The determination of the test protocol was based on planning calculations by participants of the QUESA project: EDF (MAAP5), GRS (ATHLET-CD), IBRAE (SOCRAT), Jon Birchley (SCDAP-SIM) and LEI (RELAP/SCDAPSIM).

The test objectives were agreed following discussions between XJTU, EDF and KIT. The primary aims were to examine the oxidation of M5® claddings (OD=9.5 mm, wall thickness 570 µm) in air/steam mixture following a limited pre-oxidation in steam, and to achieve a long period of oxygen and steam starvations to promote interaction with the nitrogen. QUENCH-18 was thus a companion test to the earlier air ingress experiments, QUENCH-10 and QUENCH-16 (in contrast to QUENCH-18, these two bundle tests were performed without steam flow during the air ingress stage). Additionally, the QUENCH-18 experiment investigated the effects of the presence of two Ag/In/Cd control rods on early-phase bundle degradation (companion test to the QUENCH-13 experiment), and two pressured unheated rod simulators (60 bar, He). The low pressurised heater rods (2.3 bar, similar to the system pressure) were Kr-filled.

In common with the previous QUENCH experiments, the bundle was heated by a series of stepwise increases of electrical power from room temperature to a maximum of ≈900 K in an atmosphere of flowing argon (3 g/s) and superheated steam (3.3 g/s). The bundle was stabilised at this temperature, the electrical power being ≈4.1 kW.

In a first transient, the bundle was heated by power increase to the peak cladding temperature of T_{pct} ≈ 1400 K, reached at 4000 s (with the heat-up rate 0.3 K/s). During this heat-up, claddings of the two pressurised rods were burst at temperature of 1045 K. The attainment of T_{pct} ≈ 1400 K marked the start of the pre-oxidation phase to achieve a maximum cladding oxide layer thickness of up to 150 µm. The power was controlled via small variations between 8.8 kW to 9.4 kW, to maintain more or less constant temperatures. In line with pre-test planning calculations about 11.5 g of hydrogen were produced in this phase which lasted until 6310 s. At this point the power was reduced to 3.8 kW which effected a cooling of the bundle to T_{pct} ≈ 1080 K, as a preparation for the air ingress phase. This phase lasted about 1100 s, until 7400 s. Towards the end of this phase, one of the corner rods was extracted from the test bundle for determination of the oxide thickness axial distribution. Preliminary measurement showed an oxide layer thickness of less than 160 µm (at bundle elevation of 950 mm) indicating maximum oxidation in the bundle was within the target band.

In the subsequent air ingress phase, the steam flow was reduced to 0.3 g/s (7411 s), the argon flow was reduced to 1 g/s (7424 s), and air was injected at 7540 s with the flow rate of 0.2 g/s. The power was maintained at 3.8 kW. The change in flow conditions had the immediate effect of reducing the heat transfer so that the temperatures began to rise again.

After some time measurements demonstrated a gradual increasing consumption of oxygen, starting at about 9000 s. The first Ag/In/Cd aerosol release was registered at 10530 s with corresponding T_{pct} = 1350 K (at 950 mm) and T_{550mm} = 1300 K. The first aerosol release was dominated by Cd bearing aerosols. Later in the transient, a significant release of Ag was observed along with continued Cd release, as well as a small amount of In. High aerosol concentration of several g/Nm^3 was measured until the isolation of the aerosol measurement system at the time of the quench initiation. The sharp increase of the steam consumption with simultaneous hydrogen release was registered at 10550 s. In contrast to the QUENCH-16 test (performed with the air ingress stage without steam flow), oxidation of bundle parts in steam caused release of additional chemical energy (power about 4 kW) and consequently acceleration of bundle heat-up. A strong temperature escalation started at about 10590 s at the bundle elevation of 550 mm and propagated to the upper and lower elevations between 150 and 850 mm. A period of oxygen starvation started at about 10700 s and was followed (about 300 s later) by almost complete steam consumption. Shortly before that time (10640 s), partial consumption of the nitrogen was first observed, indicating the possibility of local oxygen and steam starvation which promoted the onset of nitriding of claddings, shroud, corner rods and absorber guide tubes. Following this the temperatures continued to increase and stabilised at melting temperature of Zr bearing materials until water injection was initiated at 12330 s. Thus there was a period of 1630 s of strong steam and complete oxygen consumptions and hence starvation in at least part of the bundle. The total uptakes of oxygen, steam and nitrogen were 100±3, 450±10 and 120±3 g, respectively. During this starvation period a noticeable production (about 25 mg/s, totally 45±1 g) of hydrogen was measured. Significant release of Kr beginning at 10730 s and continued until quench initiation indicated failures of fuel rod simulators, probably mostly due to melting of claddings.

Toward the achievement of the cladding melting point, a lower part of the second corner rod (below elevation 550 mm) was removed (11014 s). Some local spalling of the oxide scale was observed from preliminary examination at the top of the withdrawn rod segment. The shroud failure with penetration of additional argon flow into the bundle was registered at 11253 s.

The reflood was initiated simultaneously with turning off the air and steam flows, switching the argon injection to the top of the bundle, followed by rapidly filling the lower plenum of the test section with 4 kg of water, and continuing by injecting 50 g/s of water. The power remained at 3.8 kW during the reflood.

Almost immediately after the start of reflood there was a temperature excursion in the mid to upper regions of the bundle (750 to 1150 mm), leading to maximum measured temperatures of about 2450 K. Cooling was established at the middle bundle elevation (550 mm) ca. 70 s after the start of injection, but was delayed further at upper elevations. Reflood progressed rather slowly, perhaps due to the high temperatures and partial bundle melting, and final quench was achieved after about 800 s. A significant quantity of hydrogen was generated during the reflood (238±2 g). Nitrogen release (>54 g) due to re-oxidation of nitrides was also registered.

The facility is now being dismantled prior to post-test examinations. The first videoscope inspection at the position of withdrawn corner rods shows absorber melt relocation to the bundle bottom. The overview of the bundle top shows strong cladding oxidation. In parallel, the recorded test data as well as the results of the aerosol measurements are being processed for further analyses by participants within the ALISA and QUESA projects.



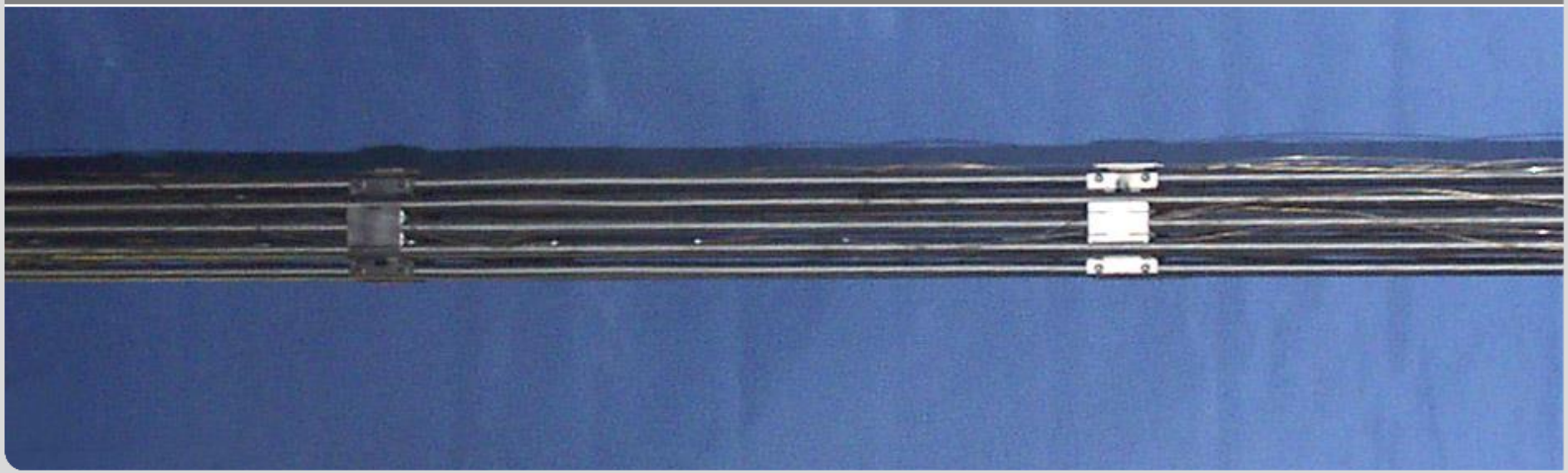
Karlsruhe Institute of Technology

Institute for Applied Materials

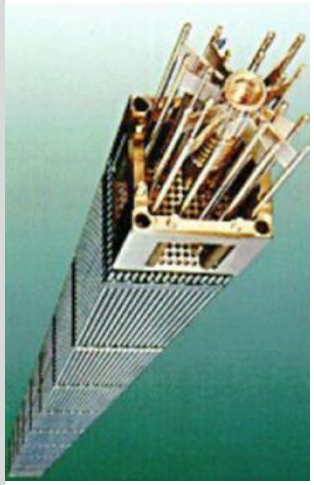
First results of the QUENCH-ALISA bundle test

J. Stuckert, M. Große, J. Laier, J. Moch, U. Peters, U. Stegmaier, M. Steinbrück (KIT),
T. Hollands (GRS), J. Birchley,
J. Kalilainen, T. Lind (PSI),
Y. Zhang (XJTU/China)

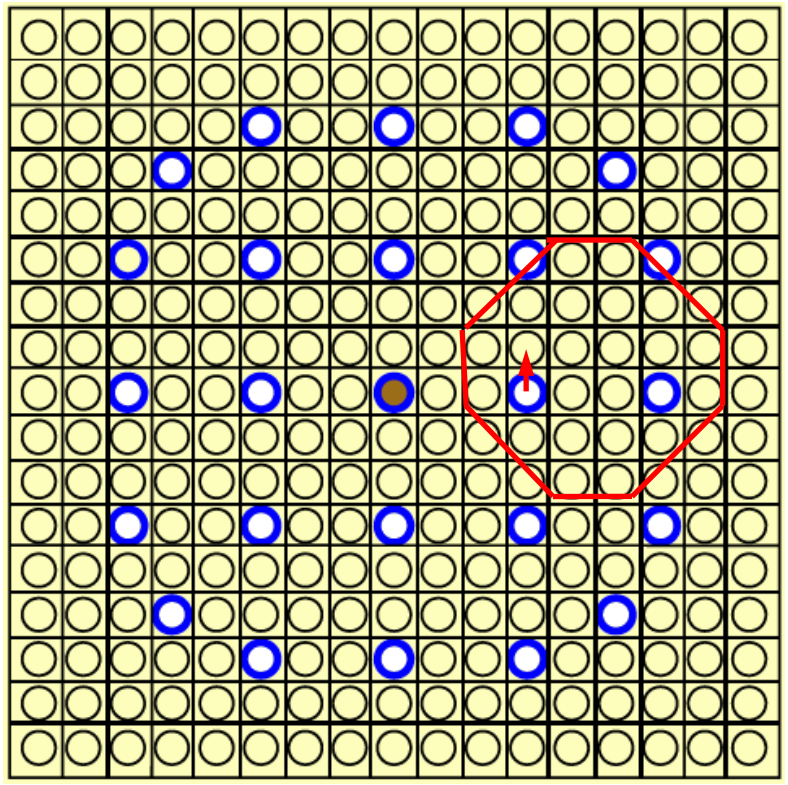
QWS-23, Karlsruhe 2017



Composition of test bundle



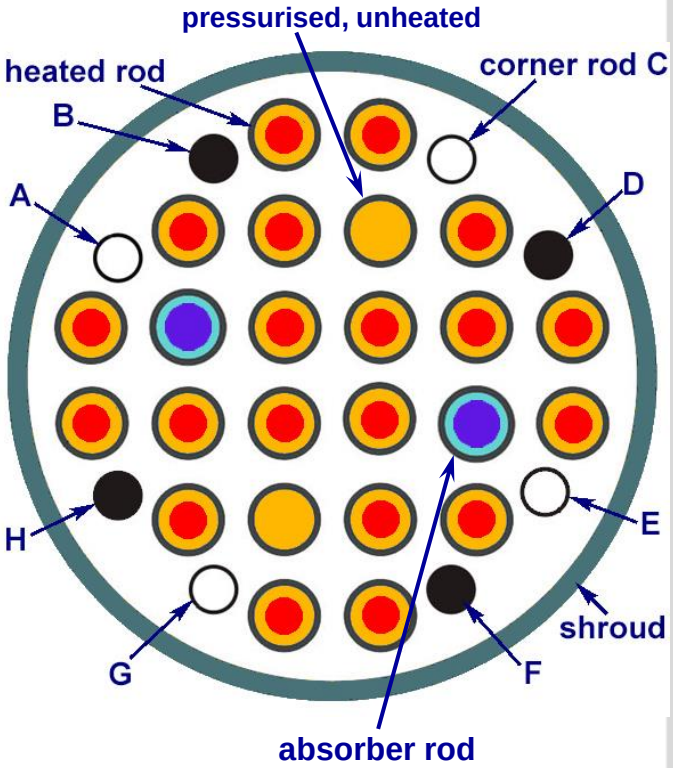
fuel assembly **17x17**



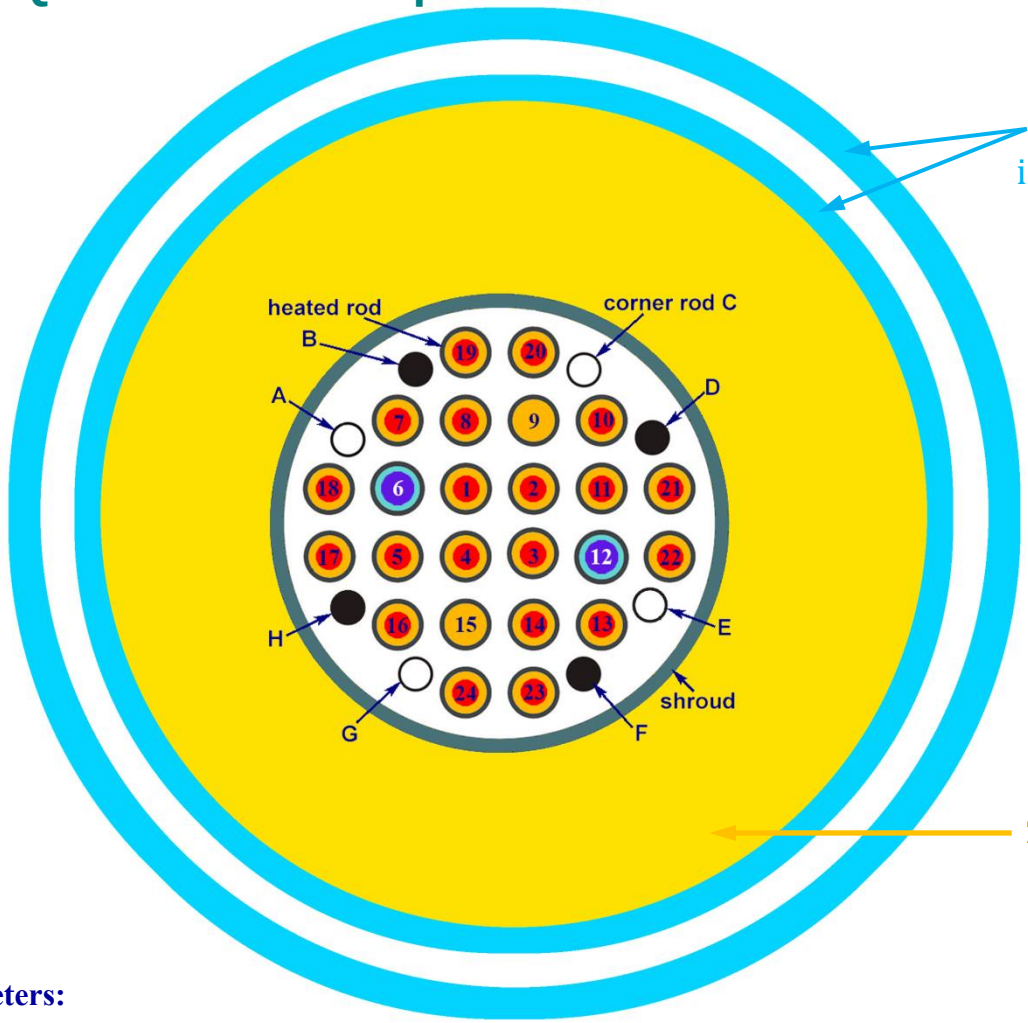
- fuel rod (264)
- guide tube for AgInCd control rod (24)
- instrument thimble

10:1

proposed QUENCH test bundle



QUENCH-ALISA: parameters of test section



Inconel cooling jacket, inner tube ID=158.3 mm

ZrO_2 porous thermal insulation

Bundle geometrical parameters:

- **heated** rods (20): cladding M5® 2200 mm, OD=9.50 mm, ID=8.36 mm; ZrO_2 pellet 10 mm, OD=8.2 mm, ID 5.2 mm; W heater OD=5 mm;
- **pressurized** /to 60 bar/ *unheated* rods (2): M5®cladding; ZrO_2 pellet with OD 8.2 mm;
- **absorber** rods (2): Ag/In/Cd absorber bar with OD = 7.65 mm; stainless steel cladding with OD = 9.68 mm and ID = 7.72 mm; guide tube M5® OD = 12.45 mm, ID = 11.25 mm;
- **corner** rods (8): Zry-4; OD = 6 mm.

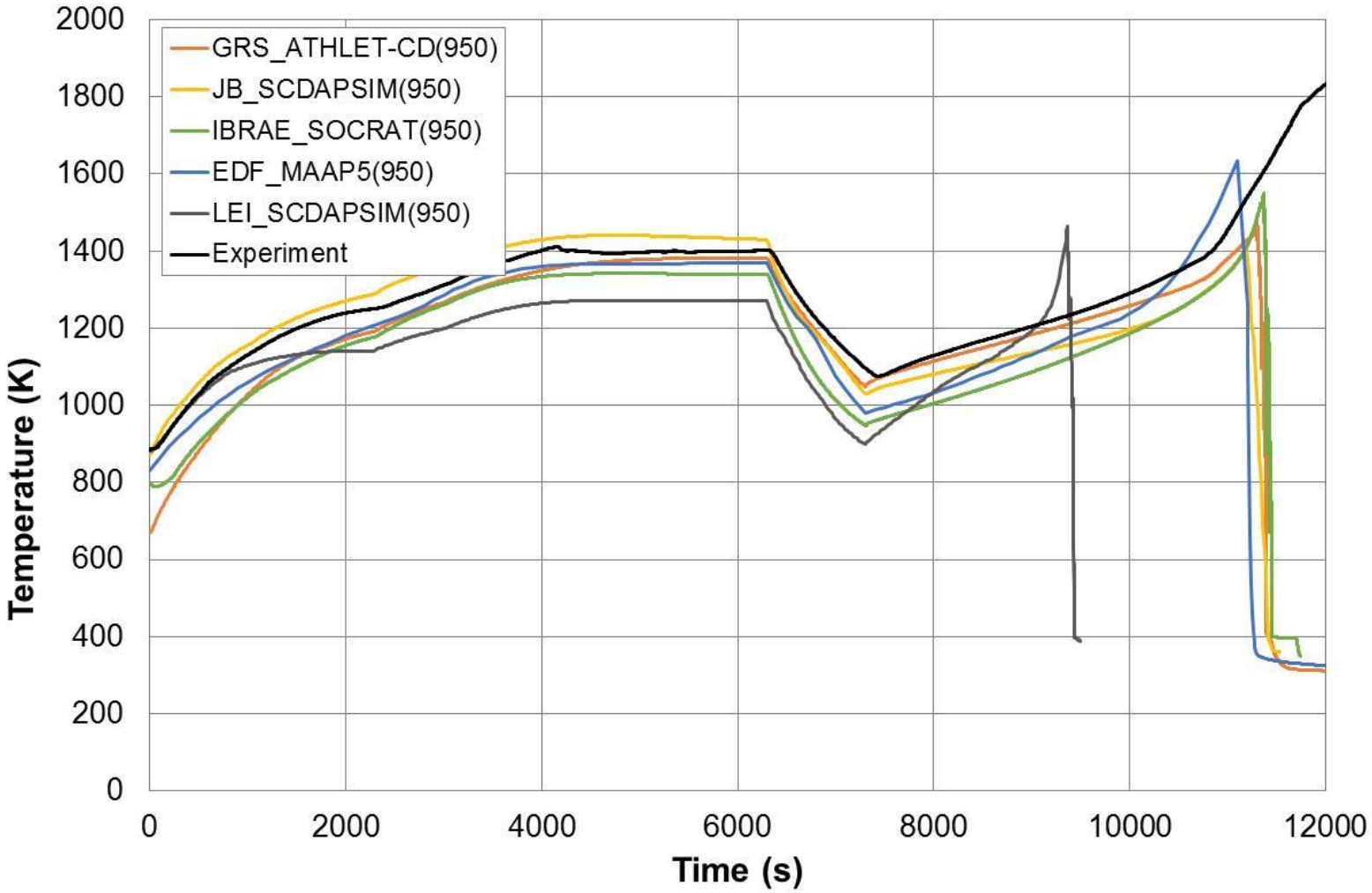
Absorber rod features



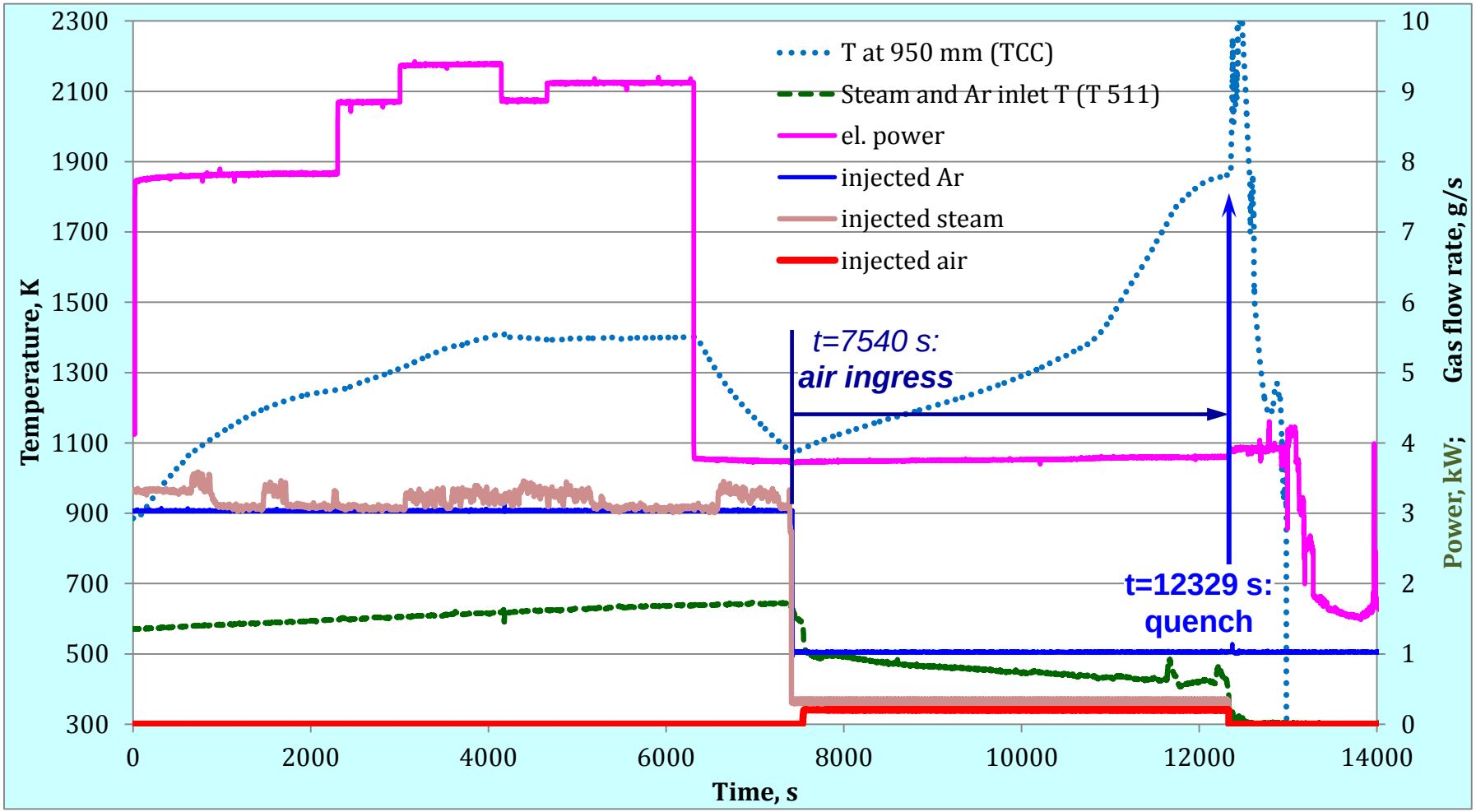
Absorber rod	**material**	**80 Ag, 15 In, 5 Cd (wt-%)**
	dimensions	∅ 7.65 mm, L=1068 mm (Elev. -15 to 1053 mm), ***M ≈ 488 g***
Cladding of absorber rod		**SS**, ∅ 9.68 / 7.72 mm L = 1083 mm (Elev. -20 to 1063 mm)
Guide tube of absorber rod		**M5®** , ∅ 12.45 / 11.25 mm L = 1187 mm (Elev. -42 to 1145 mm) Holes for coolant penetration (2x4): ∅4 mm (Elev. -34 and +1137 mm)
Internal rod pressure of absorber rod		0.12 MPa abs. (**He**)

Results of pre-test simulations

Temperatures at 950 mm

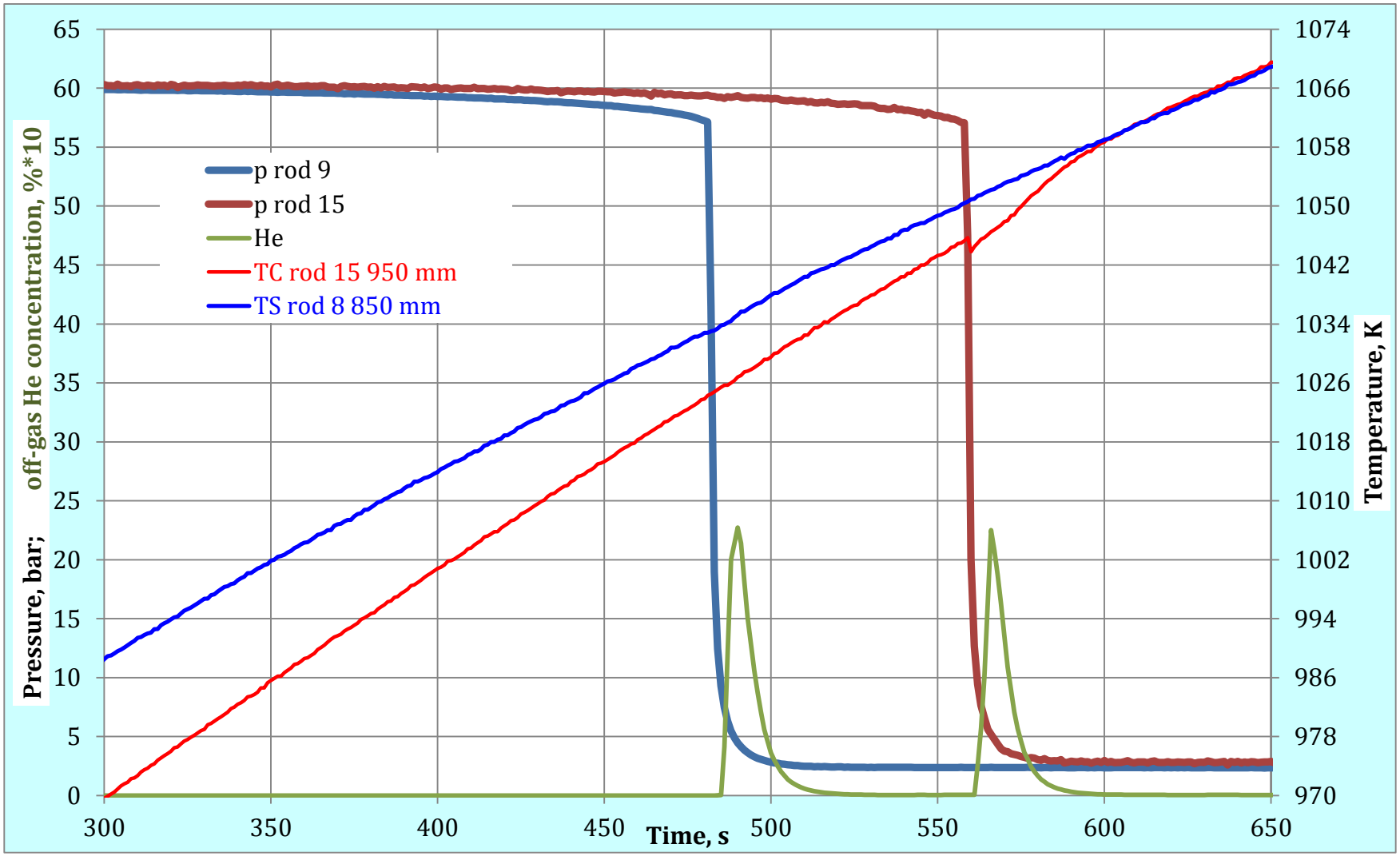


Input parameters of the QUENCH-ALISA (QUENCH-18) test



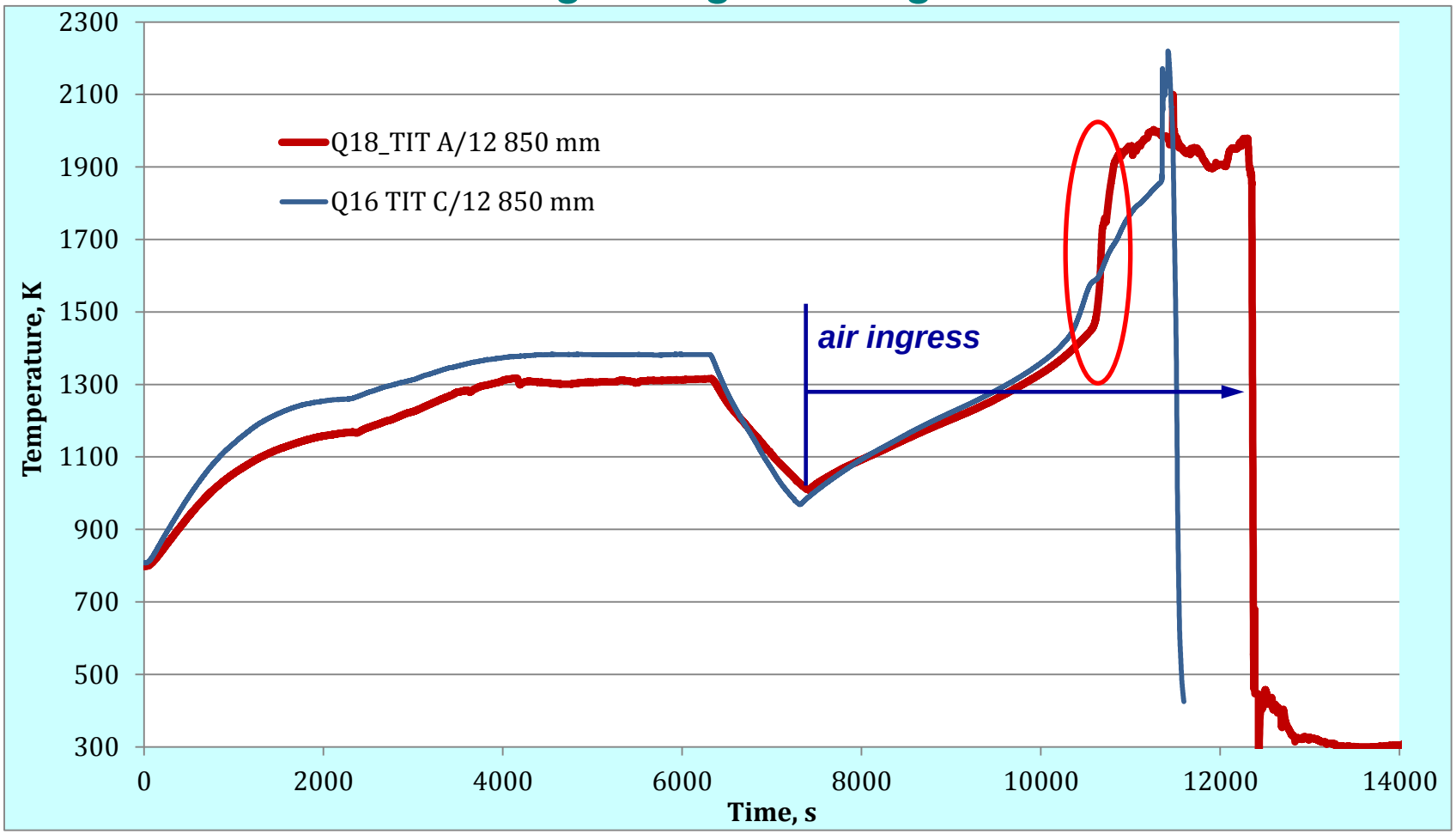
Important feature of QUENCH-18: steam flow during air ingress

Burst of pressurised rods #9 (at 850 mm?) and #15 (at 950 mm)



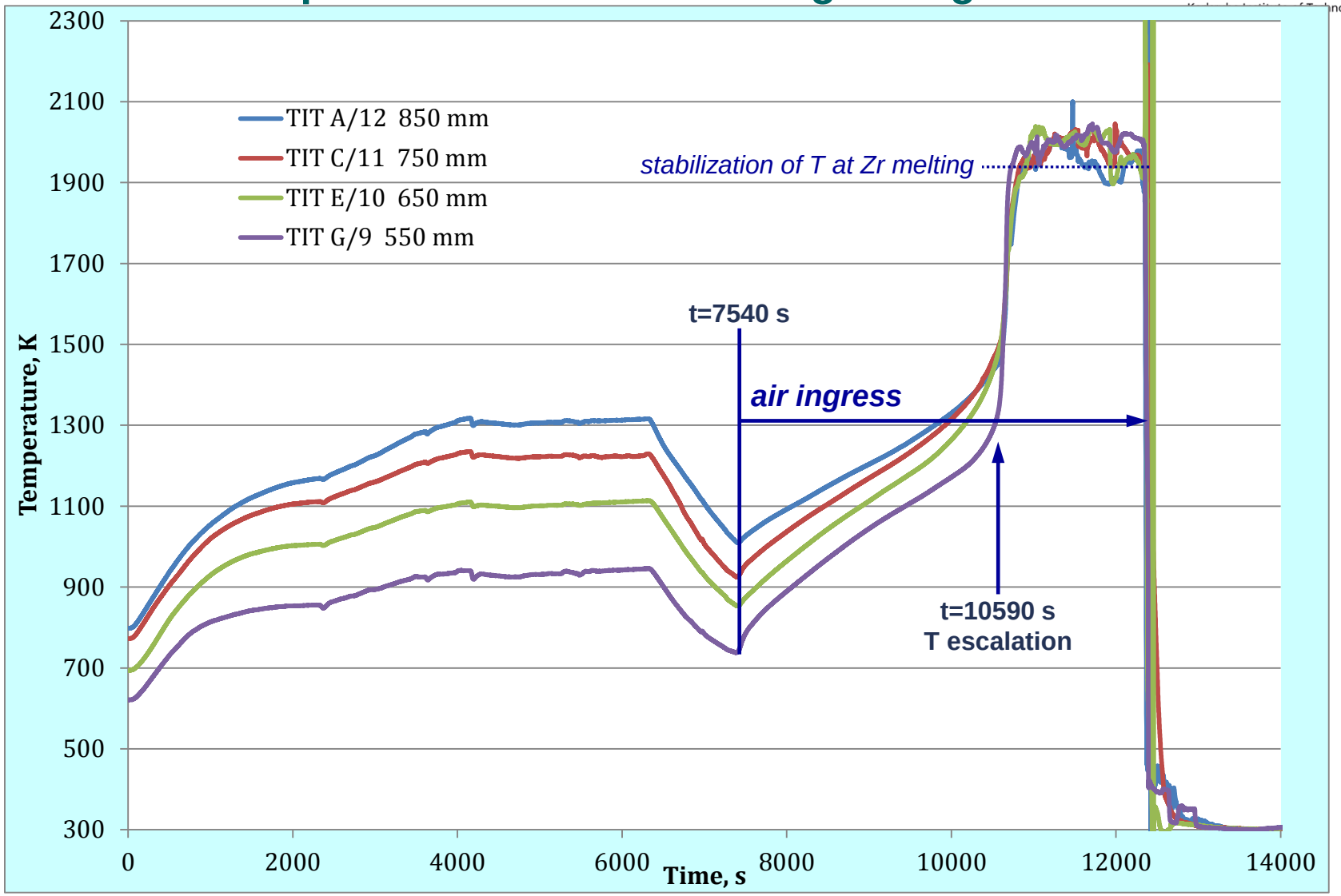
Burst temperature: 1045 K (rod #15) **at heat-up rate 0.3 K/s**

QUENCH-18 (air + steam) vs. QUENCH-16 (only air): accelerated temperature escalation during air ingress stage

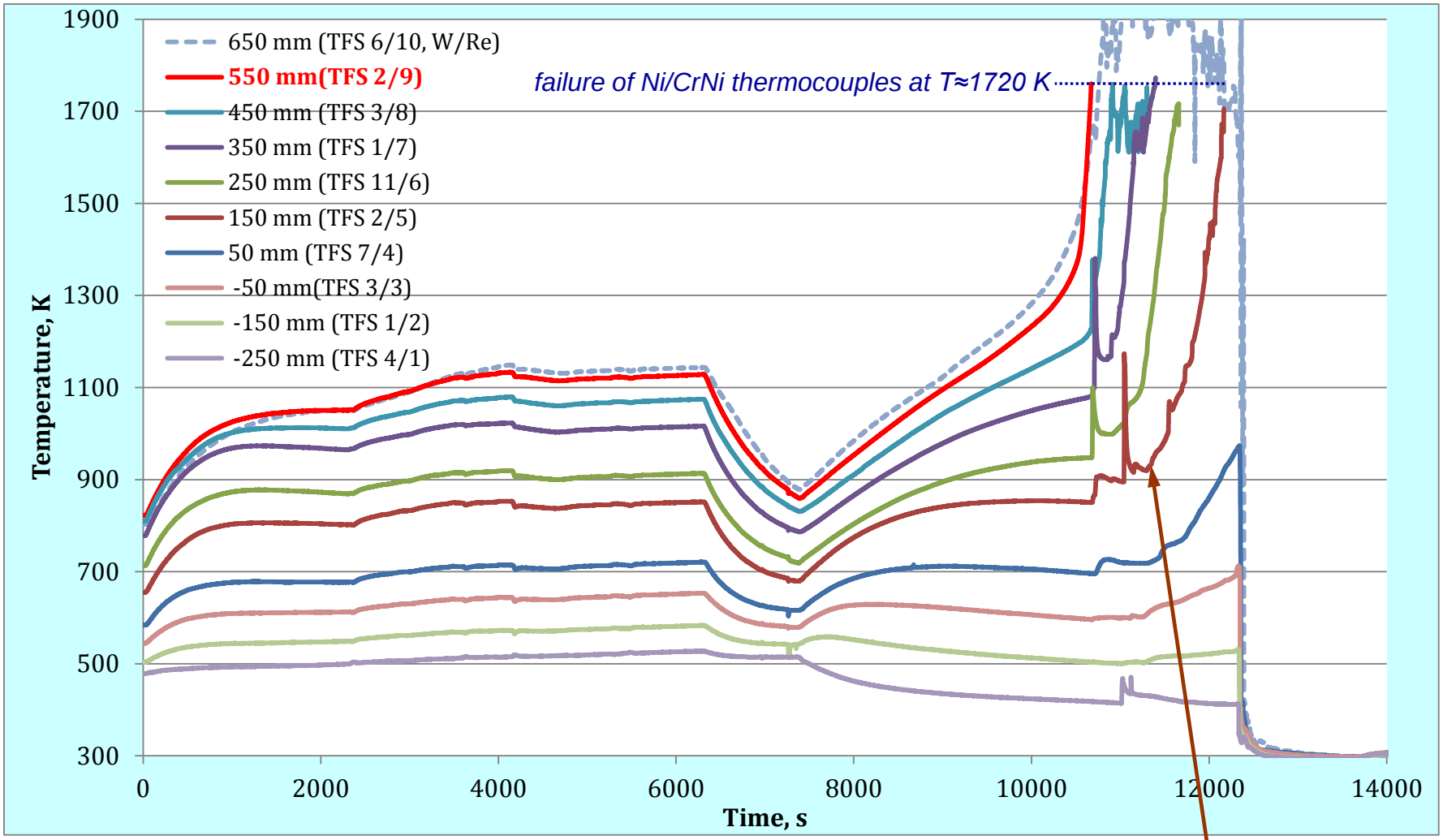


Additional chemical power (≈ 4 kW) due to cladding oxidation in steam during air ingress

Readings of sheathed thermocouples (corner rods): temperature escalation during air ingress



Readings of clad surface thermocouples (Ni/CrNi) at lower elevations

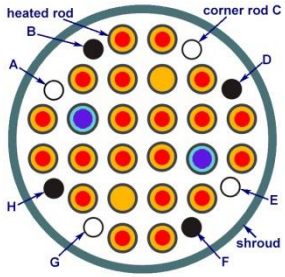


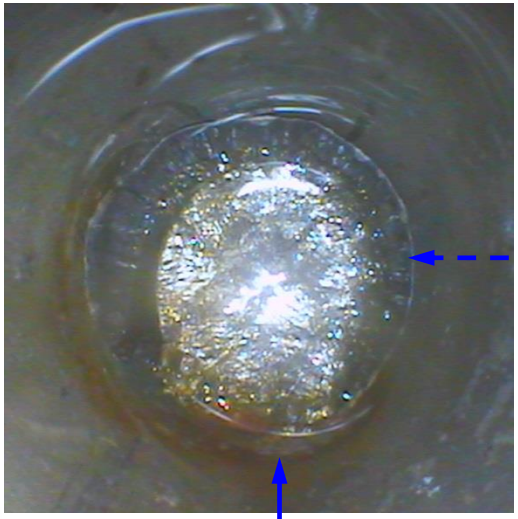
Lowest elevation with strong temperature escalation during air ingress: 150 mm

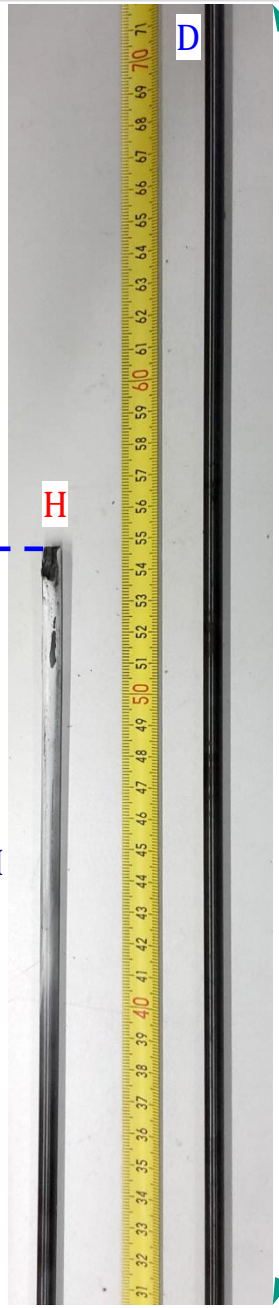
Withdrawn corner rods:

rod D before air ingress,

rod H on cessation of T escalation at 300 mm

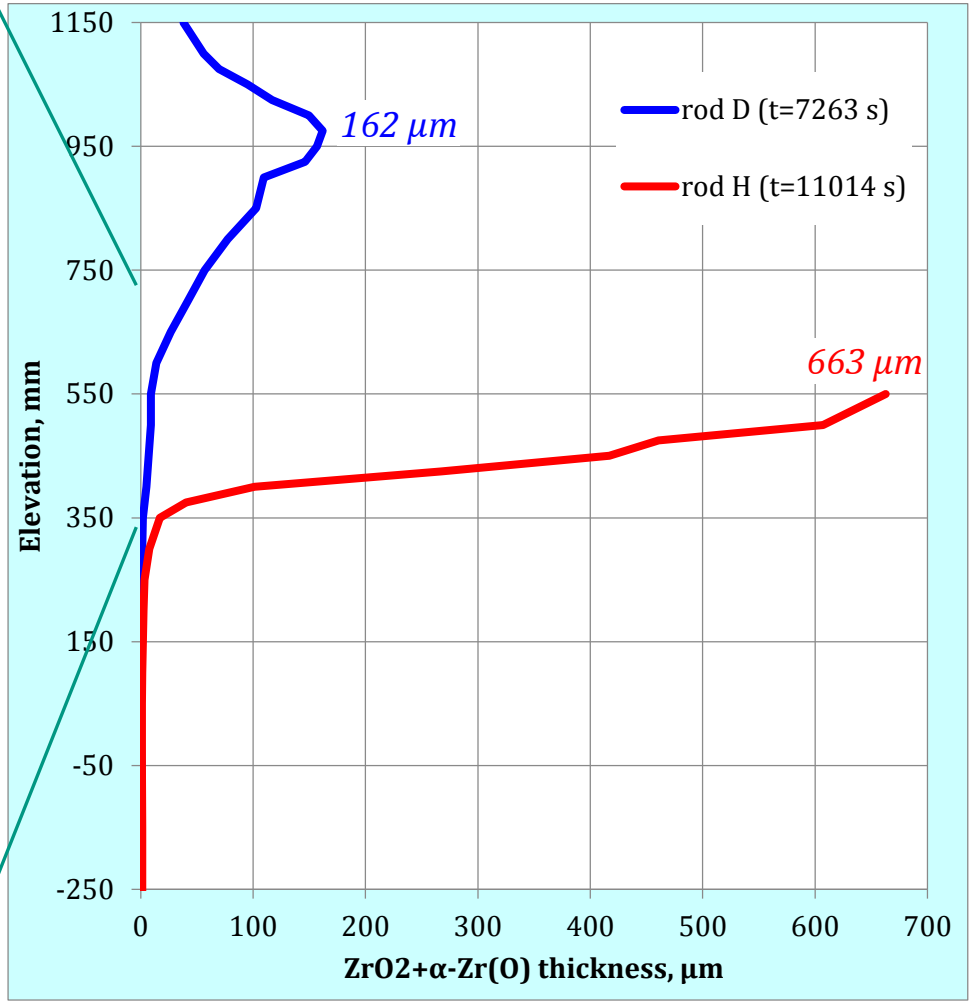






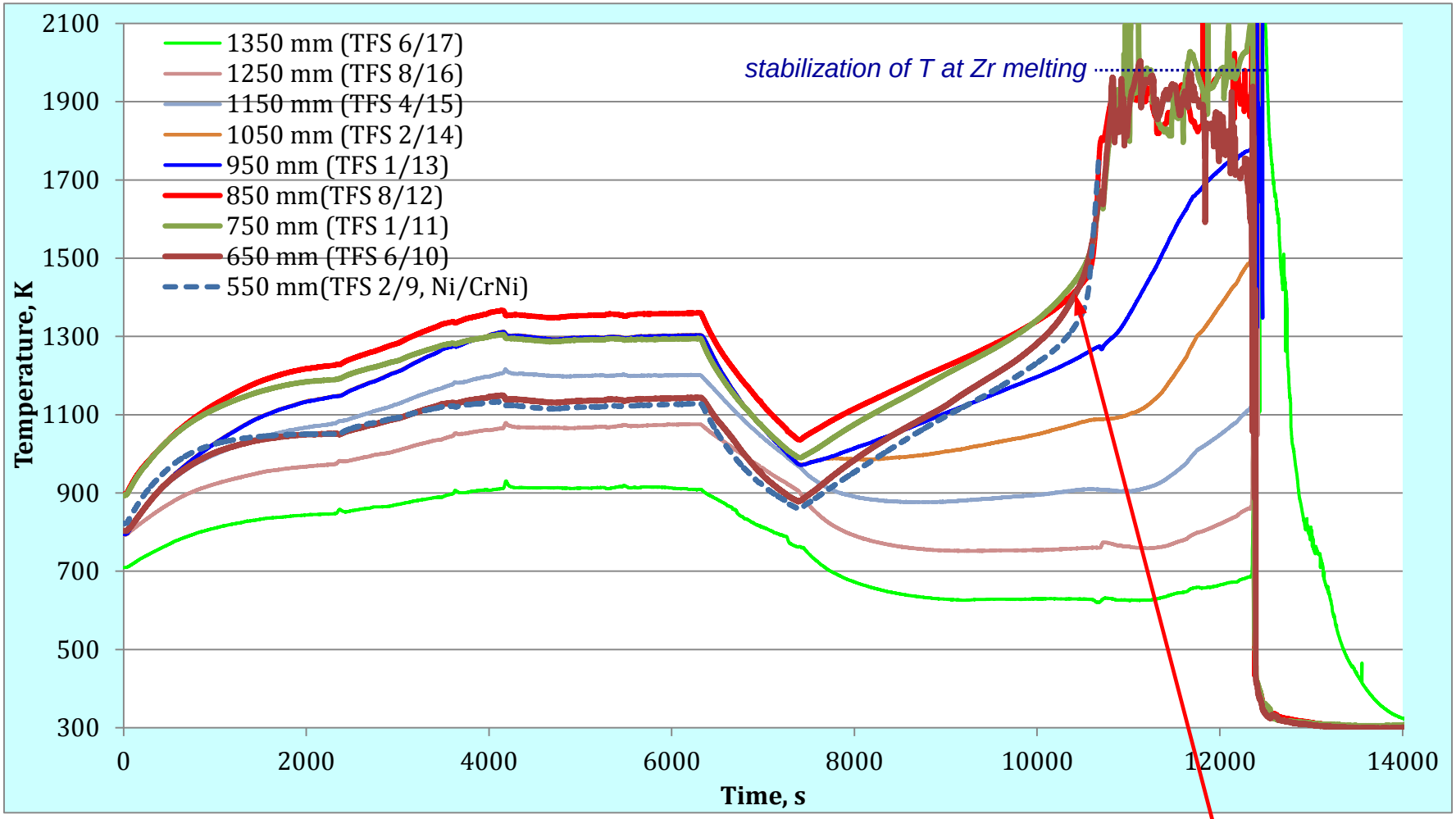
rupture position of corner rod H (550 mm)
relocated to -475 mm during the rod withdrawn

(videoscope observation through the guide tube of the corner rod H)



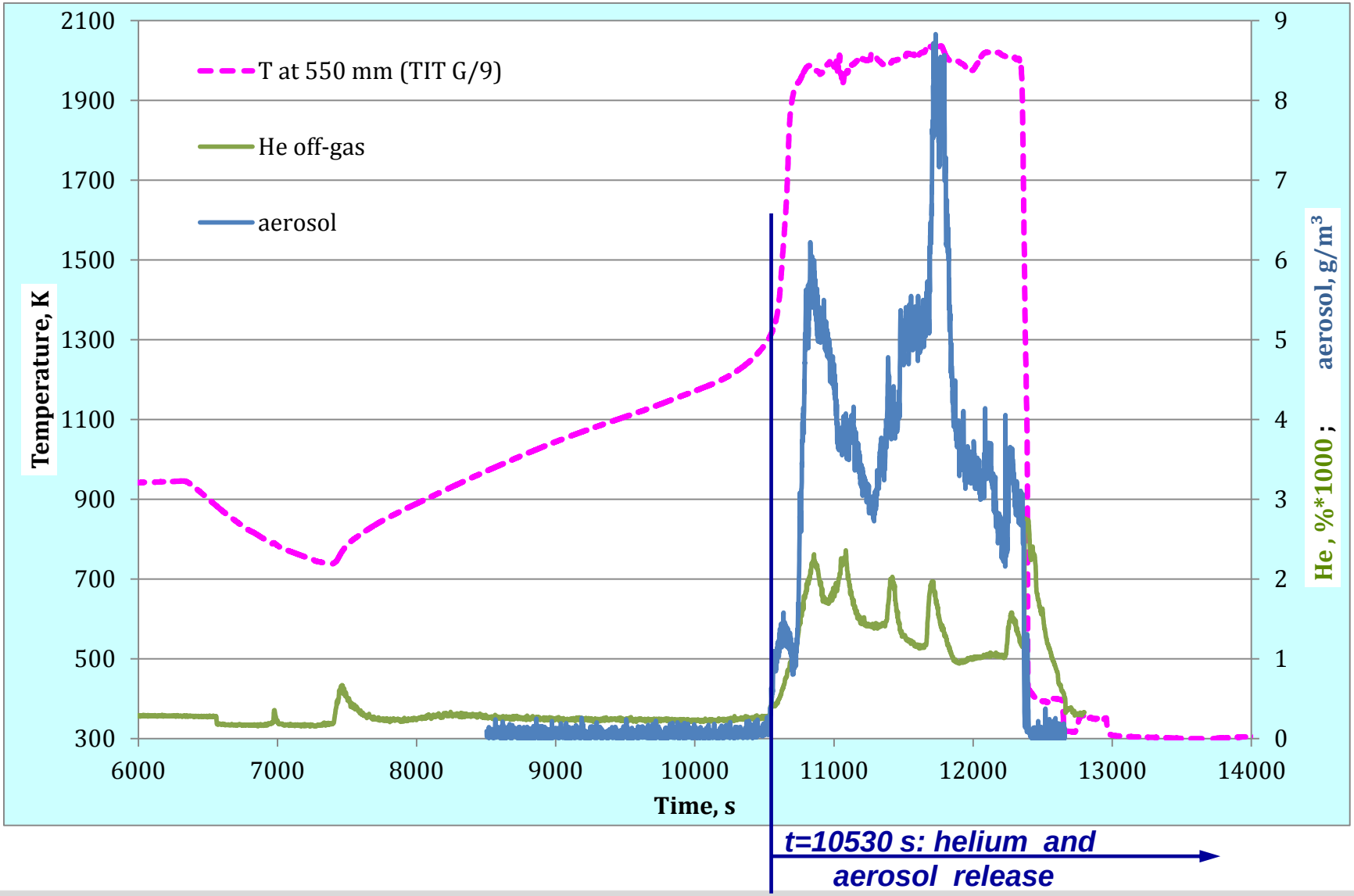
increased oxidation above 250 mm

Readings of clad surface thermocouples (W/Re) at upper elevations

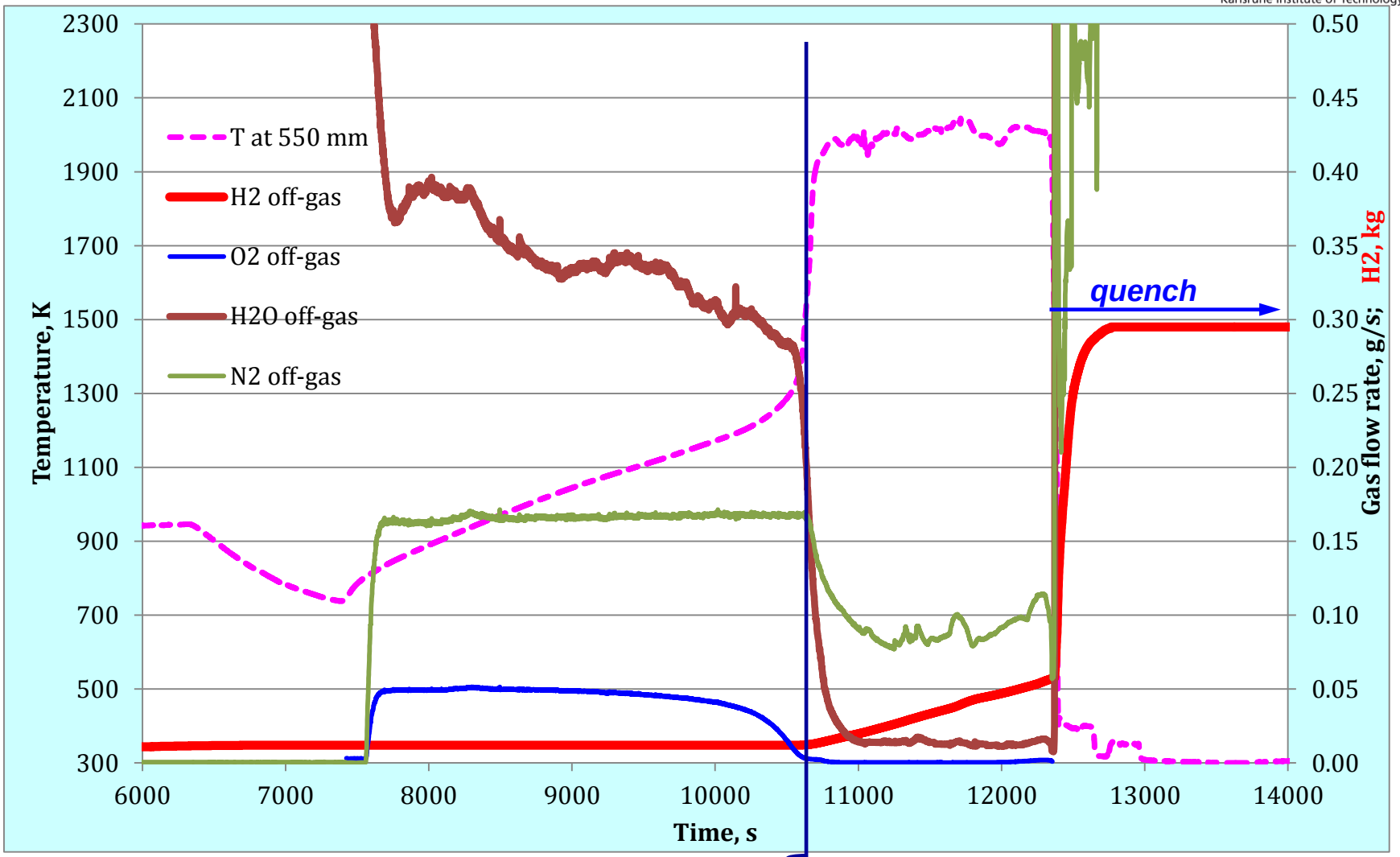


Highest elevation with strong temperature escalation during air ingress: 850 mm

Failure of absorber rods



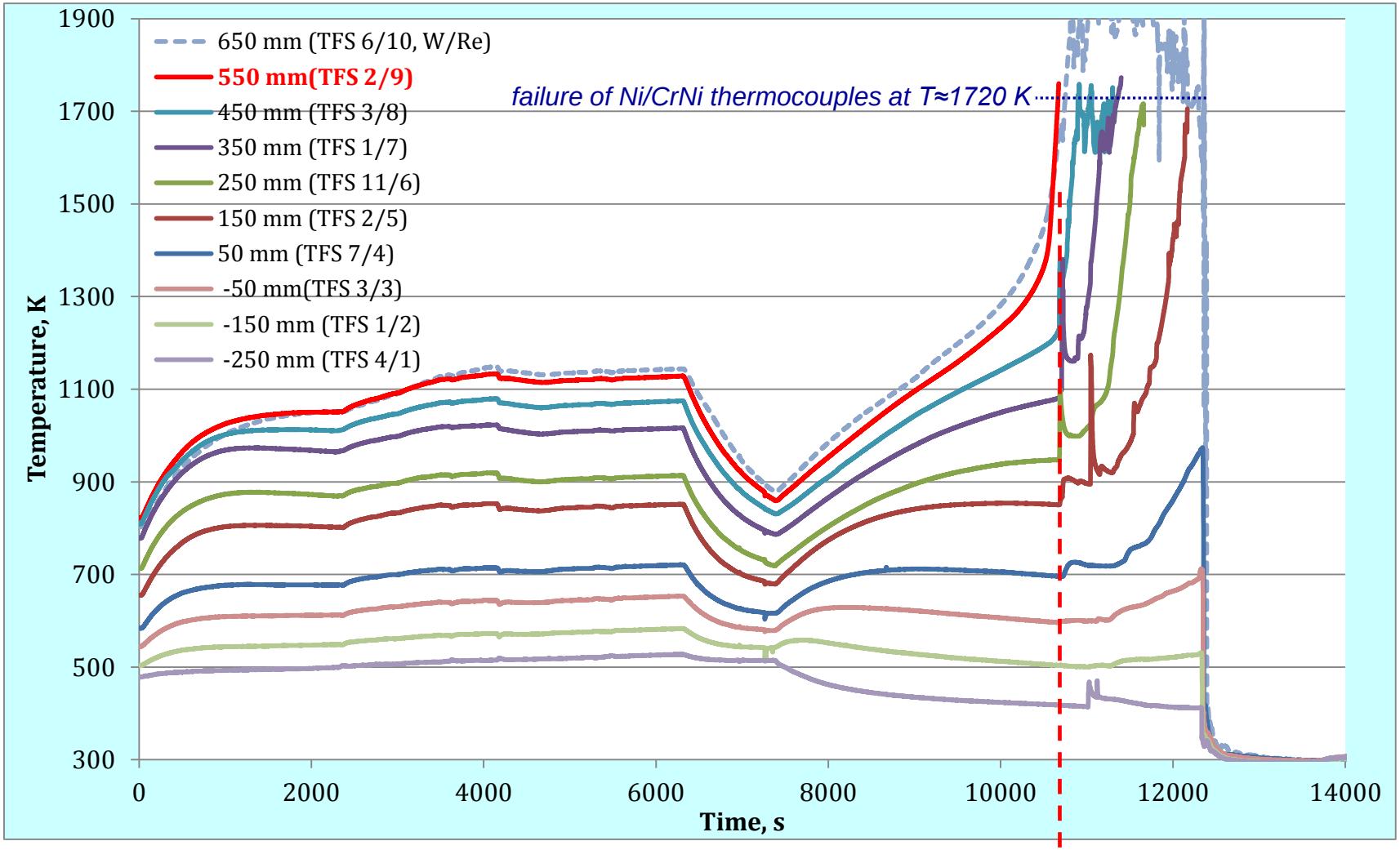
Outlet gas behaviour during air ingress: starvation phenomena



consumed oxygen: 100 g; consumed nitrogen: 120 g;
consumed steam: 450 g; released hydrogen: 45 g

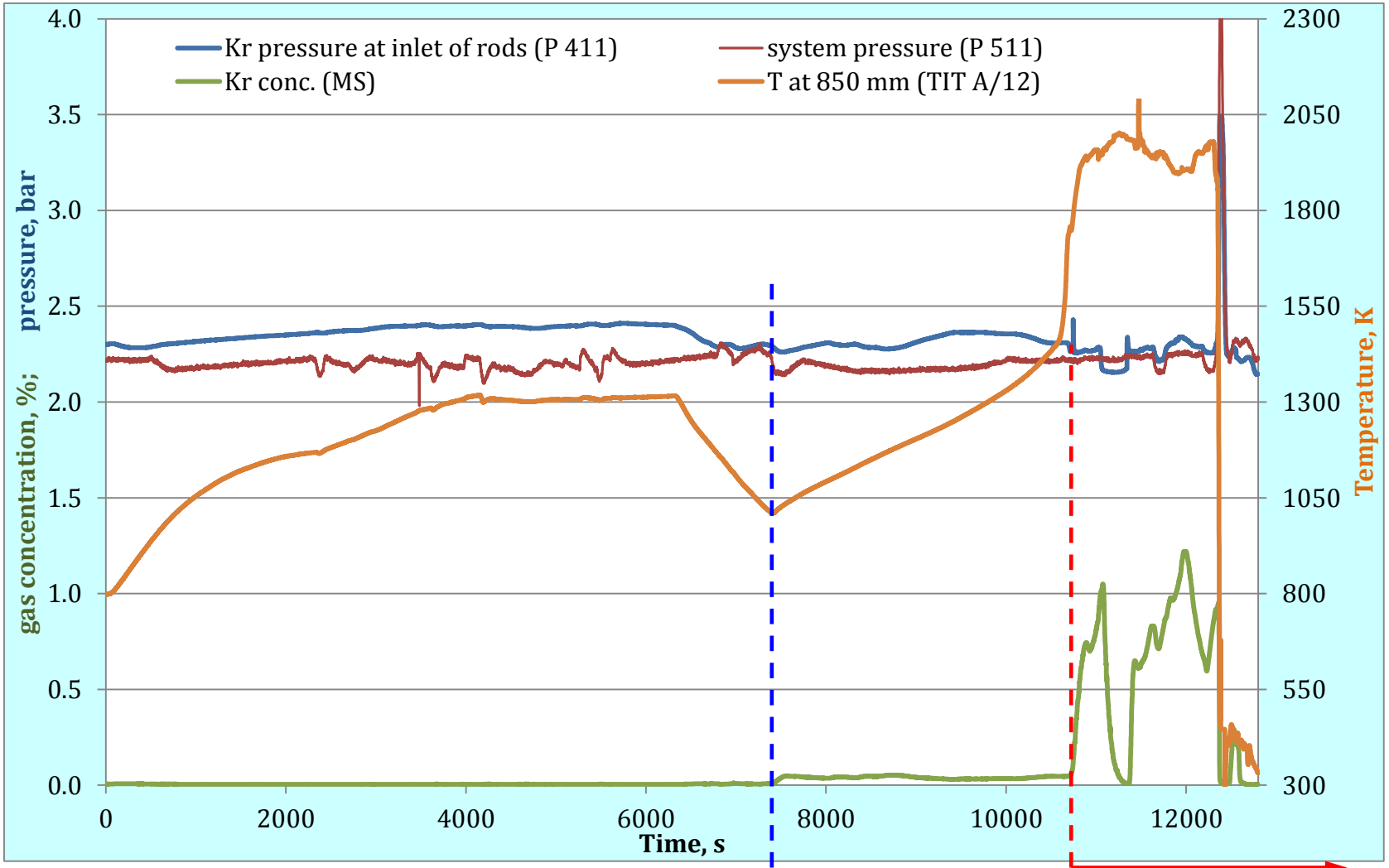
t≈10600 s: oxygen starvation, H_2 increase steam and nitrogen consumption

Readings of thermocouples at bundle elevations from -250 mm (bundle bottom) to +550 mm (strongest T escalation)

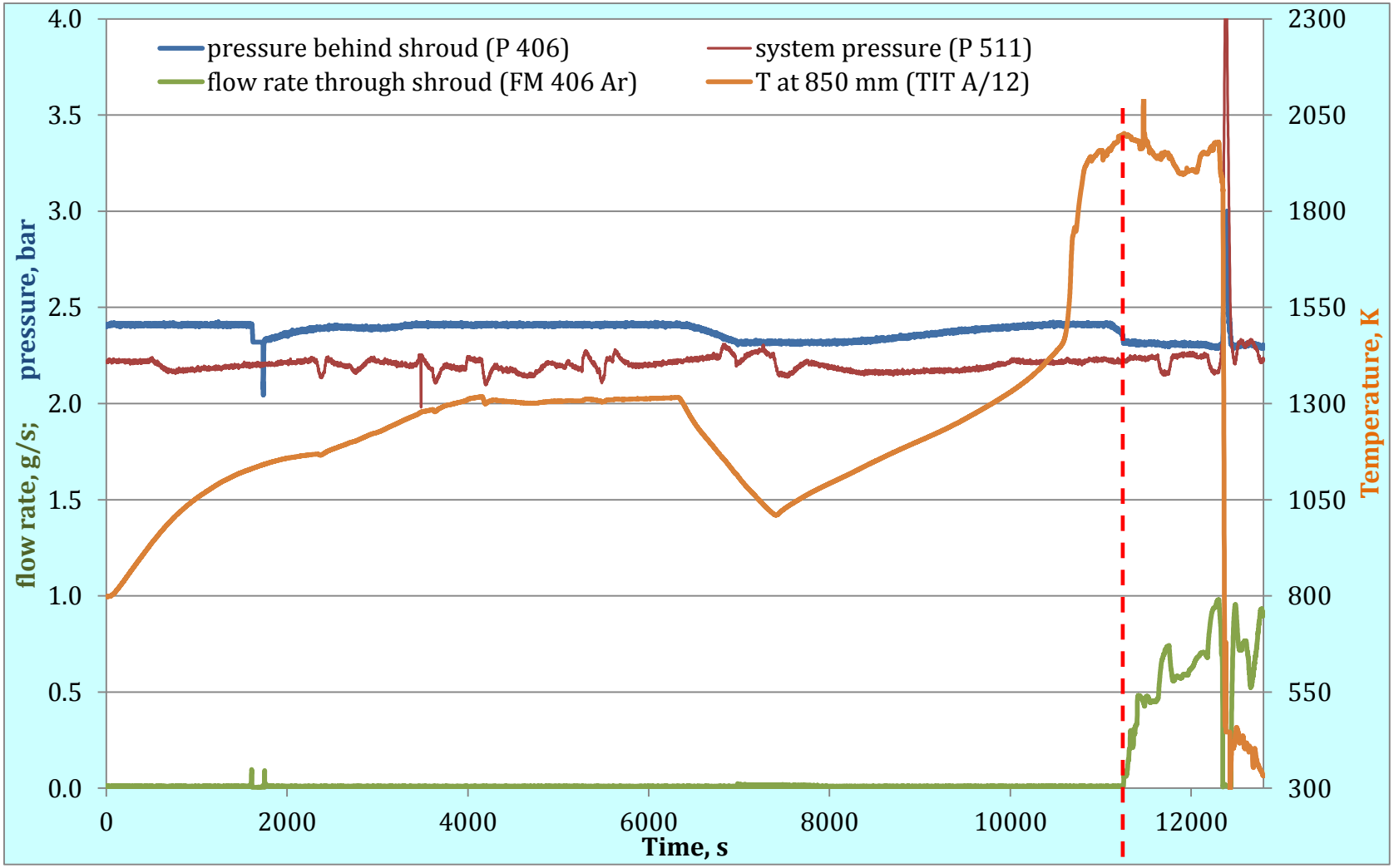


t=10680 s: absorber melt relocation from ≈ +550 mm to lower elevations

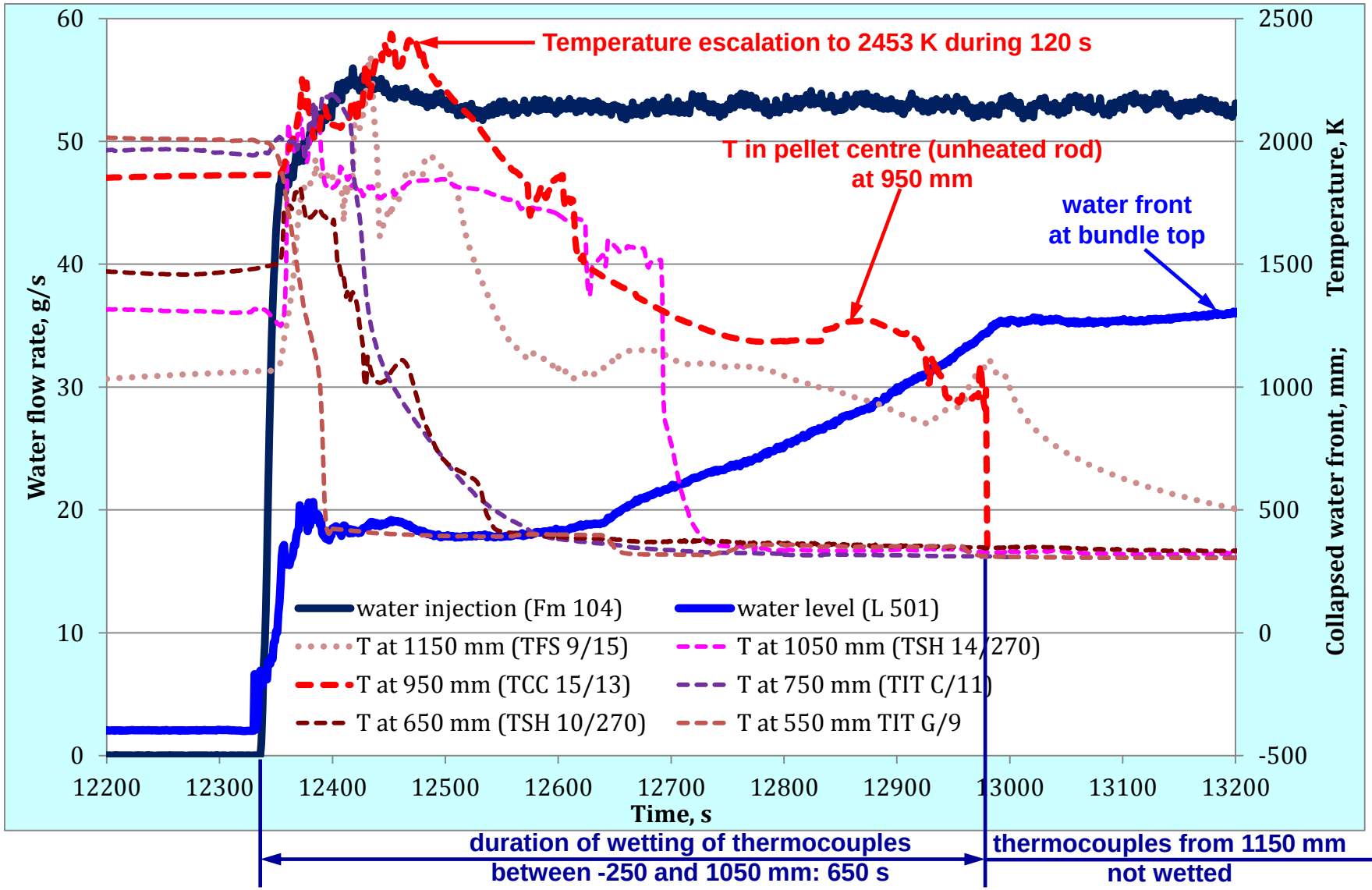
Failure of claddings (Kr release)



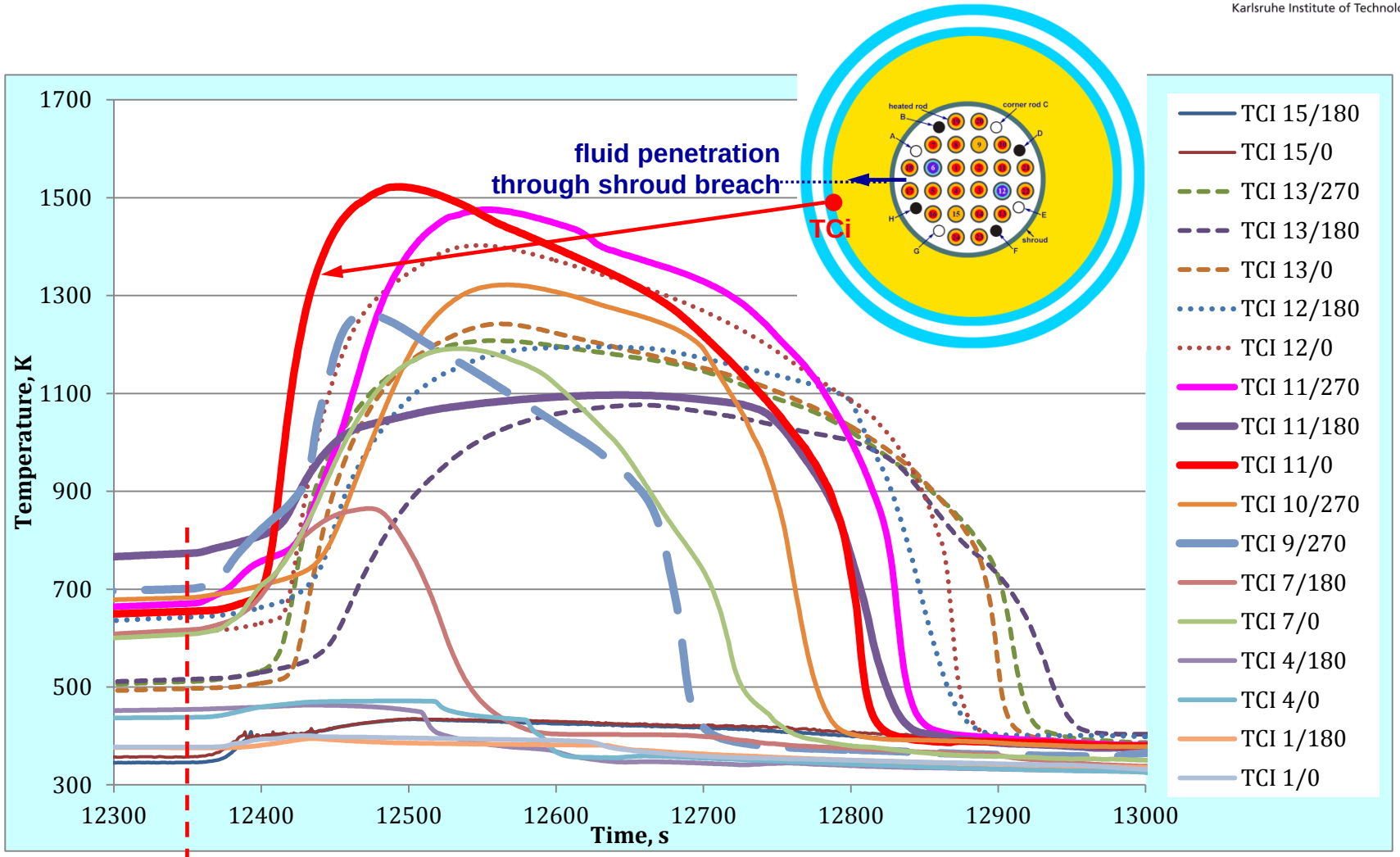
Failure of shroud (additional Ar flow)



Water and temperatures during the quench stage

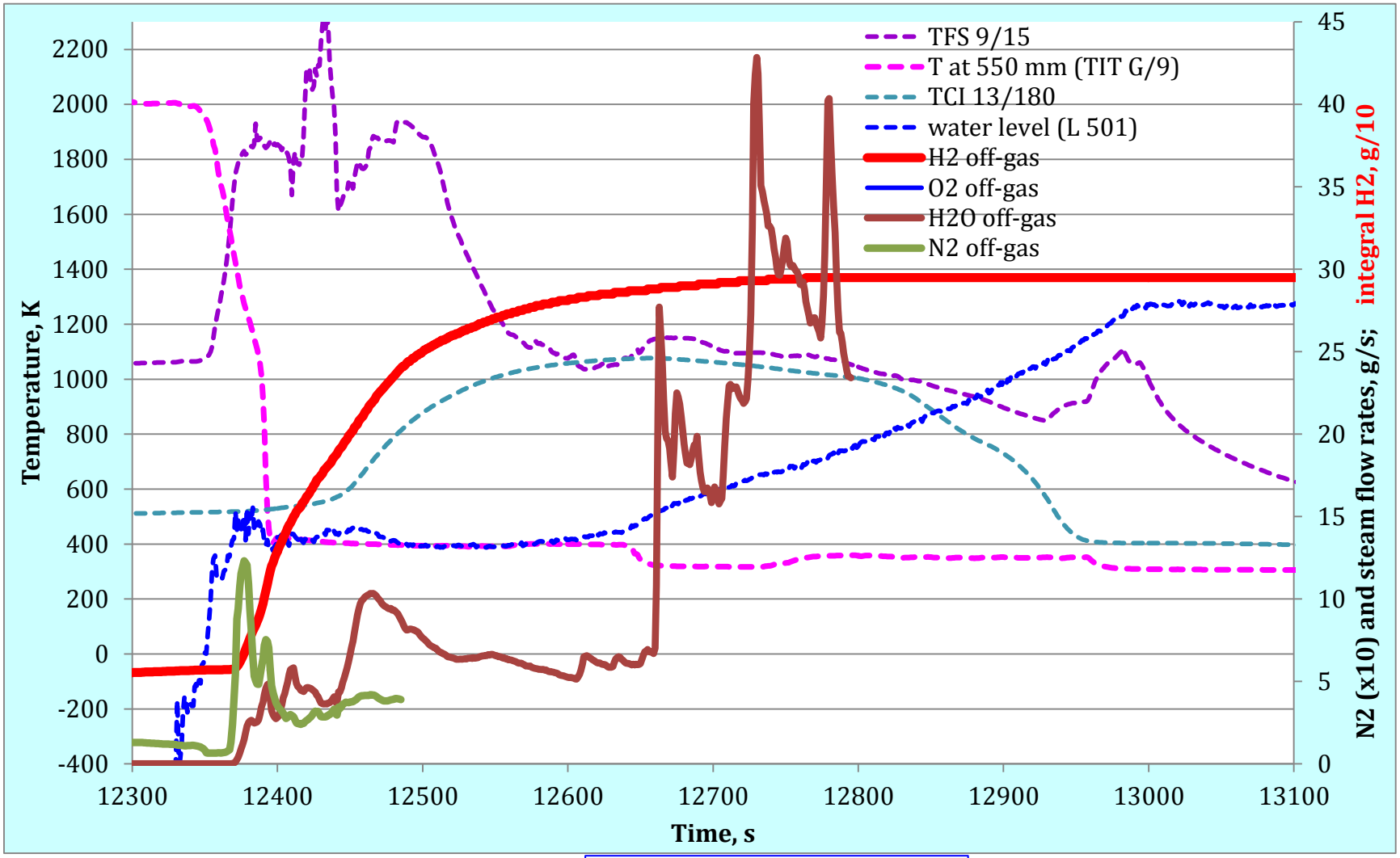


Readings of thermocouples at inner surface of cooling jacket after quench initiation (water injection from bottom)



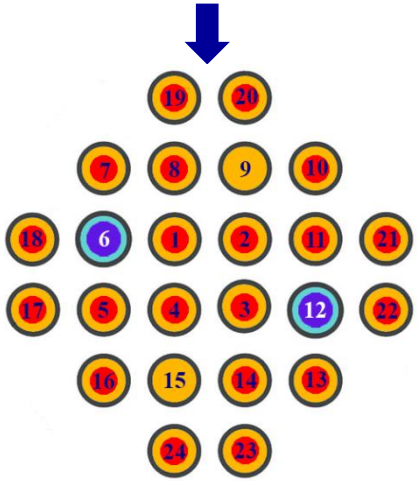
t=12330 s: quench initiation

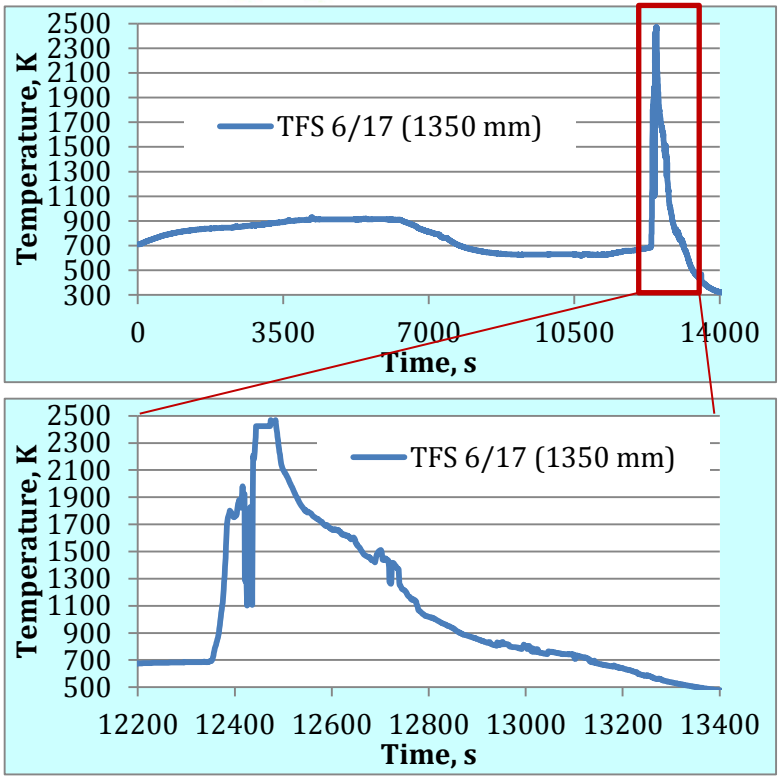
Gas release during the quench stage

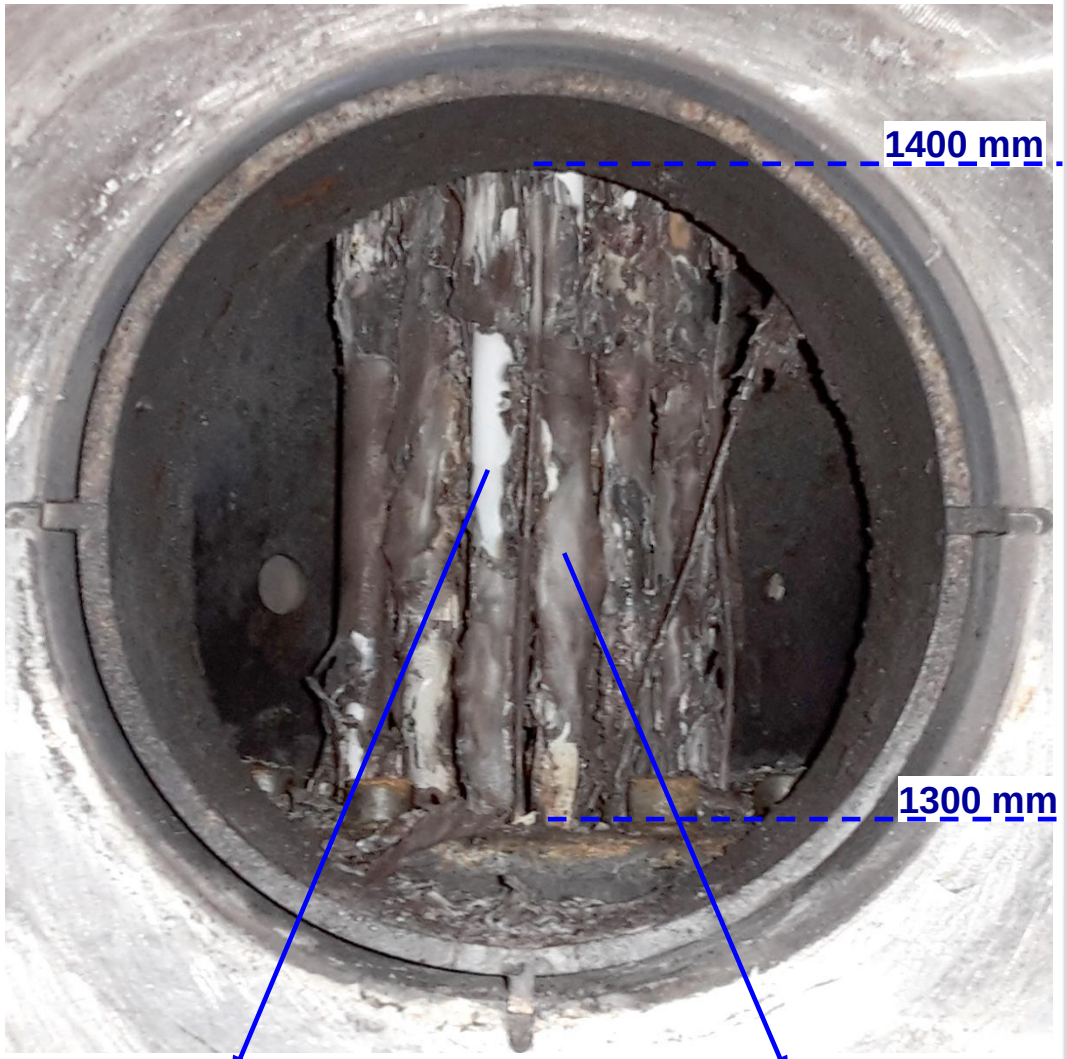


released hydrogen: 238 g
released nitrogen: >54 g

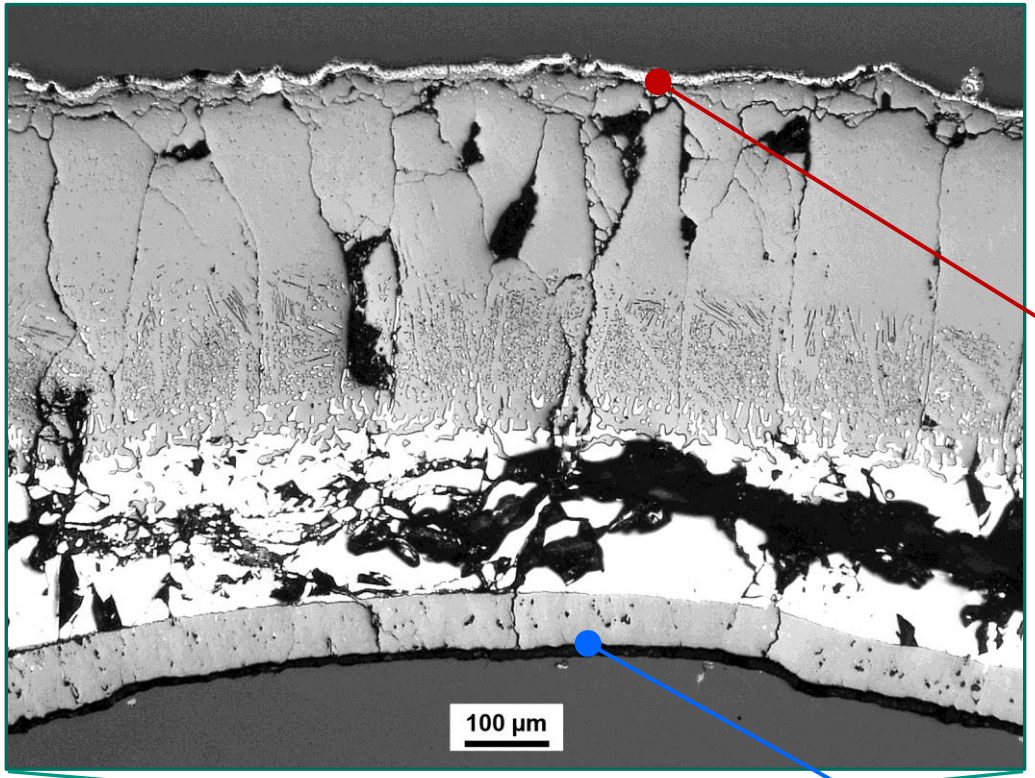
Overview of the bundle at bundle top

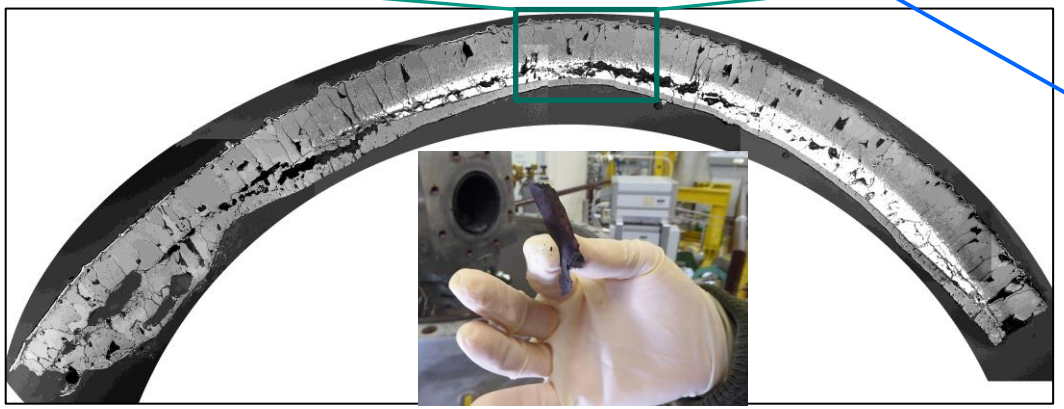


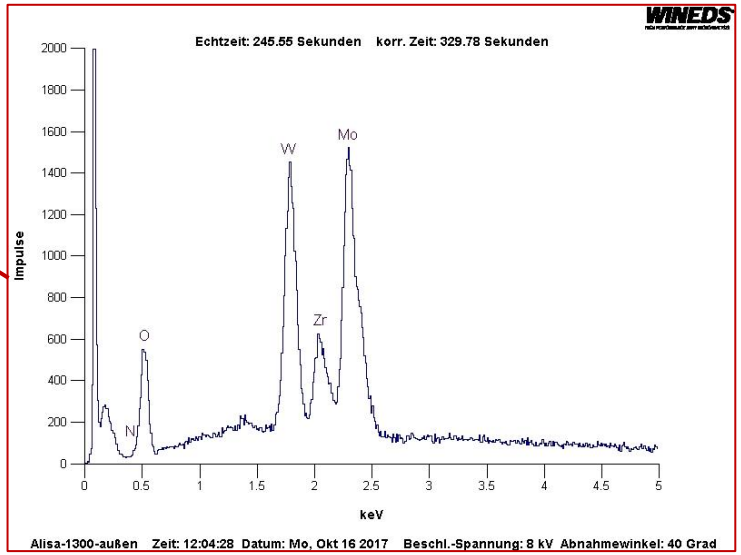


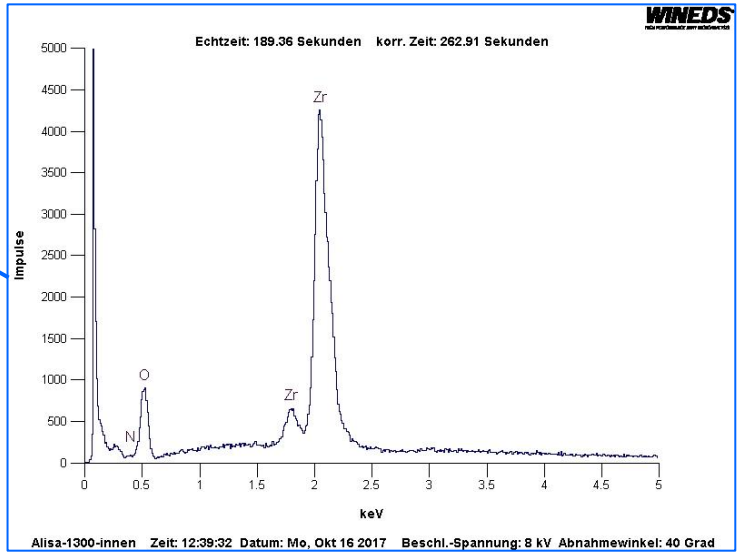


Analysis of cladding segment spalled at 1350 mm









Summary

- The QUENCH-18 test with bundle containing 20 heated and 2 unheated rods with M5® cladding as well as 2 Ag/In/Cd absorber rods was performed at KIT on September 27, 2017 in the framework of the ALISA project.
- Three typical features of QUENCH-18: moderate pre-oxidation to ≈130 µm of oxide layer (similar to QUENCH-16), a long period of oxygen starvation during the air+steam ingress phase (1770 s instead 800 s for the QUENCH-16 test performed without steam injection during air ingress), and reflood initiation at the melting point of the cladding (≈ 2000 K instead of 1700 K for QUENCH-16).
- The claddings of unheated rods burst at 1045 K at heat-up rate 0.3 K/s.
- Temperature escalation during the air ingress between elevations 150 and 850 mm was significantly stronger than for QUENCH-16 due to additional exothermal cladding oxidation in steam (corresponding additional chemical energy of ≈4 kW was even slightly higher than electrical power).
- Releases of aerosols and helium were registered on the beginning of temperature escalation (failure of absorber rods). Simultaneously, the readings of cladding surface thermocouples below elevation of 550 mm indicated the relocation of absorber melt.

Summary (cont.)

- During starvation period about *100 and 450 g oxygen and steam* correspondingly were consumed. During the steam consumption about *45 g hydrogen* were released. In the same period the partial consume of *nitrogen (about 120 g)* was registered.
- The bundle temperatures between elevations 550 and 850 mm stabilised at melting of Zr alloys.
- Initiation of reflood with 50 g/s water caused strong temperature escalation to about 2450 K at elevations between 750 and 1150 mm. As result, about *238 g hydrogen* were released (128 g for QUENCH-16). During re-oxidation of zirconium nitrides more than *54 g nitrogen* were released.
- First observations of bundle at elevations between 1300 and 1400 mm showed spalling of cladding segments from rods. Metallographic investigations of spalled parts showed strong oxidation of cladding outer surface (about 350 µm ZrO_2) and moderate oxidation of cladding inner surface (about 110 µm ZrO_2). No nitride and melt formation were indicated at these upper elevations.
- The bundle will be prepared for further investigations.

Acknowledgment

The QUENCH-18 experiment was supported and partly sponsored by the ALISA project. The bundle materials were provided by AREVA.

The authors would like to thank all participants of the QUESA project for the pre-test calculations.

Thank you for your attention

http://www.iam.kit.edu/awp/666.php
http://quench.forschung.kit.edu/

Aerosol measurements in the QUENCH-ALISA test

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Aerosol measurements were performed during the air ingress phase of the QUENCH-ALISA test. An aerosol sample was obtained from the QUENCH facility exhaust pipe and diluted using an ejector diluter and measured using several measurement devices. Electrical Low Pressure Impactor (ELPI) was used to monitor the particle size distribution online. One back ground and three filter samples were collected during the measurements. In addition, two Berner Low Pressure Impactors (BLPIs) were used for obtaining particle mass size distributions during the test.

The ELPI measurement indicated, that the particle release from the SIC control rods commenced approximately at the experiment time 10500 s. It coincided with a temperature increase measured in the QUENCH bundle, as well as a helium release detected by the mass spectrometer observing the exhaust gas from the facility. The aerosol release was observed for approximately 30 minutes and the measurement was stopped just before the quenching of the bundle took place. Figure 1 shows the mass concentration obtained from the ELPI as well as the mass concentrations calculated from the filter and BLPI samples. The number concentration determined by ELPI has been converted to mass concentration using the assumption of spherical particles with the unit density.

The preliminary Scanning Electron Microscopy and Energy Dispersive X-ray (SEM/EDX) analysis of the filters indicated that during the first filter sampling, mostly cadmium was released from the facility. Filters 2 and 3 contained also silver and from these results, a first approximate for the materials release from the control rods, shown in Table 1 could be obtained. It should be noted that a significant fraction of the aerosols are expected to have been deposited in the facility upstream of the aerosol measurement position in the off-gas pipe. Therefore, the total release fractions for Ag, In and Cd from the bundle should be higher than those detected by aerosol measurements.

Future work contains SEM/EDX analysis of the BLPI substrates as well as the ICP Mass Spectrometer study of the samples to further improve the elemental analysis.

Table 1. First estimate of the materials release from the SIC rods, obtained from the aerosol samples

	Released [g]	**Fraction from the total mass in the SIC rods**
Cadmium	7.5	15 %
Indium	0.7	0.5 %
Silver	6.9	1.0 %
Total	**15.1**	**1.5 %**

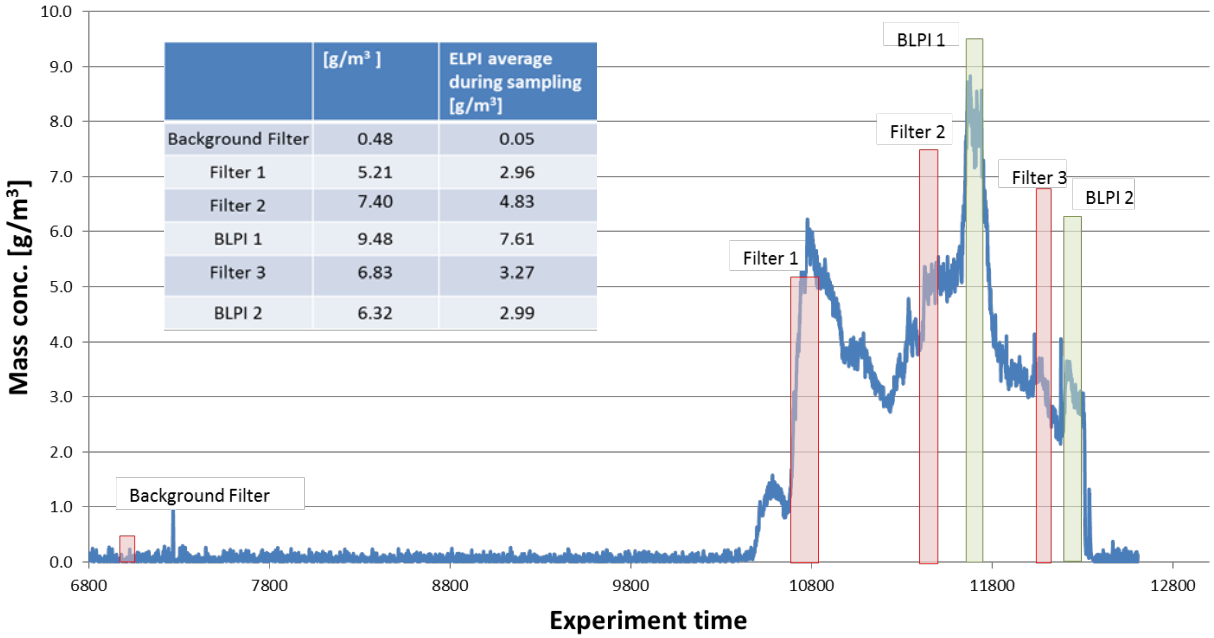


Figure 1. Mass concentration during the aerosol release at the QUENCH-ALISA test.



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PSI



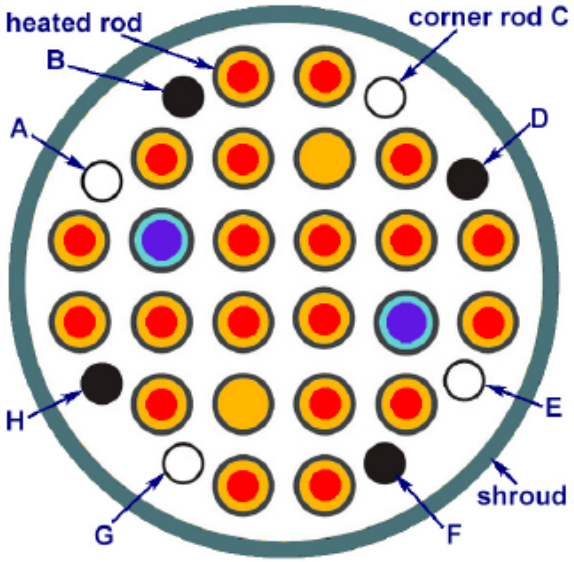
Jarmo Kalilainen, Terttaliisa Lind, Ulrike Stegmaier :: Paul Scherrer Institut

Aerosol measurements in the QUENCH-ALISA test

23rd International QUENCH Workshop, 17.10.2017, Karlsruhe, Germany

QUENCH-ALISA test

Component	Number	Material properties	Outer diameter (mm)	Inner diameter (mm)
Rod Cladding	22	Zirlo	9.50	8.36
SIC Absorber Rod	2	Ag 80%, In 15%, Cd 5% density: 10.17 g/cm³	7.65	
Absorber Cladding	2	Steel AISI 316	9.68	7.72
Absorber Guide Tube	2	Zr alloy	12.45	11.25
Corner Rod	8	Zry-4	6.00	
Shroud	1	Zry	89.00	83.00

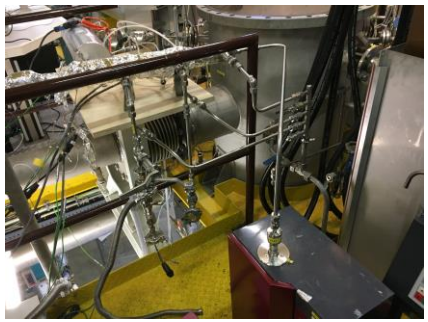


Presentation : Thorsten Hollands, 4th Nugenia-Sarnet rev. meeting 2017

- Air ingress:
  - Steam: 0.3 g/s
  - Argon: 1 g/s
  - Air: 0.2 g/s

Measurement set-up



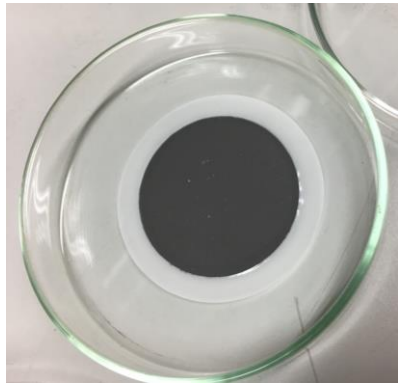


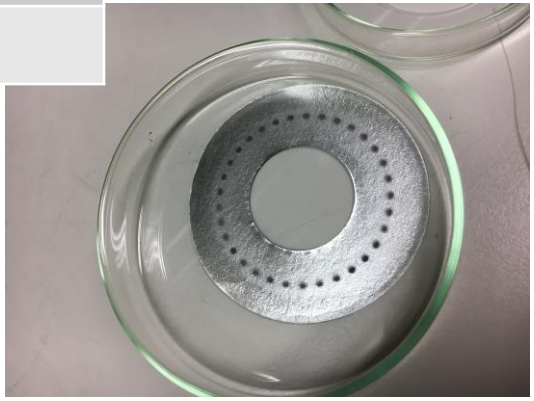
Measurement Matrix

Continues particle size distribution measurement with ELPI



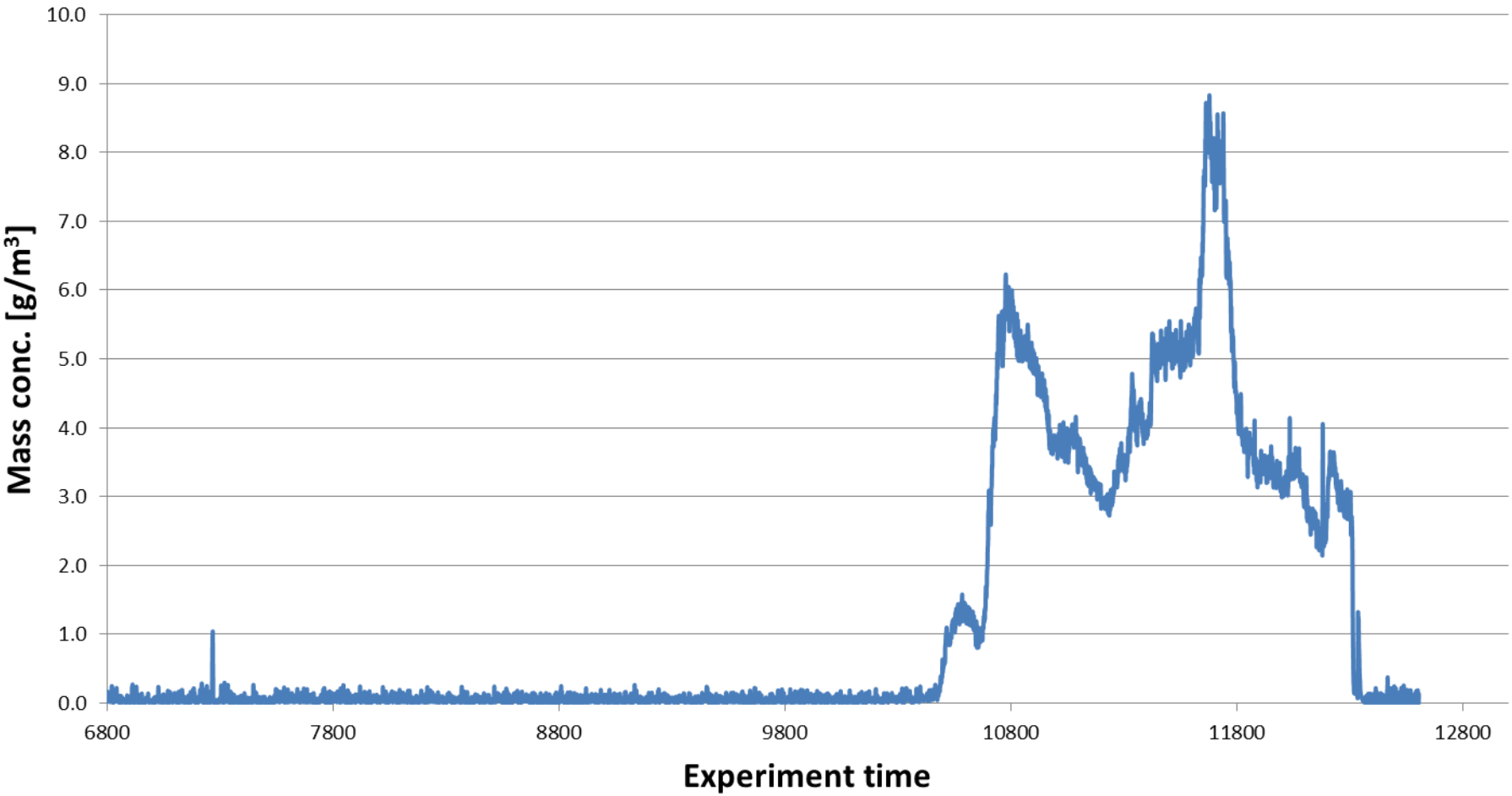
Experiment time [s]	Sample
6892 (13:49)	Background filter
10612 (14:51)	Filter 1
11392 (15:04)	Filter 2
11632 (15:07)	BLPI 1
11992 (15:14)	Filter 3
12172 (15:17)	BLPI 2





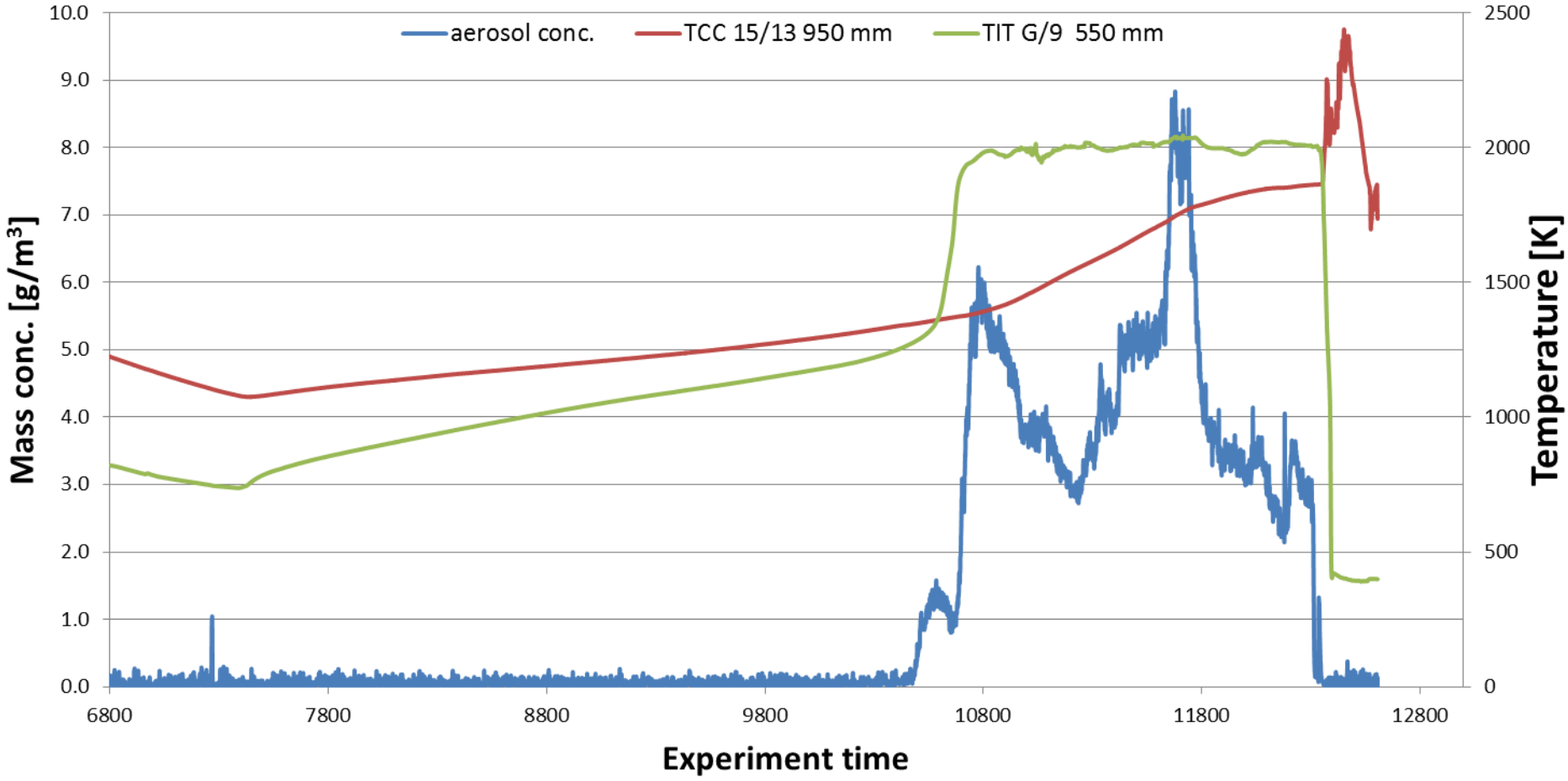
Results: Mass concentration from ELPI

- Aerosol released during the Air Ingress phase
  - Duration approx. 32 min.



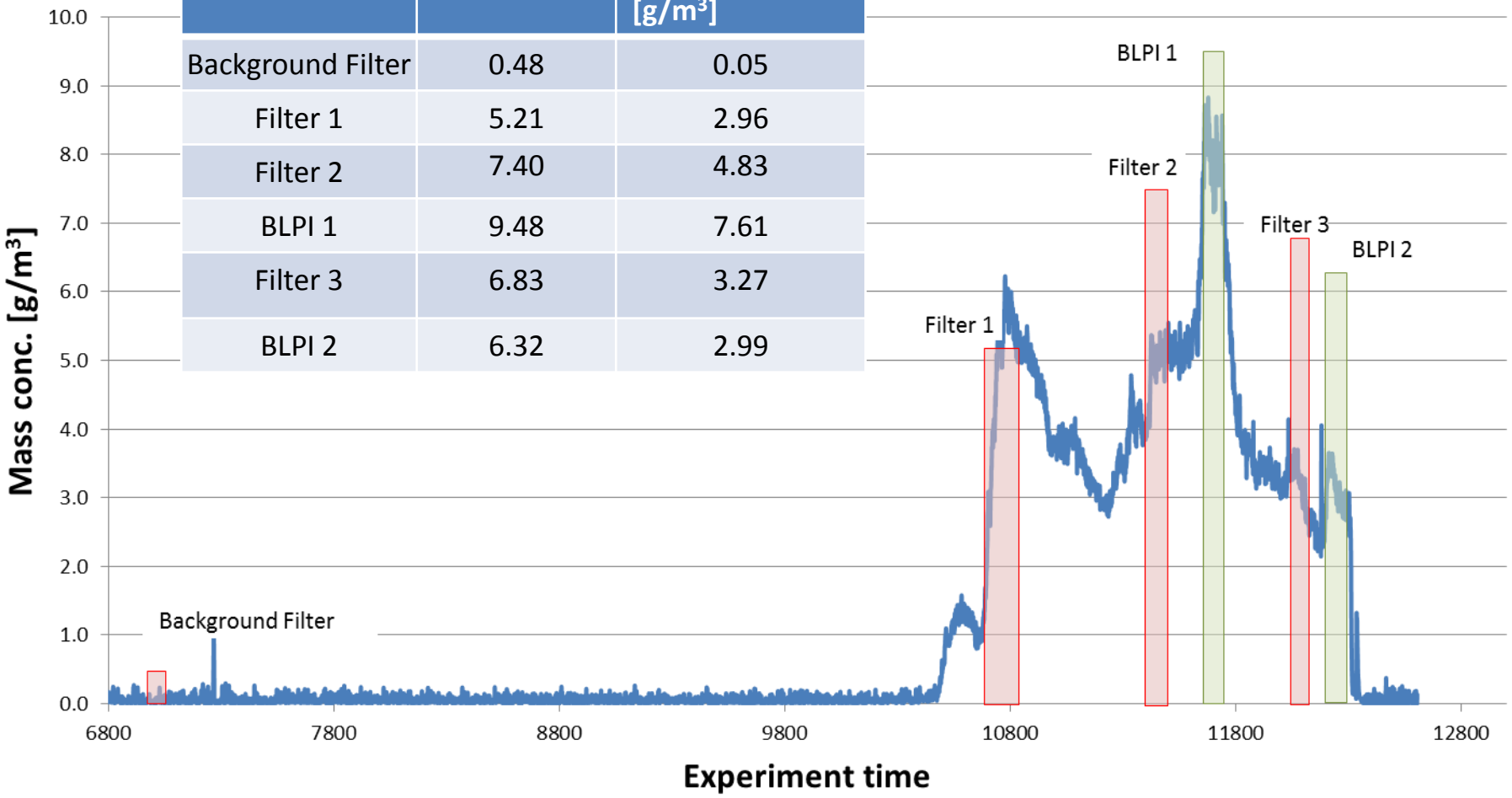
Results: Mass concentration from ELPI

- Evolution of aerosol mass compared to the QUENCH facility temperatures



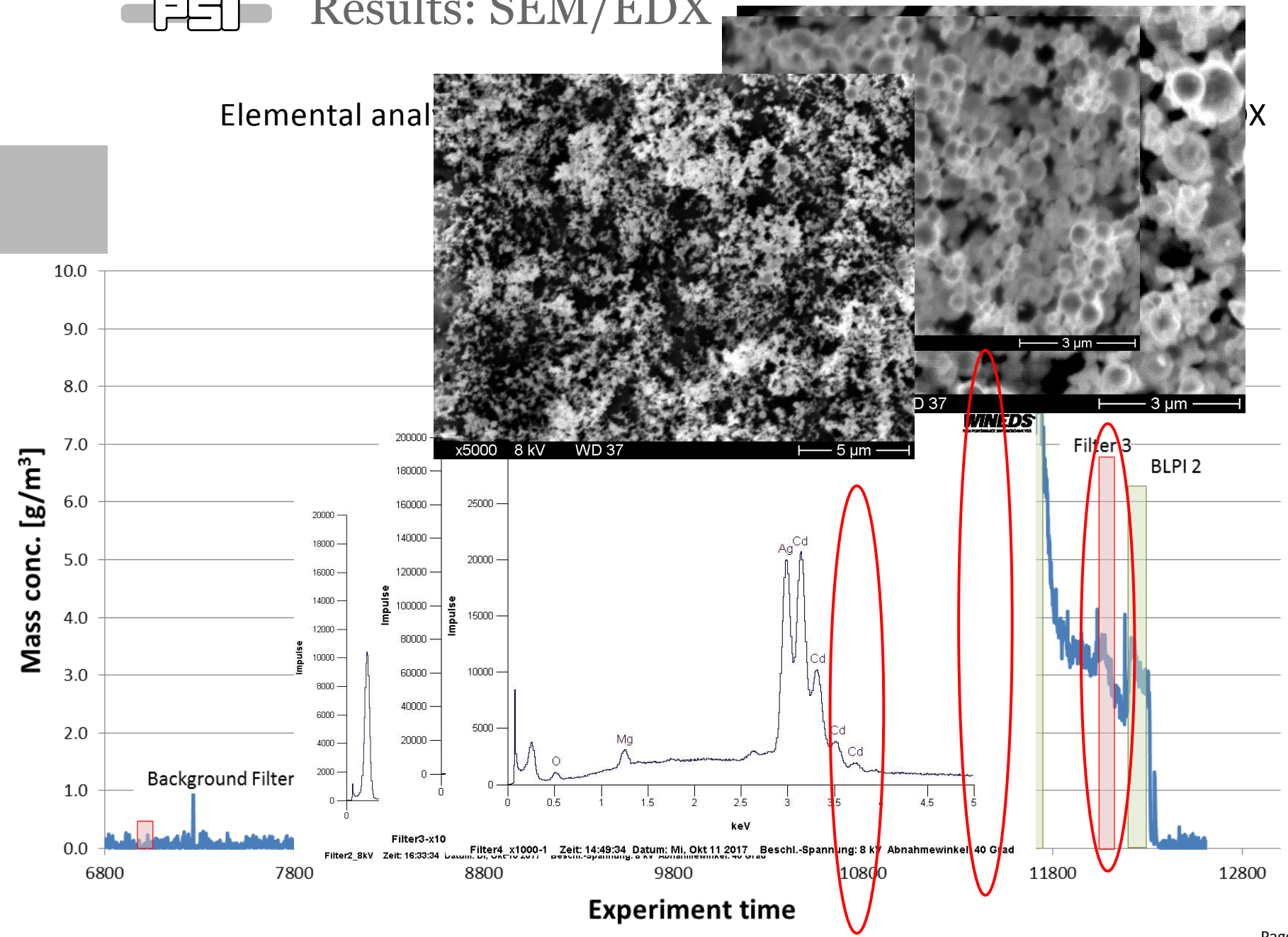
Results: Mass concentrations from samplings

	[g/m³]	ELPI average during sampling [g/m³]
Background Filter	0.48	0.05
Filter 1	5.21	2.96
Filter 2	7.40	4.83
BLPI 1	9.48	7.61
Filter 3	6.83	3.27
BLPI 2	6.32	2.99



Results: SEM/EDX

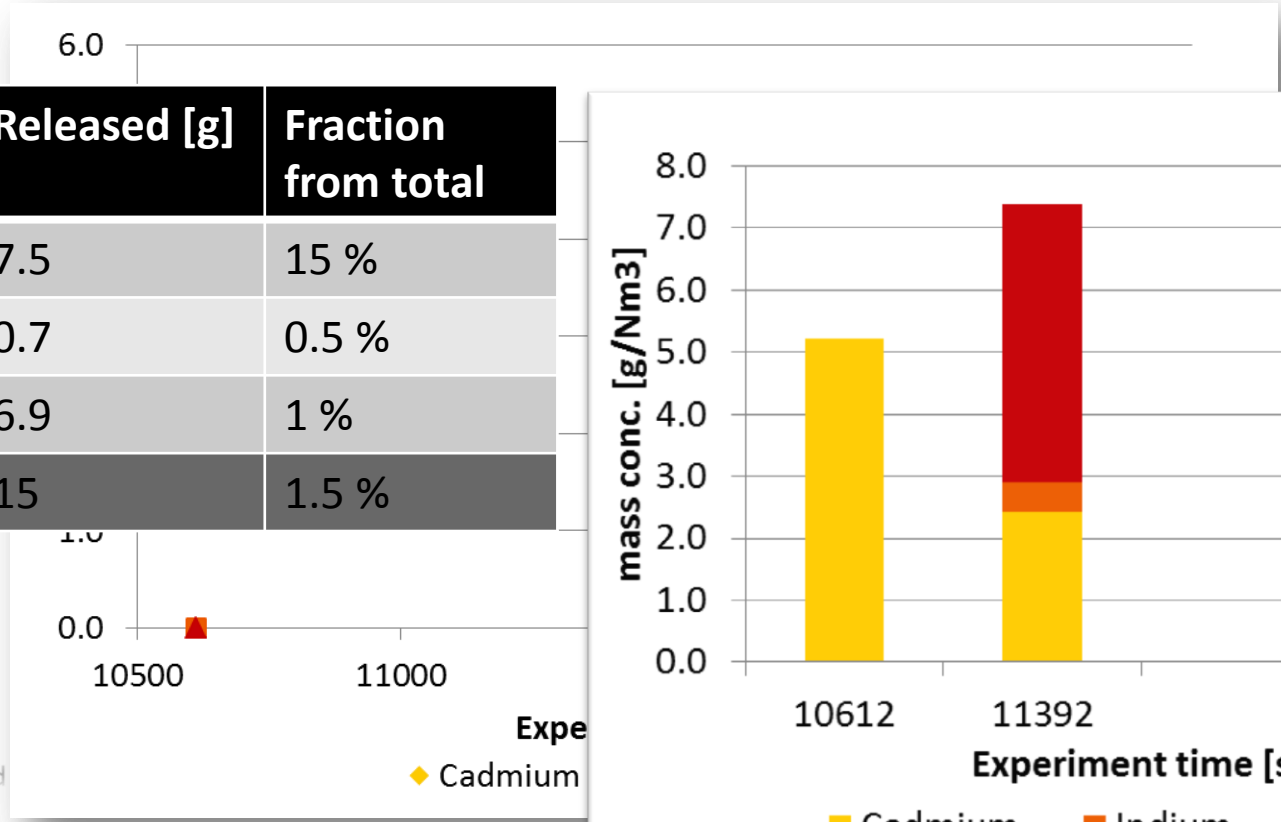
Elemental anal X



Results: Cd/In/Ag release

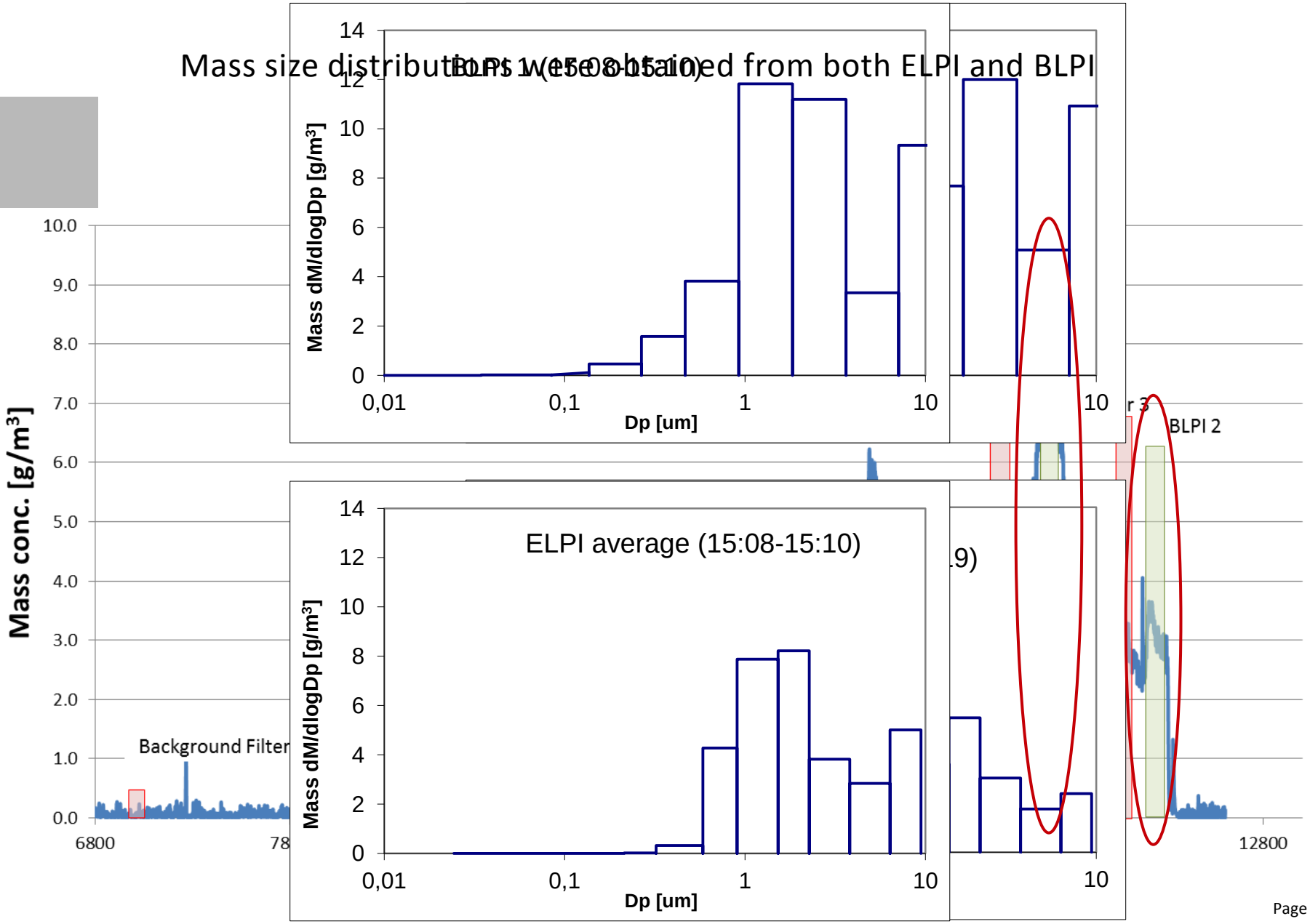
- Based on the EDX analysis, a crude estimate can be made for the SIC rod material releases

	Released [g]	Fraction from total
Cadmium	7.5	15 %
Indium	0.7	0.5 %
Silver	6.9	1 %
Total	15	1.5 %



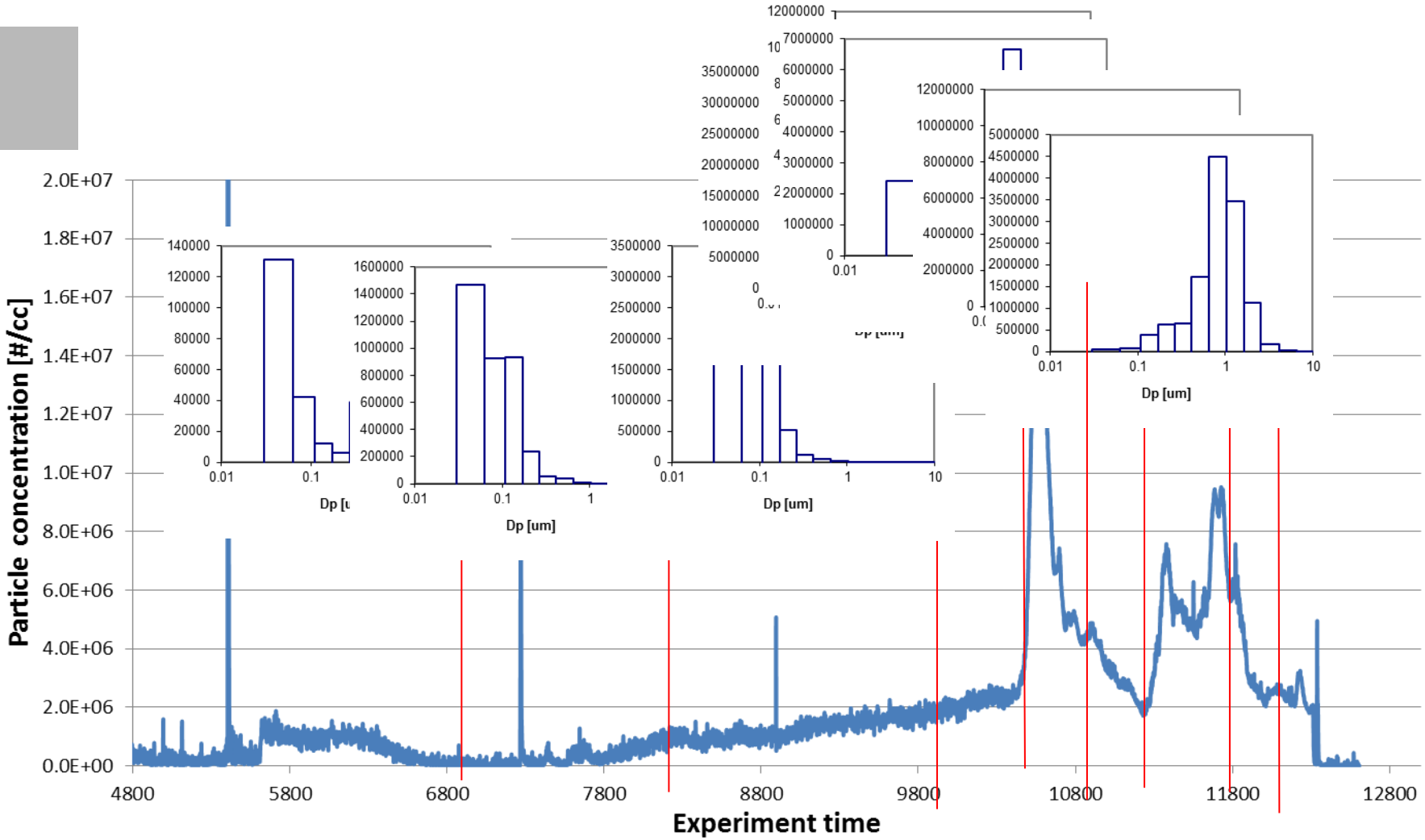
Results: Mass size distributions

Mass size distributions were obtained from both ELPI and BLPI



Results: Number size distributions from ELPI

The particle number size distribution evolved during the measurement



Summary

- Aerosol measurements were performed during the air ingress phase at the QUENCH-ALISA test
- Preliminary results available:
  - Estimates for the particle mass concentration during the air ingress were obtained from the filter samplings and BLPI/ELPI measurements
  - Good match of the mass distributions from both BLPI and ELPI
  - Elemental analysis by SEM/EDX:
    - The change in Cd, In and Ag concentrations during the air ingress can be estimated
    - First estimate was obtained for the release fraction of elements from the SIC rods
- Further analysis will include:
  - SEM/EDX of the BLPI samples
  - ICP-MS for element masses from the filter and BLPI samples
  - Possible further analysis with Raman and/or XRD (for example Cd speciation?)

Thank you for your attention



IAEA activities in the area of Nuclear Power Reactor Fuel Engineering

M.S. Veshchunov

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Abstract

The main IAEA program implementation tools in the area of fuel engineering are Coordinated Research Projects (CRP), Technical Meetings (TM), Expert Reviews, and Databases - IAEA-HOTLAB Database of PIE Facilities and NEA-IAEA International Fuel Performance Experiments (IFPE) Database. This report provides information about organization and implementation practices of these activities, and summarizes their major outputs. Since 2011 a special attention is being paid to projects related to the IAEA Nuclear Safety Action Plan initiated after Fukushima accident, that include Coordinated Research Projects “Fuel Modelling in Accident Conditions” (CRP FUMAC) and “Analysis of Options and Experimental Examination of Fuels for Water-Cooled Reactors with Increased Accident Tolerance” (CRP ACTOF).


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IAEA 60 Years
Atoms for Peace and Development

IAEA Activities in the Area of Nuclear Power Reactor Fuel Engineering

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23rd International QUENCH Workshop

Karlsruhe Institute of Technology

October 17-19, 2017

Outline

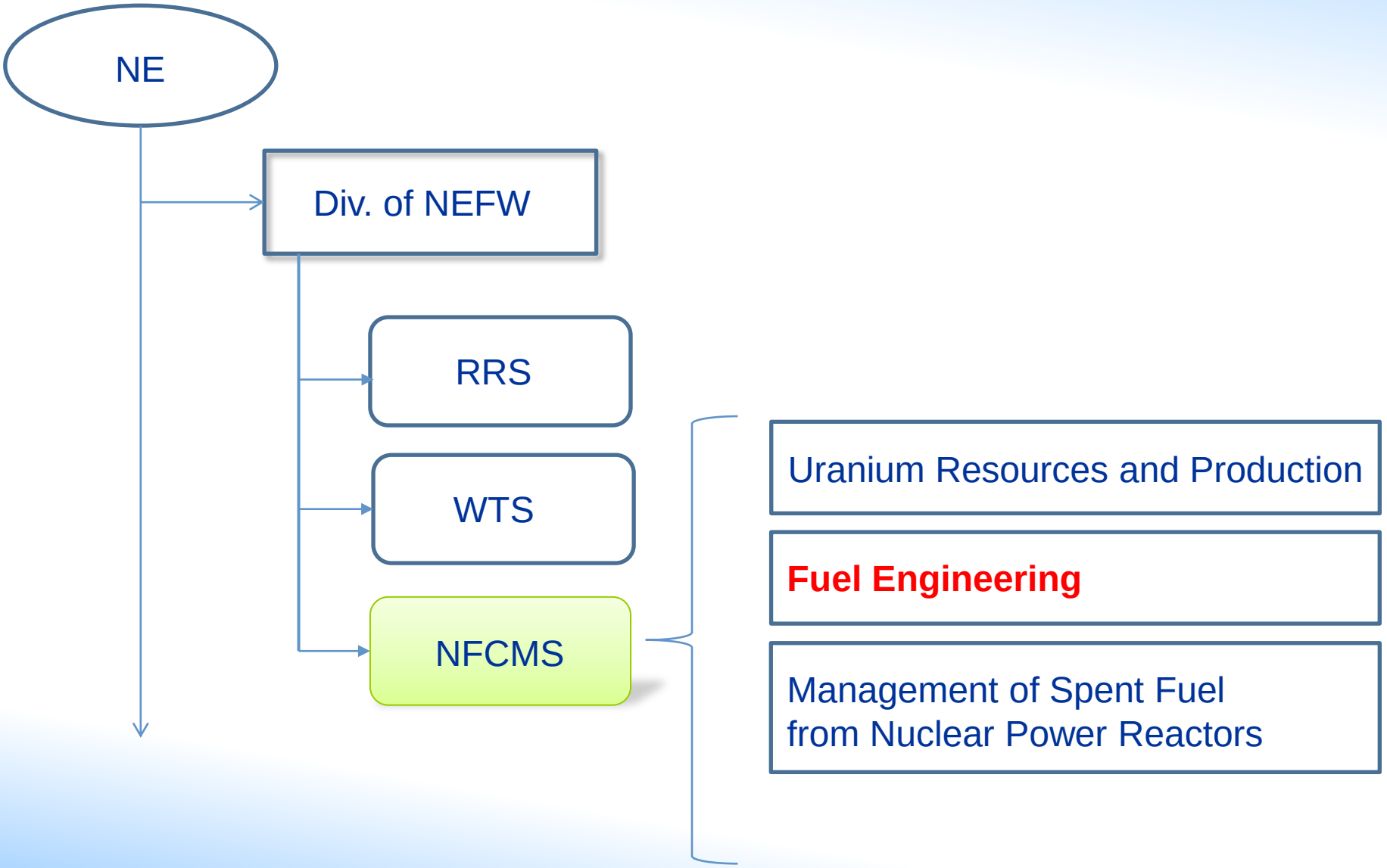
- IAEA Fuel Engineering Programme
- Coordinated Research Projects (CRP)
  - CRP on FUel Modelling in Accident Conditions (FUMAC)
  - CRP on Analysis of Options and Experimental Examination of Fuels with Increased Accident Tolerance (ACTOF)
- Consultancy and Technical Meetings
- Database systems
- Conclusion

Main Objectives of the IAEA Fuel Engineering Programme

- Major Programme (MP) 1: Nuclear Power, Fuel Cycle and Nuclear Science
  - Programme (P) 1.2: Nuclear Fuel Cycle and Materials Technologies
    - Subprogramme (SP) 1.2.2: Nuclear Power Reactor Fuel
      - ✓ Project 1.2.2.1: Nuclear Power Reactor Fuel Engineering

- To enable Member States to organize adequate R&D programmes to support effective design and manufacturing technologies and to optimize in-pile performance of current and advanced fuels and materials for reliability and efficiency

Who we are


NE
Div. of NEFW
RRS
WTS
NFCMS
Uranium Resources and Production
Fuel Engineering
Management of Spent Fuel
from Nuclear Power Reactors

Mandate

- Support MS to understand and address factors affecting the design, fabrication and in-pile behaviour of currently operating and innovative **nuclear fuels and materials** for power reactors,

- Support MS to technically implement IAEA Safety Standards when operating or upgrading existing **nuclear fuel cycle facilities**, and to understand and address factors affecting the ageing of these facilities.

Via

- Organizing TMs, Workshops, CRPs
- Technical cooperation programmes
- Maintaining database on nuclear fuels and fuel cycles

Major tools of the Fuel Engineering Programme implementation

- Consultancy Meetings (Expert Reviews)
- Technical Meetings
- Coordinated Research Projects (CRP)



Fuel engineering programme overview

Advised by TWG FPT

- The Technical Working Group on Fuel Performance and Technology (TWGFPT) is a group of recognized experts from MSs to provide advice and support programme implementation, reflecting a global network of excellence and expertise in the area of nuclear power reactor fuel engineering (25 Members)

CRPs (On-going)

- Fuel Modelling in Accident Conditions, FUMAC (2014-2018)
- Analysis of options and experimental examination of fuels for water cooled reactors with increased accident tolerance, ACTOF (2014-2018)
- Reliability of high power, extended burnup and advanced PHWR fuels (2015-2018)
- CRP on Accelerator Simulation and Modelling of Radiation Effects, SMoRE-2 (2016-2020)
- Fuel materials for fast reactors (2018-2021, planned)

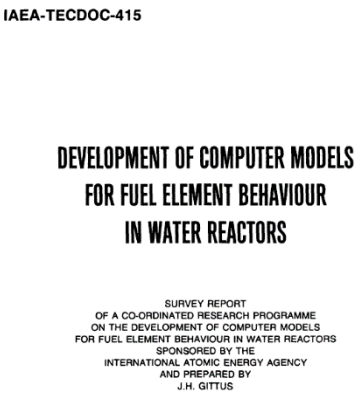
Coordinated Research Projects (CRPs)

- CRPs are a mechanism for organizing international research work within the IAEA programme of work
- A CRP theme is scoped in consultation with experts, and authorized by the IAEA for proposals of work from Member States
- Typically ~4 years, organized around 3 research coordination meetings (RCMs)
- A very good opportunity to work with many groups around the world
- CRP direction is largely defined in the first RCM by the CRP participants
- Finalized as an IAEA TECDOC
- Results are available, free of charge, to scientists, engineers and other users from all Member States

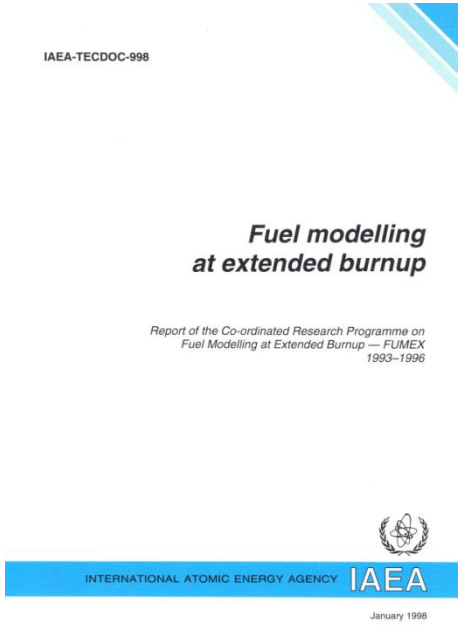
CRP Series on Fuel Modelling (1982-2012)

Joint NEA-IAEA International Fuel Performance Experiments (IFPE) Database

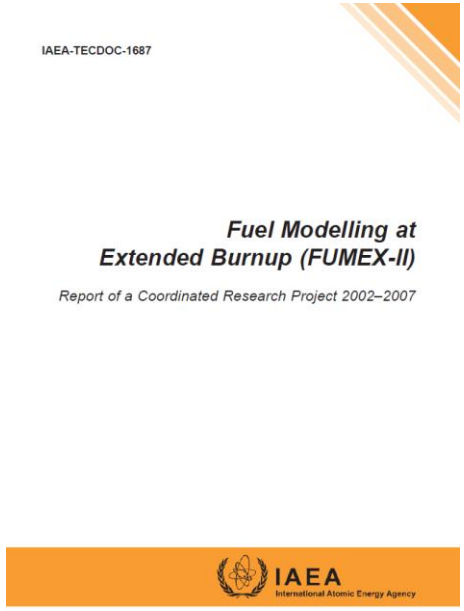
Blind exercises



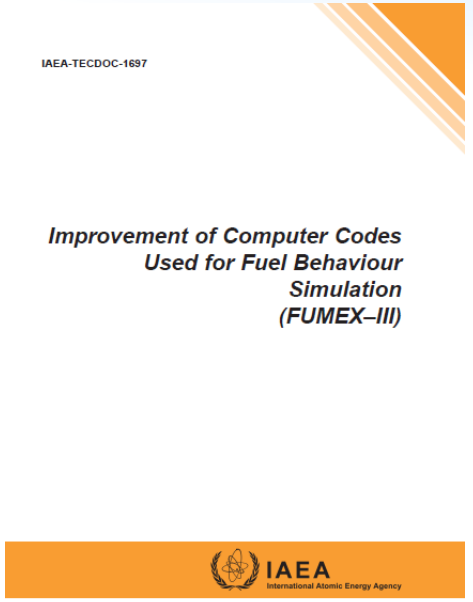
CRP D-COM
1982-1984
published in 1987



CRP FUMEX
1993-1996
published in 1998



CRP FUMEX-2
2002-2007
published in 2012



CRP FUMEX-3
2008-2012
published in 2013

AP support: New CRP FUMAC (Fuel Modelling in Accident Conditions), 2014-2018

CRP on FUel Modelling in Accident Conditions (FUMAC)

- IAEA CRP **T12028** (2014-2017)
- Regular Budget (Project 1.2.2.003: AP Support Related to Nuclear Power Reactor Fuel)
- Continuation of the previous FUMEX projects, but focus on the accident conditions, following the IAEA Nuclear Safety Action Plan at enhancing safety of nuclear reactor operations after the Fukushima accident

❑ Objectives

- Better understanding of fuel behaviour in accident situations through identification of best practices in the application of relevant physical models and computer codes, used in different Member States, and enhancement of their predictive capacities
- Selecting well checked results of accident simulation experiments, integrating into the joint NEA-IAEA International Fuel Performance Experiments (IFPE) Database and using codes for verification and benchmarking

CRP FUMAC

- **Participants (**24 organizations from 18 countries)

  CNEA (Argentina), Suez-Tractabel (Belgium), IPEN-CNEN (Brazil), INRNE (Bulgaria), CIAE (China), CNPTRI (China), EC IRC/ITU, VTT (Finland), CEA (France), KIT (Germany), MTA EK (Hungary), Poli di Milano (Italy), NSR (Japan), KAERI (Korea), IFE (Norway), IBRAE (Russian Federation), VNIINM (Russian Federation), CIEMAT (Spain), Quantumtech (Sweden), Energorisk (Ukraine), SSTCNRS (Ukraine), Batelle INL (USA), Westinghouse (USA), NRC (USA)

- **Observers**

  OECD-NEA, GRS (Germany)

- **Status Update**
  - TM “Fuel behaviour and modelling under severe transient and LOCA conditions”, Mito, Japan, 2011
  - TM “Modelling of water-cooled fuel including design basis and severe accidents”, Chengdu, China, 2013
  - Consultants Meeting on FUMAC outline and organization, Vienna, March 2014
  - RCM-1, Karlsruhe, 11-14 November 2014
  - RCM-2, Vienna, May 30 - June 2, 2016

CRP FUMAC

❑ Scope

- Focus on LOCA (DBA) fuel behaviour, in line with the early stage of the scenario of the Fukushima accident (BDBA)
- Preparation of experimental data sets for code benchmarking:
  - Separate-effect tests with fuel cladding segments (MTA EK, Hungary)
  - Out-of-pile single-rod tests (NRC Studsvik LOCA tests 192 and 198)
  - Out-of-pile SA and LOCA bundle tests (QUENCH and CORA facilities, KIT, Germany)
  - In-pile LOCA tests (IFA 650.9-11, HALDEN)
- Benchmark exercises against HALDEN tests by participants using various fuel performance codes: ALCYONE (France), BISON, FRAPCON (USA), DIONISIO (Argentina), TRANSURANUS (JRC-ITU), START-3, SFPR (Russia), etc.
- Thermal hydraulic boundary conditions for benchmark exercises

FUMAC Benchmark Exercises

IFA 650.9	IFA 650.10	IFA 650.11	Studsvik 192 198	QUENCH LOCA	CORA 15	MTA-EK	Provide TH information for benchmark		Uncertainties analysis
With dispersal	PWR	VVER	Out-of-pile Single fuel rod	Out-of-pile Bundle	Out-of-pile Bundle	Fuel rod segments	QUENCH	IFA650	DAKOTA, SUNSET, etc.

- Each participant was asked to indicate which of the various experiments they would be able to evaluate
- The thermal hydraulic boundary conditions are provided for IFA650 and QUENCH tests by SOCRAT
- Uncertainty analysis will be performed in the second phase
- Collection and preparation of all data (FUMAC Project website):

  https://nucleus.iaea.org/sites/nefw-projects/fumaccrp/_layouts/15/viewlsts.aspx?BaseType=0

CRP on Analysis of Options and Experimental Examination of Fuels with Increased Accident Tolerance (ACTOF)

- IAEA CRP T12030 (2014-2018)
- Regular Budget (Project 1.2.2.003: AP Support Related to Nuclear Power Reactor Fuel)
- The 1st RCM was held from 2 to 4 November 2015 in Vienna
- The 2nd RCM was held from 20 to 22 June 2017 in Vienna
- Collection and preparation of all data (ACTOF Project website): https://nucleus.iaea.org/sites/nefw-projects/crpactof/SitePages/Home.aspx

❑ MSs' participation (15 organizations from 12 MSs)

Argentina (CNEA), Brazil (USP), China (CNPRI, CIAE, NPIC), Finland (VTT), Germany (KIT), India (BARC), Italy (NIE), Korea (KAERI), Poland (INCT), Russian Federation (VNIINM, IBRAE), United States (Westinghouse, Batelle), Czech Republic (TU)

❑ Observers

OECD-NEA, JAEA (Japan), EPRI (USA), US NRC

CRP ACTOF

❑ Objective

- To support options for the development of nuclear fuel with an improved tolerance of severe accident conditions
- To support modelling of new fuel designs with advanced cladding or fuel
- To acquire data through experiments on new fuel types and cladding materials to support their use for fuel with improved accident tolerance

❑ Background

- Fukushima accident: zirconium cladding worsened the problem
- New and improved fuel designs are being considered by many Member States
- TWGFPT recommendation arising from post-Fukushima review
- IAEA expertise in supporting fuel modelling – FUMAC CRP considers standard fuel modelling under accident conditions
- Technical Meeting on Accident Tolerant Fuel held in US, 2014

Advanced Core Materials

IAEA 60 Years Atoms for Peace and Development

Cladding/Core Materials

- Properties of material candidates
  - Coated and improved Zr-based alloys
  - SiC and SiC/SiC composites
  - Advanced steels (ODS, FeCrAl, …)
  - Refractory metals
- Evaluation under normal operations and SA conditions

Fuel Concepts

- Properties of material candidates
  - Modified UO_2 (BeO, Cr_2O_3,…)
  - High density fuel (UN, UC, (U,Pu)N, U_3Si_2,…)
  - Composite fuel (UN-U_3Si_2,…)
  - Coated particle fuel
- Evaluation under normal operations and SA conditions
- Pellet-Cladding Interaction (PCI)

ACTOF Round Robin and Benchmark Exercises (1/2)

Round Robin tests

a. Coordinate round robin test for coated Zr alloy cladding (Westinghouse):
   i. Finalize testing conditions and sample matrix
   ii. Finalize schedule
   iii. Financial support may be available from IAEA
b. Provide Zr alloy substrate for coating (Westinghouse)
c. Provide coating samples: 3 samples per type per test condition for each test lab (INCT, KAERI, KIT, TU, VTT)
d. Perform autoclave test (INCT, TU, VTT)
e. Perform post autoclave test analysis and report, provide uncertainties in measurement
f. Perform high temperature steam oxidation test (KIT, TU, VTT) 1300°C and 1400°C max. 30 minutes
g. Perform post high temperature steam test analysis and report

ACTOF Round Robin and Benchmark Exercises (2/2)

IAEA 60 Years Atoms for Peace and Development

Benchmark calculations

- Provide material property data for:
  - FeCrAl and U_3Si_2 (INL)
  - AISI 348 (USP),
  - SiC (CGNPC, CIAE)
  - coated cladding (KAERI)
- Benchmark calculation for FeCrAl
  - Provide modeling input, normal operation case specification (UO_2-FeCrAl) and LOCA case specification, output format for FeCrAl (INL)
  - Perform benchmark calculation using different codes; include BISON (INL), FRAPCON (USP), BACO (CNEA), TRANSURANUS (NINE, Westinghouse)
  - Prepare report for benchmark calculation
- Severe accident experimental and modeling
  - Pre-test calculations for QUENCH-19 (KIT, IBRAE, EPRI (?))
  - Provide results of severe accident testing, including separate single rod test and Quench-19 FeCrAl test (KIT)
  - Calculation for high temperature QUENCH-19 FeCrAl test (IBRAE, EPRI)

New CRP Proposal on Fuel Materials for Fast Reactors

Objective

Bringing together specialists from the Member States with active fast reactor programs for a coordinated support of their national efforts in the R&D areas:

- Thermo-physical properties and irradiation behavior of fuel forms:

  oxide, nitride, carbide, metal, and others (if any)

- Thermo-physical properties and irradiation behavior of cladding materials:

  austenitic, ferritic-martensitic, high-nickel alloy, ODS steel, and others

- Validation, verification and benchmarking of FR fuel performance codes
- Trend and current status of fuel development:

  fuel design for existing and planned fast reactors, advanced fuel design for attaining high burnup, fuel development for minor actinide transmutation

- Other issues, such as compatibility of stainless steels with heavy liquid metals, etc.
- Close collaboration and information exchange with the OECD/NEA Expert Group on Innovative Fuels

Consultancy and Technical Meetings

Consultancy (CM) and Technical (TM) Meetings	2016	17	18	19
TM-1 on Beyond 5% enrichment limit for LWR: perspectives and problems (1), with NS, NE lead, 12-16 October 2015				
CM on Review of Fuel Failures in Water Reactors in 2006-1015				
TM-2 on Beyond 5% enrichment limit for LWR: perspectives and problems				
CM/TM on Pellet-Cladding Interaction-Stress Corrosion Cracking in Water Cooled Reactors: Operational experience, modelling and R&D about potential remedies			CM	TM
CM on Fuel Materials for Fast Reactors (to launch new CRP)				

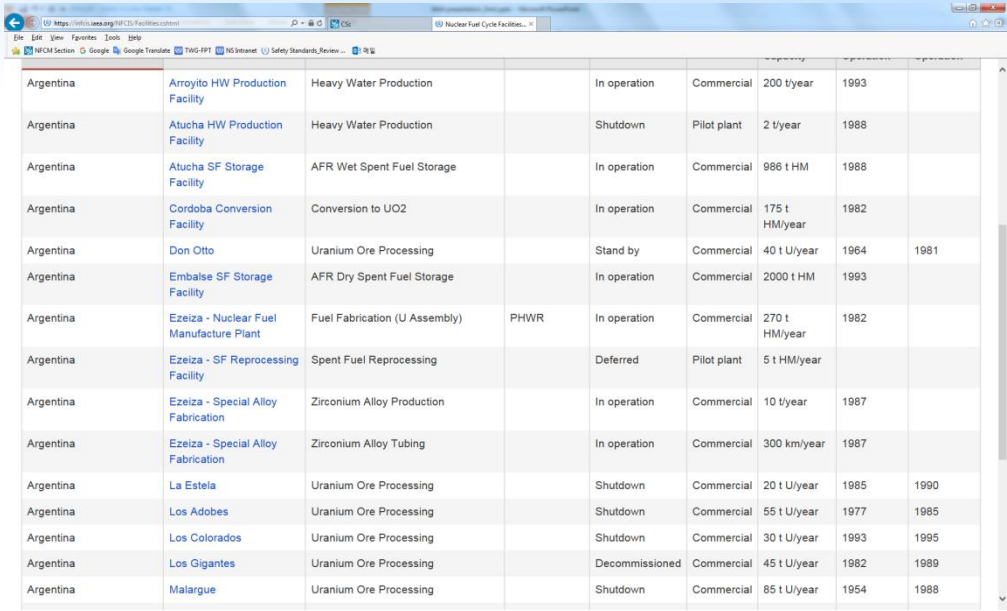
Database systems: INFCIS

- Integrated individual 6 database systems (NFCIS, NFCSS, PIE Facilities Database, UDEPO, ThDEPO, MADB)
- Individual databases were initially developed in electronic database form in 1980's
- Web based system with public access since 2001
- On IAEA NUCLEUS platform since 2012/13

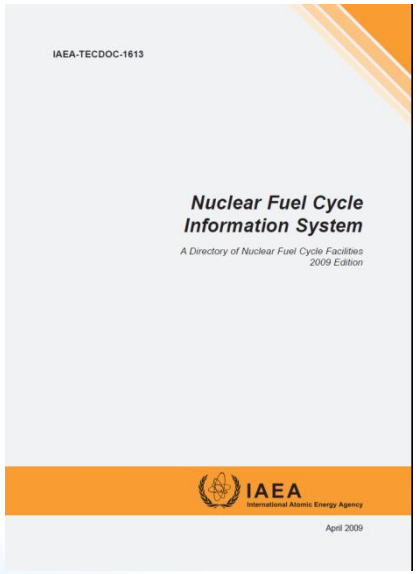


Database systems: NFCIS

- Directory of civilian nuclear fuel cycle facilities covering: mining, milling, conversion, enrichment, fabrication, spent fuel storage, SF reprocessing/recycling, heavy water production, Zircaloy facilities
- Currently including 715 facilities from 55 MS
- Annual update through questionnaire to officially nominated contact points
- TECDOC published in 2009
- New TECDOC is under preparation to support nuclear fuel cycle management analysis and understanding

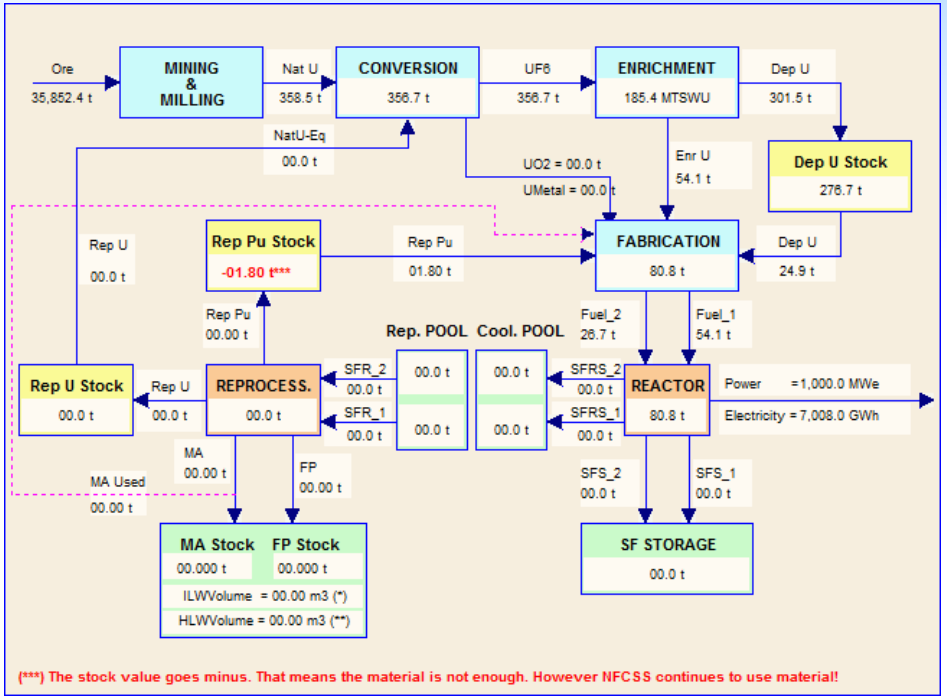


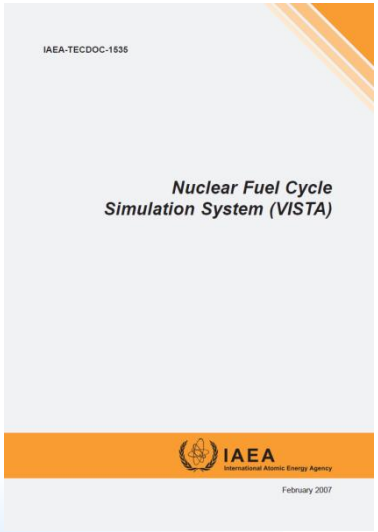
Argentina	Arroyito HW Production Facility	Heavy Water Production		In operation	Commercial	200 t/year	1993	
Argentina	Atucha HW Production Facility	Heavy Water Production		Shutdown	Pilot plant	2 t/year	1988	
Argentina	Atucha SF Storage Facility	AFR Wet Spent Fuel Storage		In operation	Commercial	986 t HM	1988	
Argentina	Cordoba Conversion Facility	Conversion to UO2		In operation	Commercial	175 t HM/year	1982	
Argentina	Don Otto	Uranium Ore Processing		Stand by	Commercial	40 t U/year	1964	1981
Argentina	Embalse SF Storage Facility	AFR Dry Spent Fuel Storage		In operation	Commercial	2000 t HM	1993	
Argentina	Ezeiza - Nuclear Fuel Manufacture Plant	Fuel Fabrication (U Assembly)	PHWR	In operation	Commercial	270 t HM/year	1982	
Argentina	Ezeiza - SF Reprocessing Facility	Spent Fuel Reprocessing		Deferred	Pilot plant	5 t HM/year		
Argentina	Ezeiza - Special Alloy Fabrication	Zirconium Alloy Production		In operation	Commercial	10 t/year	1987	
Argentina	Ezeiza - Special Alloy Fabrication	Zirconium Alloy Tubing		In operation	Commercial	300 km/year	1987	
Argentina	La Estela	Uranium Ore Processing		Shutdown	Commercial	20 t U/year	1985	1990
Argentina	Los Adobes	Uranium Ore Processing		Shutdown	Commercial	55 t U/year	1977	1985
Argentina	Los Colorados	Uranium Ore Processing		Shutdown	Commercial	30 t U/year	1993	1995
Argentina	Los Gigantes	Uranium Ore Processing		Decommissioned	Commercial	45 t U/year	1982	1989
Argentina	Malargue	Uranium Ore Processing		Shutdown	Commercial	85 t U/year	1954	1988



Database systems: NFCSS

- Scenario based simulation system to estimate fuel cycle material flow and service requirements
- Capable of calculating actinide accumulation in spent fuels
- TECDOC published in 2007
- Thorium fuel options will be available shortly
- TECDOC is under revision

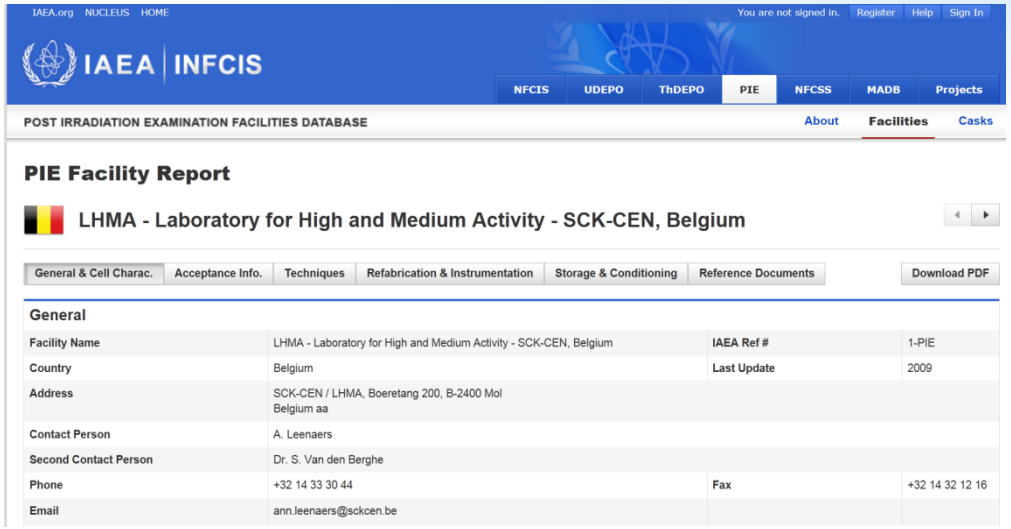




Database systems: PIEDB

The DB consists of 5 main areas describing PIE facilities:

- Acceptance criteria for irradiated components
- Cell characteristics
- PIE technology
- Refabrication/instrumentation capabilities
- Storage and conditioning capabilities
- As well as major technical and licensing data of casks



423 facilities from 22 countries and 21 casks

Conclusion

- **Fuel Engineering Team** at IAEA continues to provide assistance to MS so as to ensure the reliability of power reactor fuels and associated adequate safety margins
- The performance of nuclear fuel is key to the safe operation of nuclear power plants
- Development and modelling of advanced fuel types that can better withstand the extreme conditions found in severe accident conditions (FUMAC, ACTOF) will improve safety margins

IAEA 60 Years
Atoms for Peace and Development

THANK YOU,

and welcome
to the IAEA projects!



Dry storage of spent nuclear fuel and high active waste in Germany

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Abstract:

With the Site Selection Act in 2013 the discussion about a final repository in Germany started again from scratch. According to this act the site selection procedure should be finished in 2031. Considering the necessary time for erection and commissioning, 2050 or even later is a likely date for realizing an operating final repository in Germany. Since 1995 spent nuclear fuel (SNF) and vitrified high active waste (HAW) is stored in casks under dry conditions. According to the existing licenses, the storage time for SNF and HAW is limited to forty years starting with the loading date. The same license limitation applies to the storage facility starting with the first stored cask. Having a date later than 2050 in mind for the begin of the operation of the final repository the interim storage period has to be extended to sixty and more years, meanwhile one hundred years are in discussion. As a consequence, the licenses have to be renewed for both, the casks and the storages facilities.

Important for dry storage is, beside the storage itself, the handling, transport and possible unloading and loading of the inventories into new casks and the conditioning for the final storage in a repository. For these handlings the integrity and the maintaining of the geometry of the cask's inventory is essential. Especially the integrity of the SNF rod cladding and the cladding of the vitrified HAW are stipulated as a barrier in the storage concept and are in charge to secure the specified geometry.

To ensure the cladding integrity of SNF the cladding hoop stress and the cladding hoop strain as well as the maximum temperature during drying and storage are limited to certain values for a forty year storage period today. Other cladding degradation mechanism like the inner and outer oxide layer, hydrogen pick up, irradiation damages in the cladding material crystal structure are well understood during fuel operation and pool storage, but the long term consequences for a prolonged storage period still are not considered due to the lack of experience and long term experiments. The same applies to the helium production from the alpha decay and long term fission gas release. Especially high burnup and mixed oxide fuels have properties like higher fission gas release, high burnup structure and a slower decrease of the post irradiation decay heat. Therefore, the number of high burnup or mixed oxide fuel assemblies is limited in most cask loading patterns. Whether these effects have an influence on fuel degradation and on the cladding integrity during the dry storage is not sufficiently examined up to now.

Within this presentation the effects mentioned above will be discussed from TSO point of view, who gave the expert opinion on fuel integrity in the German dry storage licensing procedures to address inter alia the future needs.





DRY STORAGE OF SPENT NUCLEAR FUEL AND HIGH ACTIVE WASTE IN GERMANY

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CONTENT

- Introduction
- Situation of dry Storage in Germany – A brief overview
- Verification concept for inventory's integrity during dry storage
- Prolonged dry storage period – A need for new verifications
- Conclusions

INTRODUCTION – TÜV NORD, FIELDS OF ACTIVITY

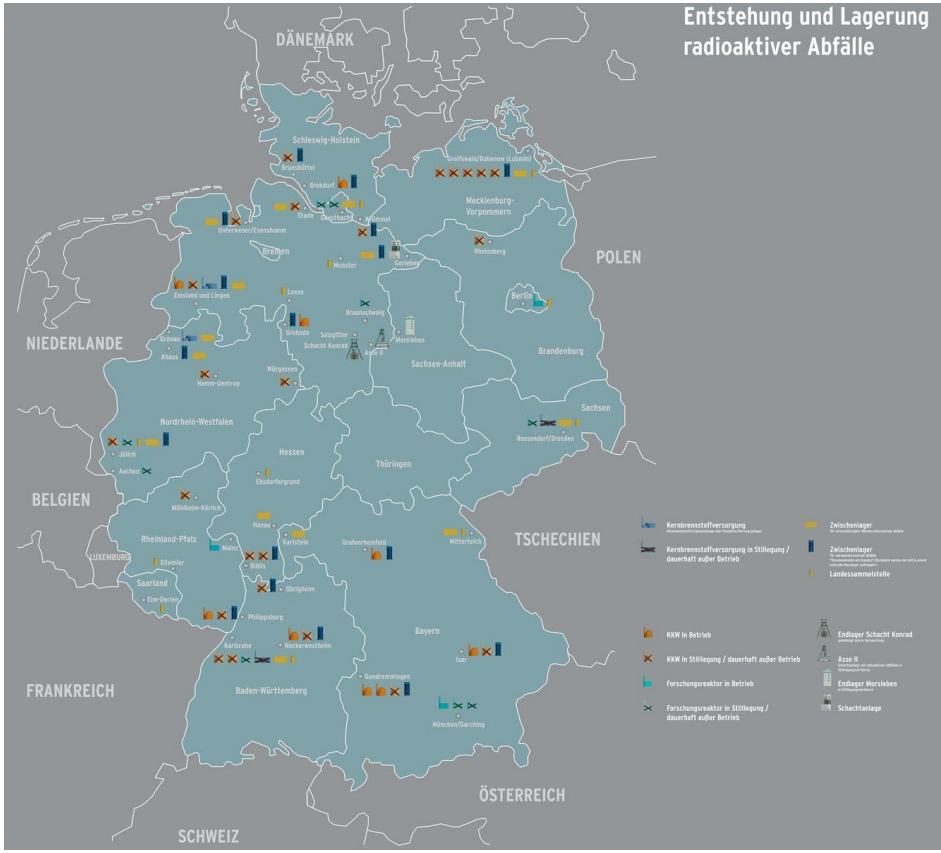
- The TÜV NORD Group is a technical support organization acting in a variety of fields as e.g. safety of conventional technology, education, quality and environmental management as well as nuclear safety.
- TÜV NORD covers all aspects of nuclear safety with an international network.
- TÜV NORD is e.g. in contract for
  - licensing and surveillance of NPPs in operation and in decommissioning,
  - Research reactors,
  - licensing of interim storage facilities,
  - licensing of casks and
  - surveillance of interim storage facilities.



Source: https://de.wikipedia.org/wiki/Liste_der_Kernreaktoren_in_Deutschland

SITUATION OF DRY STORAGE IN GERMANY – A BRIEF OVERVIEW

- In Germany Spent Nuclear Fuel (SNF) High Active Waste (HAW) is stored in casks under dry conditions since 1995:
  - 3 central interim storage facilities (Gorleben, Ahaus und Lubmin)
  - 12 interim storage facilities at the NPP sites
  - AVR-Cask storage facility Jülich
- Besides SNF casks 113 casks with HAW are stored in Gorleben and Lubmin.
- Germany has the obligation to take back some casks with HAW and MAW from fuel reprocessing.



Source: BFE, http://www.bfe.bund.de/EN/nwm/interim-storage/decentralised/sites-occupancy/sites-occupancy.html

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SITUATION OF DRY STORAGE IN GERMANY – A BRIEF OVERVIEW

- Expected amount of radioactive waste in Germany for final repositories:
  - 1100 casks with heavy metal from LWR reactors
  - Casks with HAW and MAW from reprocessing
  - App. 300000 m³ with low active waste (final repository Konrad)
  - App. 220000 m³ from retrieval from the repository ASSE II

  (Source: http://www.bmub.bund.de/themen/atomenergie-strahlenschutz/nukleare-sicherheit/sicherheit-endlager/nationales-entsorgungsprogramm/)
- Expected interim storage periods:
  - Interim storage facilities including the casks are licensed for 40 years
  - Final repository site selection act (last change on 5th May 2017): Final repository will not open before 2050
  - Need for interim storage of SNF for decades beyond 40 years

VERIFICATION CONCEPT FOR INVENTORY'S INTEGRITY DURING DRY STORAGE

- Legal provisions / Technical Rules and Regulations for nuclear issues in Germany:
  - Basic Law / Atomic Energy Act
  - Ordinances
  - General Administrative Provisions
  - Safety Requirements for Nuclear Power Plants
  - Regulatory Guidelines by the BMUB
  - Reactor Safety Commission Guidelines (RSK-, SSK-, ESK-Leitlinien)
  - KTA Safety Standards (KTA-Regelwerk)
  - EU legislation

  (Source: http://www.bmub.bund.de/en/topics/nuclear-safety-radiological-protection/nuclear-safety/legal-provisions-technical-rules/)
- Important for the interim storage of SNF and HAW:
  - German Nuclear Waste Management Commission (ESK)
    ESK Guidelines and Recommendations (ESK-Leitlinien)

VERIFICATION CONCEPT FOR INVENTORY'S INTEGRITY DURING DRY STORAGE

- Cask Types
  - CASTOR cask types
  - TN cask type in licensing process (PWR fuel)
- Main fuel designs
  - PWR fuel from KWU Type NPPs from different vendors and PWR fuel from VVER 440
  - BWR fuel from several vendors
  - Fuel from pebble bed reactors (THTR and AVR)
- Burnup limitations (example for PWR fuel, CASTOR type cask)
  - Uranium and mixed oxide fuel with an average burnup up to 55 MWd/kgHM
  - High burnup fuel with an average burnup up to 65 MWd/kgHM is limited to a number of four assemblies on special positions in the cask
  - Number of MOX fuel with an average burnup up to 65 MWd/kgHM is limited to a number of six assemblies on special positions in the cask

VERIFICATION CONCEPT FOR INVENTORY'S INTEGRITY DURING DRY STORAGE

- ESK Approach: Barrier Concept in dry storage of SNF and HAW in casks
  - Function: Retention of fission products and prevent ingress of matter from outer environment.
  - Barriers for spent nuclear fuel storage
    - Fuel matrix
    - Fuel rod cladding tubes
    - Canister / Cask with welded or bolted lid
  - Barriers for vitrified waste storage
    - Glass matrix
    - Coquilles cladding
    - Canister / Cask with welded or bolted lid

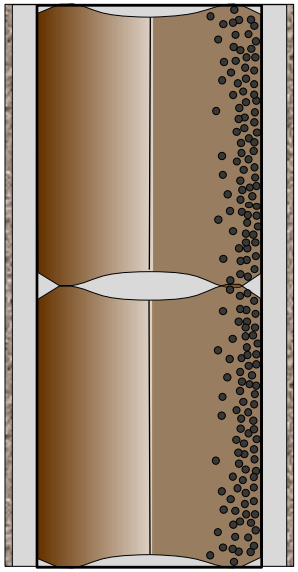
Comparable barriers for both inventory types:

- Current assessments show the integrity of the fuel rod cladding over 40 years of storage.
- For the coquilles cladding no limitations are established for the time of storage.

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VERIFICATION CONCEPT FOR INVENTORY'S INTEGRITY DURING DRY STORAGE - BARRIERS

- Fuel matrix:
  - Approximately 80 % of the gaseous fission products will remain trapped in the uranium fuel pellet
    - in bubbles at grain boundaries,
    - in intergranular bubbles or
    - Is somewhere trapped in the crystal lattice e.g. at interstitials.
  - The fuel matrix has a retention function with respect to the gaseous fission products.



Source: TÜV NORD

VERIFICATION CONCEPT FOR INVENTORY'S INTEGRITY DURING DRY STORAGE - BARRIERS

- Fuel rod cladding tubes:
  - Approximately 20 % of the gaseous fission products are released to the free volume of the fuel rod and increases the inner pressure.
  - Approximately half of the fuel rod gaseous inventory results from the filling gas added during manufacture.
  - Cladding integrity can be ensured by limitation of:
    - Maximum cladding temperature during drying and storage
    - Maximum hoop stress caused by gas pressure
    - Maximum hoop strain caused by creep during storage
    - Corrosion during storage
  - The intact fuel rod cladding tube is a tight barrier for fission products.
  - Cladding integrity prevents fuel relocation which has an important impact on fuel handling and e.g. thermal or radiological assessments.
  - Releases of fission products into the cask free volume are caused by fuel rod cladding tube failure.



Source: TÜV NORD

VERIFICATION CONCEPT FOR INVENTORY'S INTEGRITY DURING DRY STORAGE - BARRIERS

- Dual purpose cask for storage and transportation:
  (example: CASTOR® – type cask)
  - Monolithic cask body made from spheroidal graphite iron.
  - Bolted double lid system with a monitored gas plenum between the lids.
  - Metal spring energized seals with defined leaking rate.
  - Neutron moderator in cask body wall, below secondary lid and at the bottom part.
  - Inner basket for positioning of spent fuel or coquilles.
  - Trunnions for handling operations at both ends.
- For long time storage the most critical points are:
  - Seals
  - Moderator material
  - Trunnions



Source: GNS Gesellschaft für Nuklear-Service mbH, www.gns.de

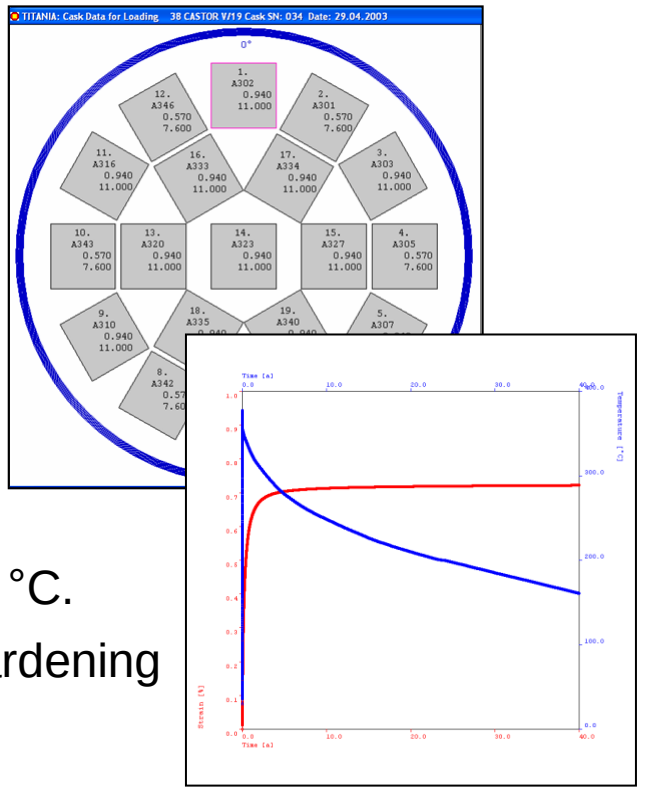
VERIFICATION CONCEPT FOR INVENTORY'S INTEGRITY DURING DRY STORAGE

- Within the limitation of the storage time to 40 years the following requirements on an exclusion of a systematic cladding failure are formulated:
  - The cladding hoop stress should not exceed 120 N/mm².

    Within this criterion stress corrosion cracking and hydride reorientation as failure mechanisms will be excluded.
  - The cladding hoop strain should not exceed 1 %.

    Within this criterion an increase of the hoop stress at higher strain levels and the failure caused by strain will be excluded.
  - The cladding temperature should not exceed 370 °C.

    Within this criterion the annealing of irradiation hardening and the dissolution of the hydrogen precipitates in the cladding will be excluded.
  - Cladding corrosion during the storage time should be limited.



Source: TÜV NORD

PROLONGED DRY STORAGE PERIOD – A NEED FOR NEW VERIFICATIONS

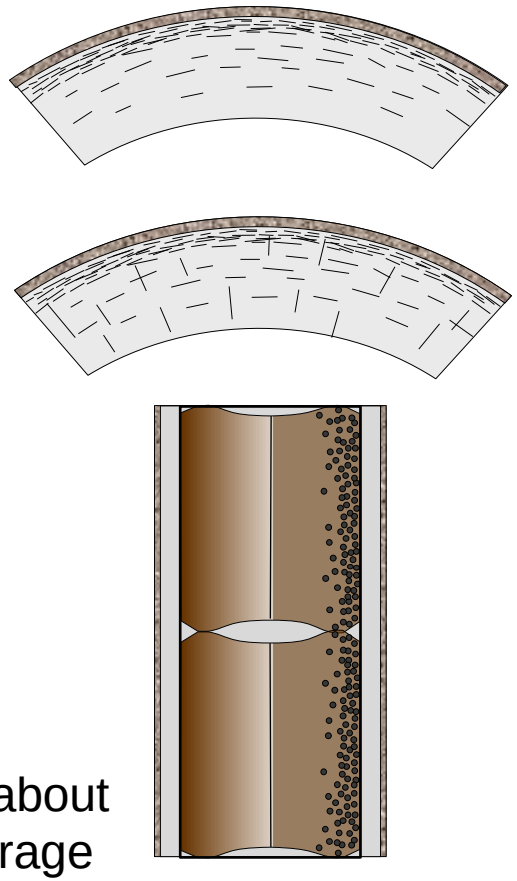
- It has to be evaluated, whether the requirements as formulated for the storage of 40 years are still valid and applicable for a longer time span.
- In the long term other effects of degradation may take the leading part in the assessment of cladding integrity.
- High burnup fuel and mixed oxide fuel are identified as the fuel with the highest loads and with the highest potential for a systematic cladding failure.
  (see also IAEA Standard on Storage of Spent Nuclear Fuel)
- Up to now the examinations carried out on spent fuel after a dry storage period do not cover high burnup fuel or mixed oxide fuel.
  (see also Examination of Spent PWR Fuel Rods after 15 Years in Dry Storage, NUREG/CR-6831 and ANL-03/17 reports, 45 MWd/kgHM)

➡ What measures are necessary to ensure SNF- and HAW-Integrity or to have knowledge about it after 100 years or more?

PROLONGED DRY STORAGE PERIOD – A NEED FOR NEW VERIFICATIONS

- Considerations for measures to assess cladding integrity during an extended storage time:
  - Reorientation of hydrogen precipitates
  - Fission gas release from high burnup structure
  - Cladding degradation by irradiation damages and fuel side chemical interactions
  - Annealing of irradiation damages and the consequences on cladding ductility and creep
  - Cladding embrittlement processes

  and
  - Handling of risks of loosing data and knowledge about the fuel which will be kept in long time interim storage after the nuclear phase out in 2022 in Germany?



Source: TÜV NORD

PROLONGED DRY STORAGE PERIOD – A NEED FOR NEW VERIFICATIONS

- Needed and ongoing R&D work:
  - Results from the instrumented cask in EPRI Extended Storage Collaboration Program to verify modeling work.
  - Accumulation of the technical knowhow among other sources in the licensing and surveillance processes (TÜV NORD participation).
  - Model development for hydrogen uptake, dissolving and precipitation to calculate the reorientation of hydrogen precipitates with fuel rod performance codes (TÜV NORD participation).
  - Investigations on data from manufacturing, in pile life and storage of the fuel which are needed to predict today's requirements and what might be needed for future investigations (TÜV NORD participation).
  - Analyzing process for data and knowledge gaps and no "wait and see" during the storage time.

CONCLUSIONS

- Interim dry storage of spent fuel and high active waste:
  Common practice in Germany and other countries.
- The validity of proofs are limited to 40 years.
- A long-term interim dry storage for much longer periods is to be expected.
- Data and knowledge management aspects are as important as technical considerations.
- Research has been initiated.
- TÜV NORD contributes its expertise in research projects.

UK Research in LOCA-Related Activities and Co-operation with the Karlsruhe Research Centre– A Historical Perspective

John Lillington (AMECFW, UK), Simon Walker (Imperial College, UK), Martin Steinbrück (Karlsruhe Institute of Technology, Germany), Juri Stuckert (Karlsruhe Institute of Technology, Germany)

ABSTRACT

From the 1970s onwards, very extensive research on Loss-of-Coolant Accidents (LOCA) was carried out in the UK, well supported by the industry and regulator, much of it in support of the safety case for the Sizewell B PWR, a Westinghouse 4-loop design. This work involved separate effect and bundle tests on Zircaloy oxidation and creep, with investigation of methods for defining and measuring bundle blockage. The dynamic bundle ballooning tests were supplemented by bundle tests with a fixed blockage to investigate in detail coolability and coolant bypass effects, and by pellet stack vibration tests investigating potential fuel slumping under LOCA conditions. There was extensive international collaboration with experimental teams particularly in the US, Germany and France, and the OECD Halden Reactor programme in Norway. Cooperation with the then Kernforschungszentrum Karlsruhe (KfK, now Karlsruhe Institute of Technology, KIT) was particularly important, concerning small-scale creep/ballooning and oxidation tests on Zircaloy cladding, along with bundle/reflood tests in the REBEKA facility. The knowledge gained was encapsulated in the UKAEA modelling codes CANSWEL-2 for clad ballooning (2D r-theta) treatment taking account of pellet/clad eccentricity and associated azimuthal temperature gradients) and MABEL-2 (with CANSWEL-2 as the deformation module) for calculating ballooning and blockage in bundle geometries and for reactor safety assessment, including mutual feedback with thermal hydraulic effects and channel blockage.

Following this major effort, research continued including small-scale material tests particularly concerning the properties of advanced cladding materials, such as binary alloys of the Zr-Nb type, so that the LOCA response of cores containing these materials could be assessed. More recently a new build programme in the UK, based on PWR and BWR technology, has been established, and research capability is being established relevant to their safety needs. The MABEL-2 code has been further developed by Imperial College into the MATARE code, with additional thermal hydraulic capability, and this remains the reference UK tool for independent assessment for clad deformation and reflood response during a LOCA.

This presentation summarises the progress made over these years, indicating the ongoing development of the MATARE code, and emphasising the cooperation with KIT. New work identified here is being taken into account in an updated CSNI SOAR on fuel behaviour during loss of coolant accidents, currently in progress.

Presentation given at the 23rd International QUENCH Workshop, Karlsruhe Institute of Technology, 17-19 October 2017.


2017
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Imperial College London

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Karlsruhe Institute of Technology

18th October 2017

UK Research in LOCA-Related Activities and Co-operation with the Karlsruhe Research Centre – A Historical Perspective

23rd International QUENCH Workshop

Karlsruhe Institute of Technology, Germany

17th - 19th October 2017

Simon Walker(1), John Lillington(2), Martin Steinbrück(3), Juri Stuckert(3)
presented by David Bottomley

(1) Imperial College, London, UK; (2) Wood Nuclear & precursor organisations, Dorchester, UK; (3) Karlsruhe Institute of Technology, Germany;

OUTLINE

- Introduction
- Zr alloy clad ballooning studies
  - Creep rupture tests
  - Clad oxidation tests
  - Tests on the interaction of creep and oxidation
  - Mechanical interaction tests
  - Pellet stack collapse tests
  - MERLIN bundle test
- Thermal hydraulic bundle tests
  - THETIS, ACHILLES (UKAEA)
  - Bundle tests (CEGB)
- Modelling studies
  - MABEL/CANSWEL
  - MATARE
- Use of KfK/FZK/KIT clad ballooning and oxidation data for modelling
- Conclusions

INTRODUCTION (1/2)

- The CSNI state-of-the art report NEA 6846 of 2009 [OECD/NEA, 2009] on LOCA is being updated in the light of newly-available data;
- One source of newly-available data is a collection of documents identified in the UK on the very extensive studies performed in the 1980s-1990s in support to the regulator of the safety case for the Sizewell B PWR (SXB, Westinghouse 4-loop, modified SNUPPS design), not then in the public domain, and this new work is summarised here:
  - CEGB and UKAEA separate-effect tests on Zry oxidation and creep, with single-rod ballooning studies, including on irradiated cladding;
  - UKAEA MERLIN multi-rod test facility, similar to REBEKA, abandoned due to technical problems;
  - UKAEA pellet stack stability studies, including 2D on-line radioscopy, including the effect of prototypic reflood vibrations;
  - Modelling studies using the CANSWEL2/MABEL2 clad ballooning code, on effects of different alloys, the interaction of oxidation and creep, and different rupture criteria;
  - These are illustrated by a sample of reports from Wood Nuclear archives (was UKAEA/AEA Technology/Serco), with a view to further investigation.

INTRODUCTION (2/2)

- Also summarised here, though not in so much detail, are the thermal hydraulic tests performed in the UK on reflood of pre-deformed bundles; THETIS & ACHILLES (UKAEA), and the CEGB series at Berkeley Nuclear Laboratories, UK;
  - THETIS experiments were summarised in NEA 6846, while these along with the other two series are accounted for in more detail in an IRSN SOAR review of 2006 [Grandjean, 2005-2008];
- The opportunity has also been taken to draw attention to the important collaboration between KfK/FZK/KIT with regard to use of relevant data for code development and validation (*note also the presentation by Juri Stuckert, this meeting, on the ongoing design-basis QUENCH-LOCA experiments*);
- Since the construction of Sizewell B, capability in the area of LOCA modelling is maintained though creep tests on various candidate alloys for this reactor etc., and in development and validation of the MATARE code.

UK separate-effects studies on Zry oxidation

- Extensive series of oxidation tests under LOCA conditions performed by UKAEA in the late 1980s to support the SXB safety case and for CANSWEL2 development;
- Three materials were examined in detail, temperatures 700°C-1000°C, exposure times 125 s – 1000 s, in the COZICAT facility, data compared with KfK measurements of Leistikow and Schanz and UKAEA PROPAT and FOURSQUARE data (referenced in the LOCA SOAR); trends consistent;
- Results were used to derive an empirical cubic/parabolic Zry oxidation model for CANSWEL2, [Haste et al., 1988], also used in ORNL BWR control blade modelling for SCDAP, MELCOR etc..

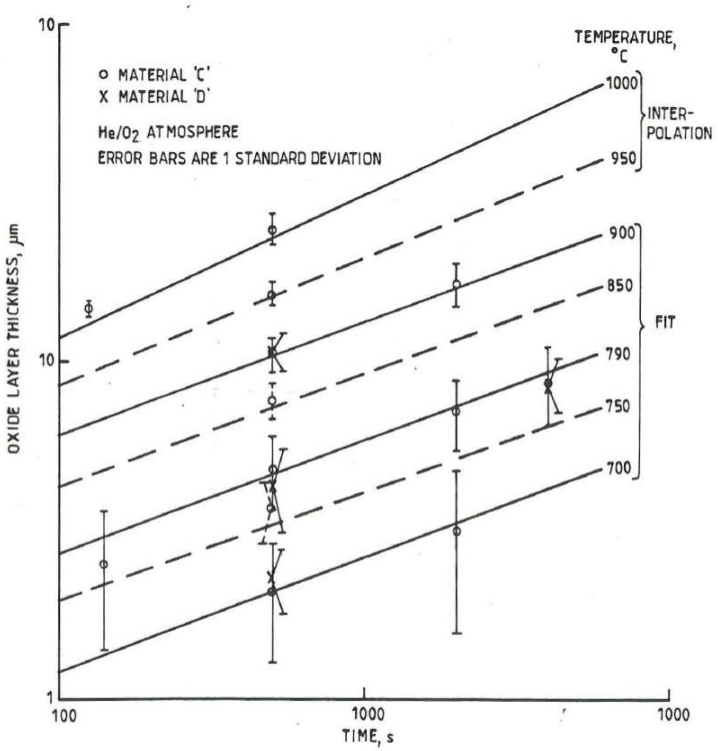
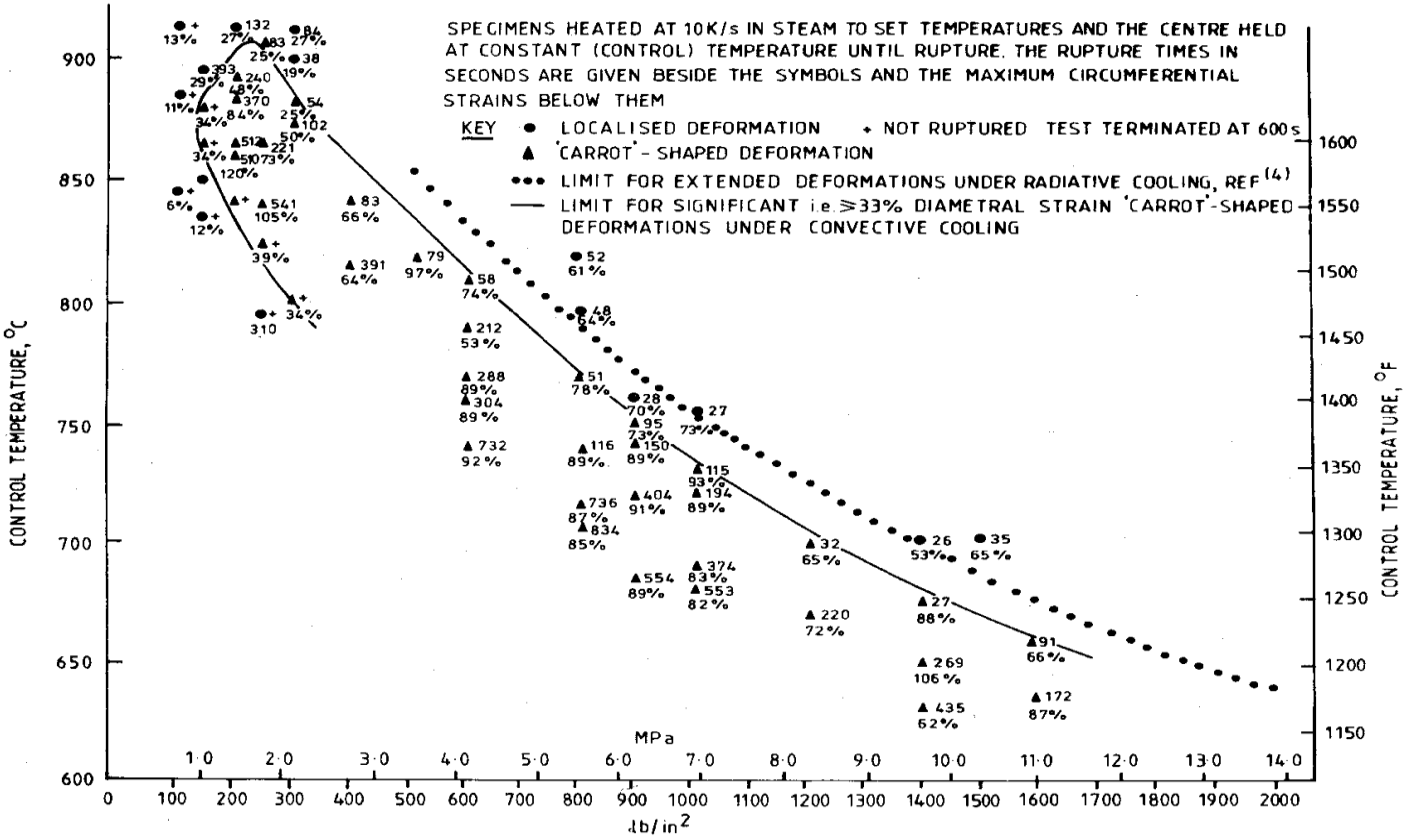


FIG. 7. COMPARISON OF MATERIAL 'C' AND MATERIAL 'D' OXIDE LAYER DATA, He/O_2 ATMOSPHERE

UK separate-effects studies on Zry ballooning (1/4)

- Extensive series of single rod cladding creep (ballooning) measurements were carried out by UKAEA SprIngfields and CEGB Berkeley Nuclear Laboratories [e.g. Donaldson et al., 1985]) on Zr alloys to characterise candidate materials for the Sizewell B PWR, and for integral experiments such as Halden IFA-54X and NRU MT3);
- Experiments up to the early 1980s are well summarised in the LOCA SOAR (e.g. SprIngfields vertical single rod facility PROPAT), steam atmosphere;
- Later CEGB studies concentrated on biaxial creep tests at constant pressure in inert Ar atmosphere, mainly in high α-phase (<820°C) of most interest to SXB design basis LOCA (slow ramp in high α-phase), but also in the phase change region (820°C–980°C) and β-phase (>980°C);
- UKAEA studies in the then new COZICAT rig studied the effect of oxidation on creep and overall ductility, using tests in inert and oxidising atmospheres (He/5%O_2 to simulate steam), including ballooning pre-oxidised rods in an inert atmosphere to examine effect of oxide strengthening and cracking; used to develop CANSWEL2 models, along with Canadian and German data [UKAEA unpublished work]:
- Also CEGB tests at constant stress (closed tube) [Donaldson and Knowles, 1983] in support of studies on rupture criteria, showed need to use ductility-based (damage fraction) rather that stress-based criteria to evaluate clad strain/channel blockage.

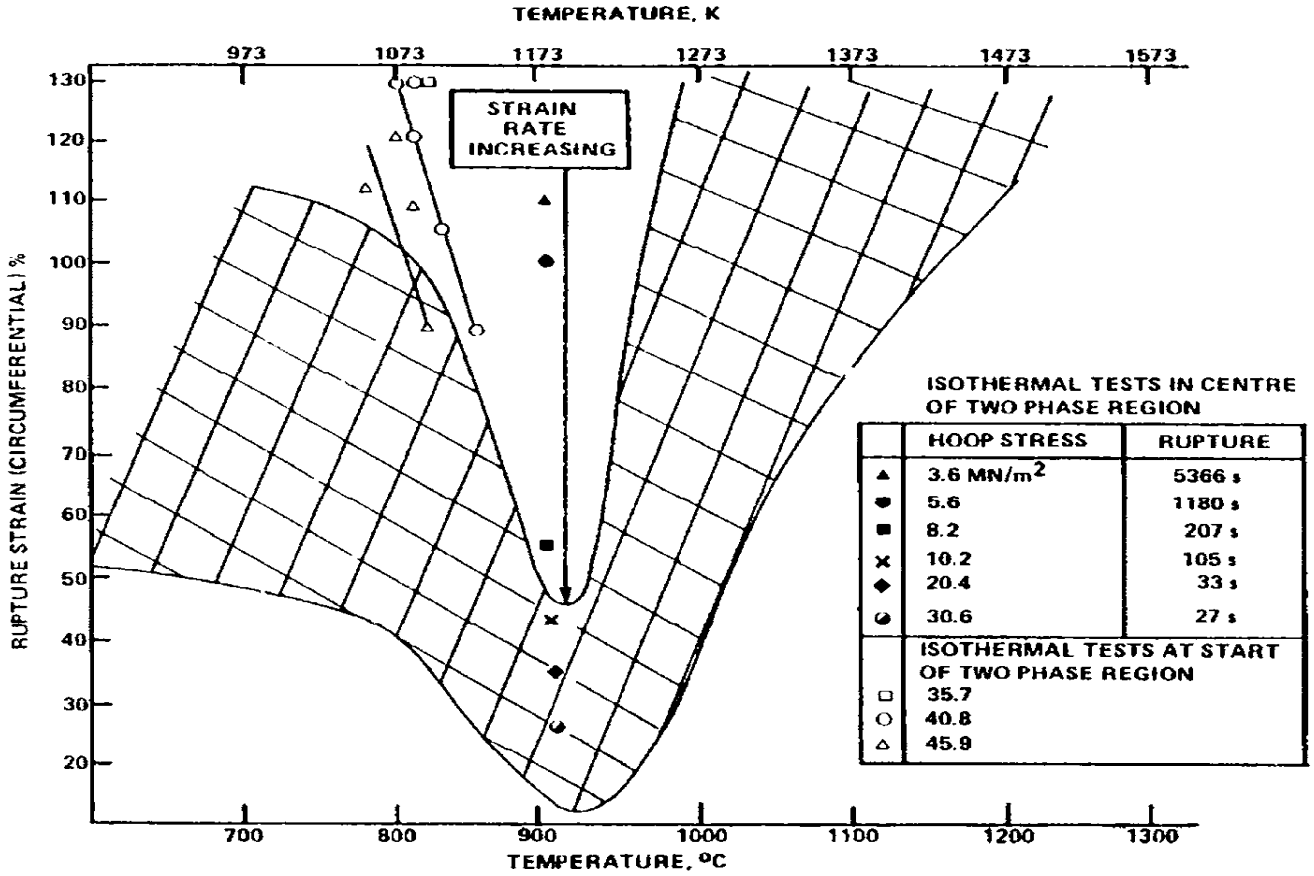
UK separate-effects studies on Zry ballooning (2/4)



Results of stylised transient testing of Zircaloy-4 PWR cladding at UKAEA Springfields (PROPAT facility, direct resistance heating, maximises ballooning) with mainly convective cooling by steam

- Later experiments used a steam substitute (He/5%O_2) that gave a similar oxidation rate to steam (but no hydrogen formation)

UK separate-effects studies on Zry ballooning (3/4)



Variation of tube rupture strain with rupture temperature in isothermal tests at UKAEA Springfields

UK separate-effects studies on Zry ballooning (4/4)

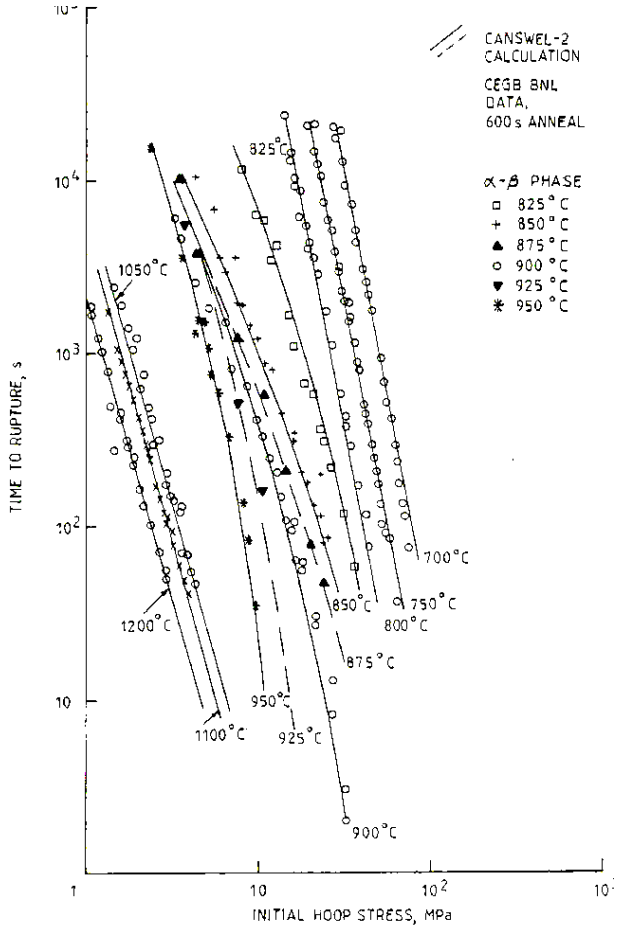


FIG. 8. DEPENDENCE OF TIME TO RUPTURE ON HOOP STRESS WESTINGHOUSE TUBING

Examples of UKAEA Springfields COZICAT single tube burst data in inert atmosphere; (left) general temperature and time dependence of time to rupture, (right) effect of grain growth in slowing superplastic creep

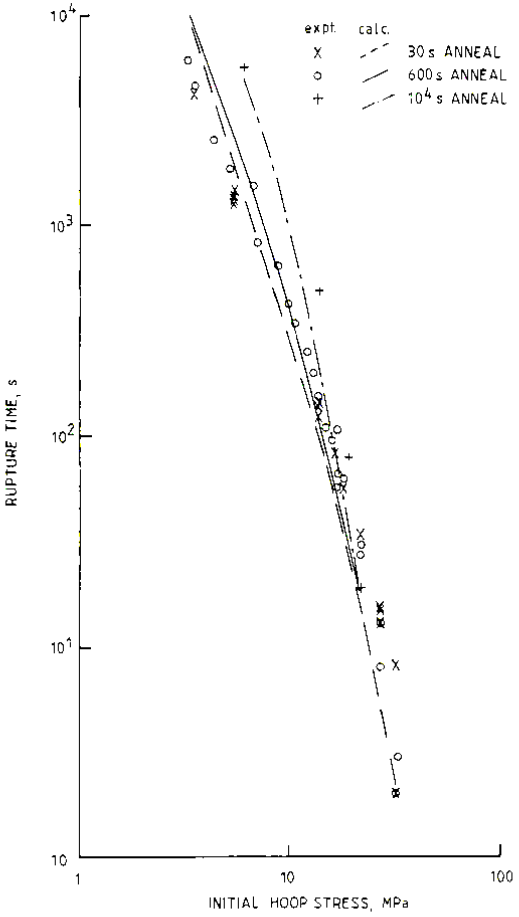
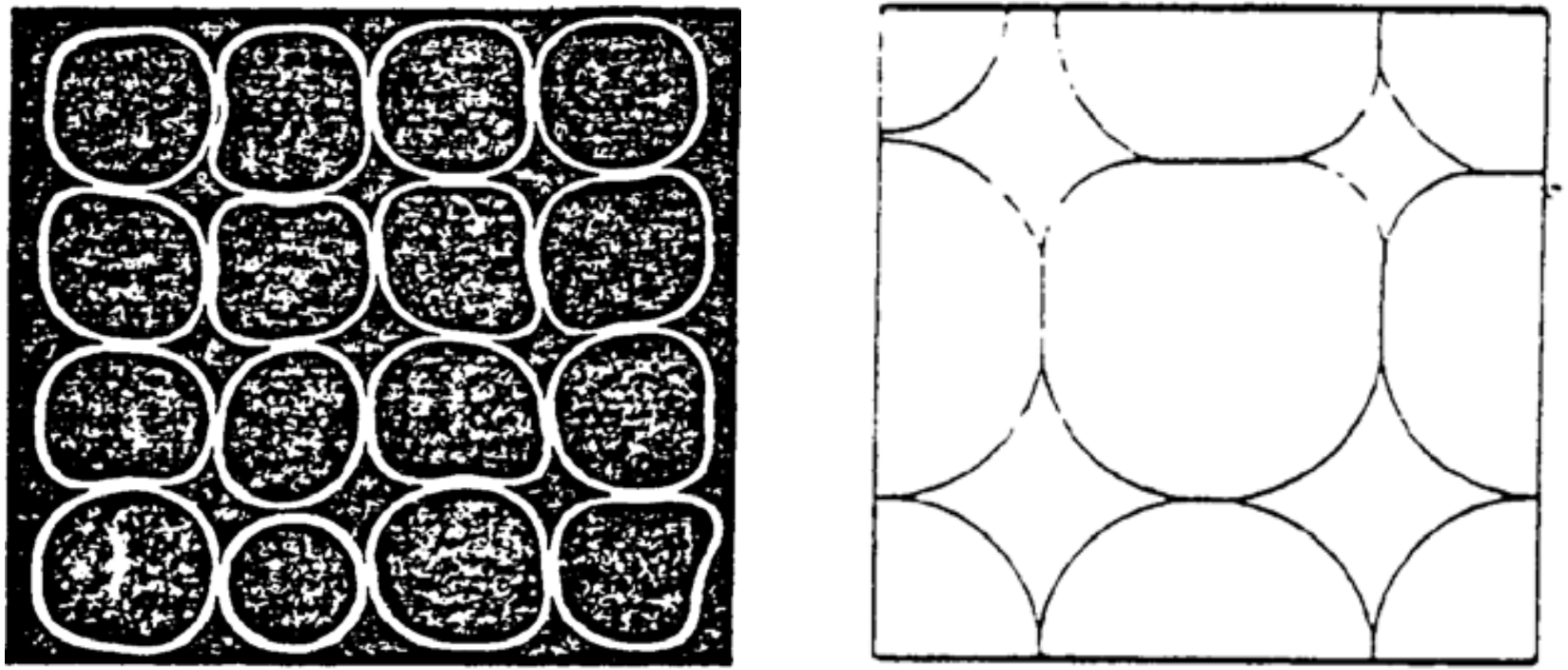


FIG. 9. DEPENDENCE OF TIME TO RUPTURE ON HOOP STRESS, WESTINGHOUSE TUBING, SHOWING EFFECT OF ANNEALING TIME AT 900°C

UK separate-effects studies on rod-to-rod mechanical interaction



Mechanical restraint by deformable neighbours:
(left) Springfields 4×4 cladding deformation cross-sections in the FOURSQUARE facility, furnace heating; (right) CANSWEL-2 idealisation [Haste, March 1982]

UKAEA pellet stack stability studies (1/2)

- Pellet stack stability studies at UKAEA Windscale in the 1980s investigated fuel stack collapse in irradiated (20 GWd/tU) ballooned single rods under LOCA conditions [Garlick et al., 1984, 2 reports];
- LOCA SOAR NEA 6846 summarises initial results up to 1983;
- Further studies performed to 1988 showed that reflood-induced vibrations promoted axial relocation of irradiated pellet fragments, that there existed a threshold strain, 50%, below which large-scale relocation did not occur and that little or no movement occurred below 36% strain (the fuel fragments are too large).

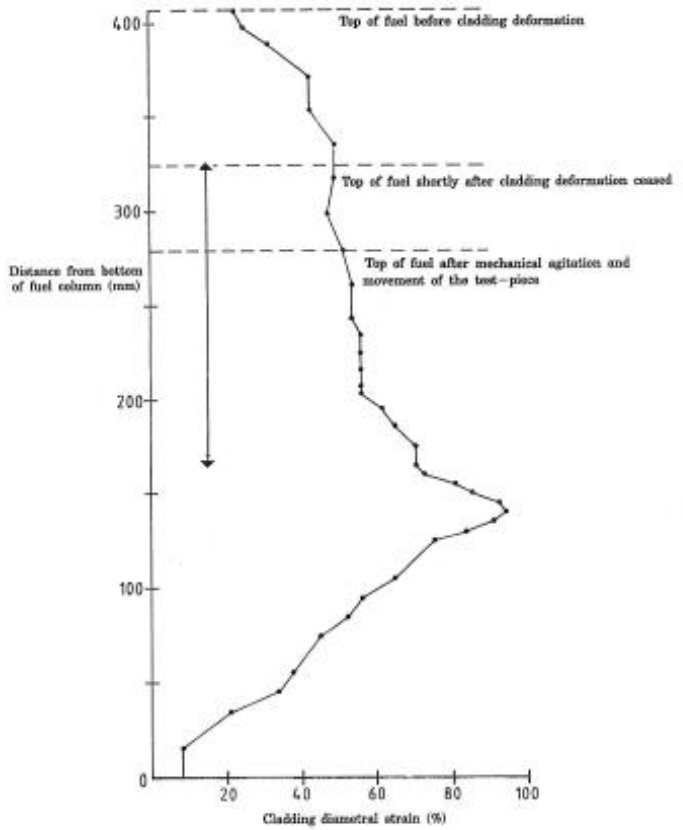


FIGURE 14 CLADDING DIAMETRAL STRAIN DISTRIBUTION AND FUEL AXIAL RELOCATION FOR TEST-PIECE K8/5. (ARROWS INDICATE THE X-RADIOSCOPY FIELD OF VIEW)

Fuel axial relocation and axial strain distribution, from X-radiographs

UKAEA pellet stack stability studies (2/2)

- Concluded that fuel collapse will not increase the likelihood of fuel channel blockage during a postulated LOCA;
- The studies also showed that there was no significant difference between the strains observed for these irradiated rods, and those for unirradiated rods performed at SprIngfields (i.e. irradiation damage anneals out).

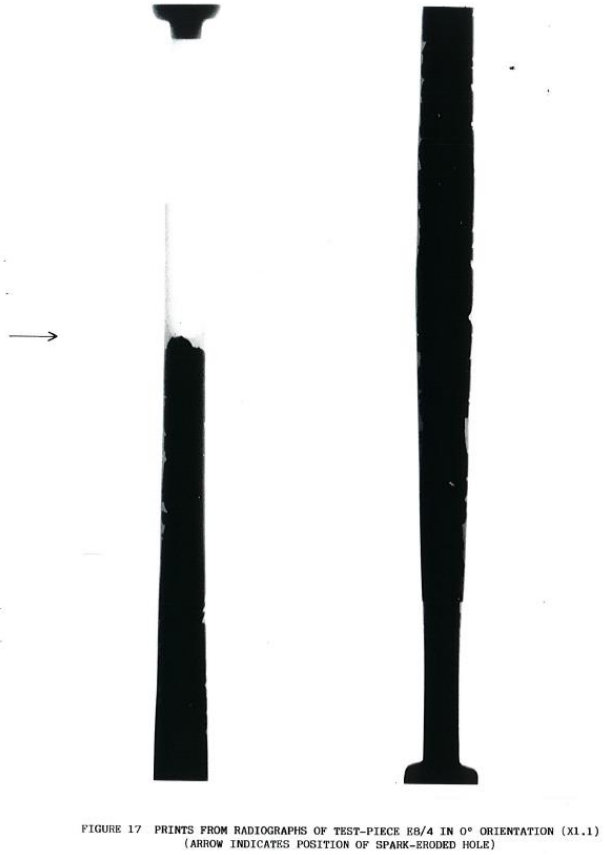


FIGURE 17 PRINTS FROM RADIOGRAPHS OF TEST-PIECE E8/4 IN 0° ORIENTATION (X1.1)
(ARROW INDICATES POSITION OF SPARK-ERODED HOLE)

X-radiographs showing fuel stack collapse, top of rod (left), ballooned region (right)

UK MERLIN experimental programme on clad ballooning)

- MERLIN [Worswick et al., 1989] was an experimental programme at UKAEA SprIngfields intended to provide data relevant to clad ballooning and reflood, particularly to assess the conditions favourable to co-planar blockage, and to provide data for MABEL2 (UKAEA) and BART (Westinghouse) code validation;
- The test section consisted of a 6x6 array of electrically-heated fuel rod simulators. The test sequences envisaged were to be based on those of NRU MT-3, i.e. slow ramp through the high alpha-phase region Zry, 750-820°C, as design basis LOCA for SXB;
- The rig experienced severe technical problems and the project was curtailed (in effect replaced by the REBEKA-7 test);
- Measurements of reflood vibrations were performed, suitable for potential use in pellet stack stability studies.

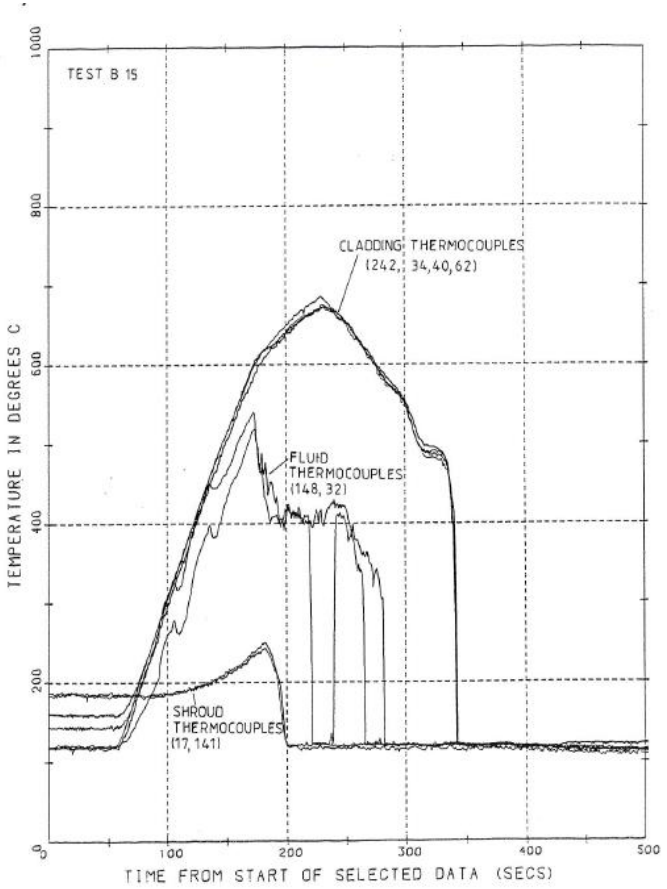
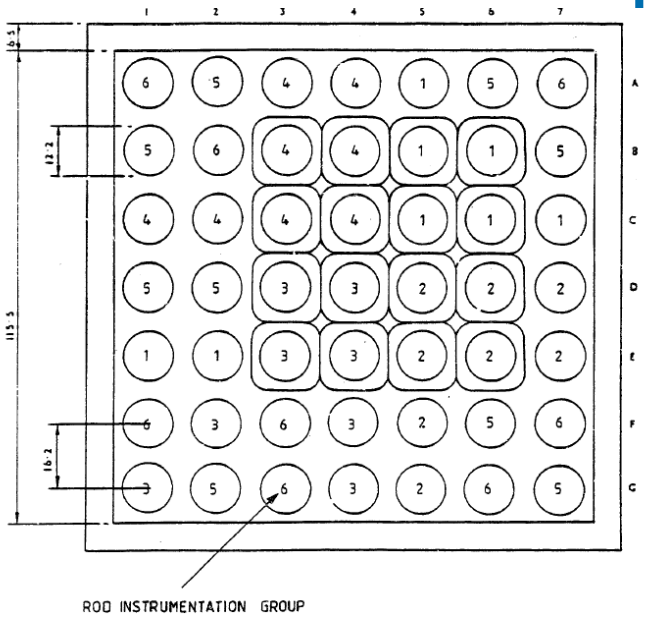


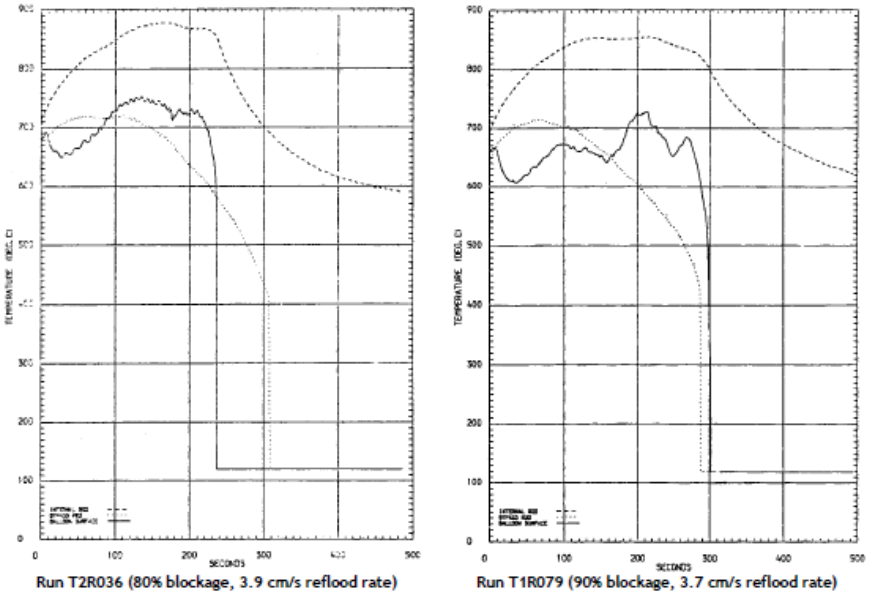
FIG. 12. TEST B15. SELECTED CLADDING, FLUID AND SHROUD THERMOCOUPLE TRACES FROM THE HIGH RATED REGION OF THE BUNDLE (TAKEN FROM REF. 6)

UK bundle reflood experiments with fixed blockage (general)

- A series of bundle reflood experiments was performed in the UK up to the years 2000 to investigate the effects of bundle blockage on the reflood response, with electrically heated bundles featuring pre-formed blockages (e.g. 80%, 90%) of prototypical geometry, see [Grandjean, 2005-2008] ;
- Aim was to investigate the coolability under postulated LOCA conditions of a fuel assembly containing a partial blockage, concentrating on large break LOCA scenarios;
- Investigated in the FEBA and SEFLEX programmes (Germany) and the FLECHT-SEASET programme (USA), summarised in NEA 6846;
- Similar experiments were performed in the UK - the THETIS and ACHILLES facilities (UKAEA Winfrith) and at CEGB Berkeley Laboratories (BNL);
  - THETIS, along with FEBA, SEFLEX & FLECHT-SEASET, already considered in NEA 6846;
  - ACHILLES and CEGB tests to be included in the update, effect on the overall conclusions to be studied (e.g. “blockages of 90% over moderate lengths (<10 cm) do no seem to pose any particular problems concerning coolability”, NEA 6846).

UK bundle reflood experiments with fixed blockage (THETIS)





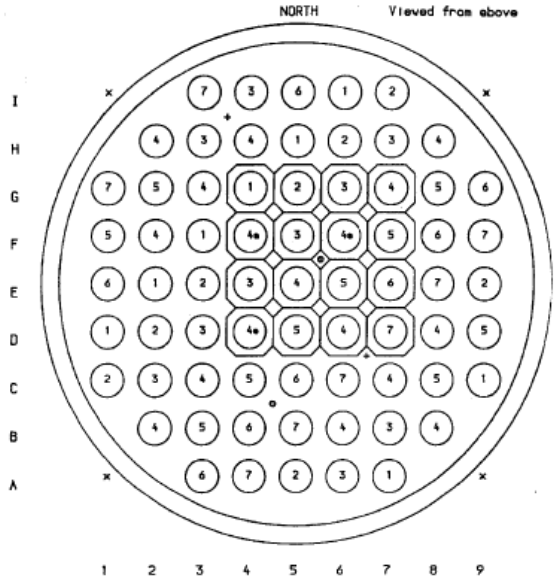
Blockage and bypass temps. for 80% (left) and 90% (right) blockages, circa 4 cm/s reflood rate

THETIS X-section, 49 rods, SGHWR geometry, rod dia. 12.2 mm (c.f. 9.5mm in a 17*17 PWR), 3.58 m heated length, 80% or 90% blockage in a 4*4 square array;
1) Single-phase (N_2) flow heat transfer tests; 2) Forced reflood tests; 3) Gravity reflood tests; 4) Level-swell tests.

Typical results: the series of forced reflood tests, 90% blockage, 2 bar pressure, inlet temperature 90°C, produced the following observations:

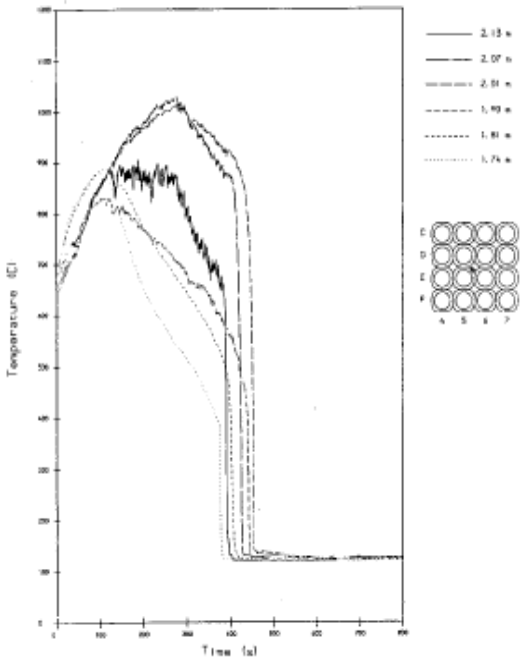
- 3 cm/s reflood rate, the blockage was found to be coolable, with the peak blockage temp. not exceeding the peak by-pass temperature by > 60°C.
- 2 cm/s reflood rate, the maximum blockage temp. rose above the facility operating limit, which made it necessary to reduce power before the complete cooling of the blockage was achieved. It may therefore be believed that these test conditions do not permit suitable blockage cooling.

UK bundle reflood experiments with fixed blockage (ACHILLES)

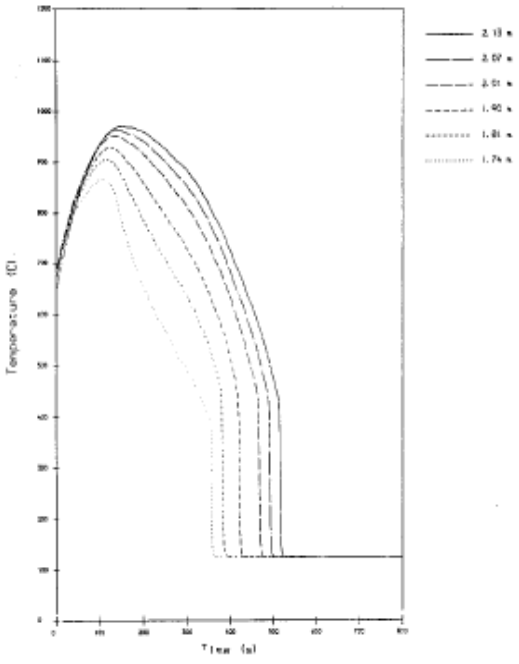


ACHILLES X-section, 69 rods, prototypic PWR geometry, mixing vanes, 3.66 m heated length, 80% blockage in a 4*4 square array, with notional transient conditions;

Pressure:	2 bar
Reflood rate:	2 cm/s (constant)
Inlet subcooling:	20°C
Initial cluster temperature:	650°C
Initial shroud temperature:	600°C
Power:	70% ANS(79)+ 2σ



Run A2R038 (blocked cluster)

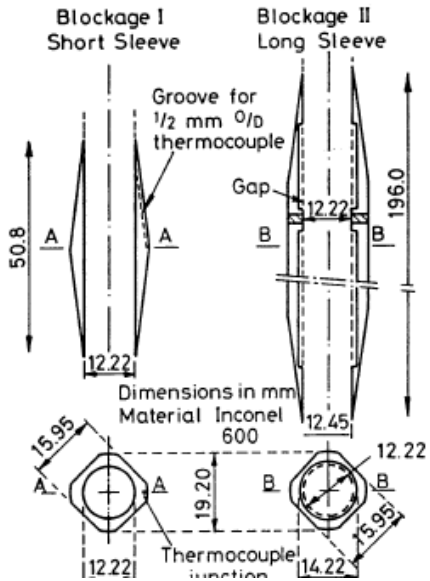


Run A1R030 (unblocked cluster)

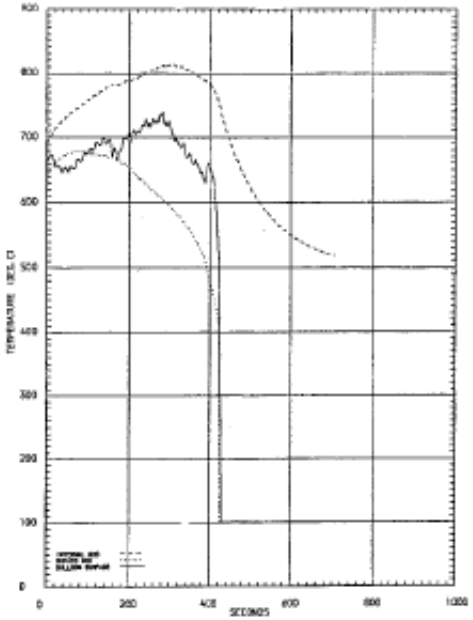
ACHILLES results showing temp. histories for blocked and unblocked bundles; the peak temp. on the rod balloons in run A2R038 was 1030°C at axial elevation 2.01m, in comparison with the peak temp. of 971°C at 2.13m axial elevation in run A1R030 in the unblocked cluster, thus the temp. penalty due to the blockage is 59°C.

UK bundle reflood experiments with fixed blockage (CEGB)

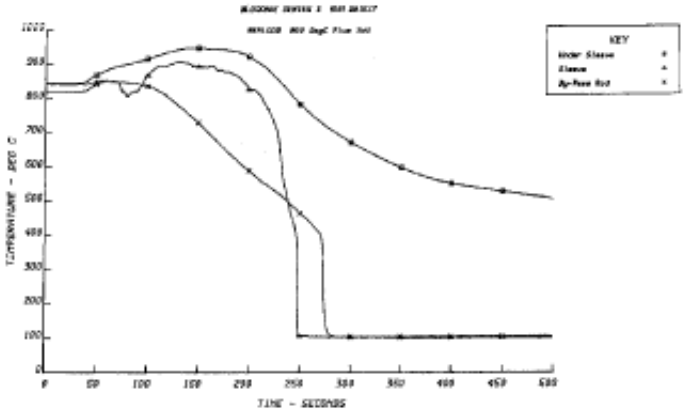
CEGB programme similar to THETIS, 44-rod bundle containing a 61% or 90% blockage on a central 4*4 group of rods; for the 90% blockage tests, one short blockage of 51 mm and two long blockages of 196 mm length were investigated



Shape of the cladding sleeves for the CEGB 90% blockage tests



THETIS Run T1R072 (90% blockage; 1.3 bar)



CEGB Blockage 3 - Run Q81KJ7

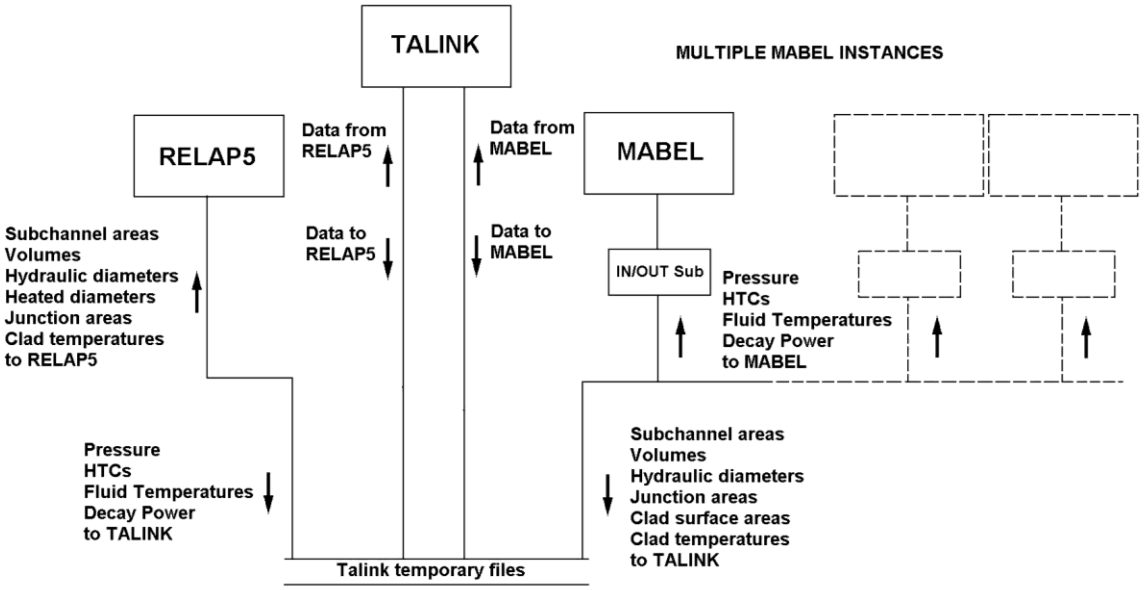
Blockage and by-pass temps.in comparative THETIS and CEGB tests; blockage sleeves rewet much earlier than the bypass rods in the CEGB long blockage 90% tests, but in THETIS they rewet at much the same time; also blockage rewetting takes place with an ascending quench front in the THETIS tests but with descending quench front in the CEGB equivalent; may be due to the shorter heated length in the CEGB case, and to the longer taper length in THETIS.

UKAEA code development on clad ballooning (1/3)

- Early version of the CANSWEL2 2-D (r-theta) clad ballooning code [Haste, July 1982] summarised in NEA 6846, the code was sponsored by the UK regulator and industry;
- CANSWEL included in the MABEL2 code [Bowring et al., 1982] that models the effects of ballooning on channel blockage, with thermal hydraulic feedback, coolant bypass and the effect of pellet stack eccentricity (or that of the electrical heating element in fuel rod simulators);
- Substantial developments since then, improved models for the effect of oxidation on creep including cracking and re-oxidation of the oxide film, different treatments of rod-to-rod mechanical interaction considering non-simultaneous burst of neighbours, superplastic creep including the effect of grain growth, choice of Zr alloy treated, choice of different rupture criteria (stress, strain, strain rate, damage fraction (development of NORA2, [S. Raff KfK 3184,1982]), damage fraction is preferred;
- Updated versions in MABEL2, also included in the BASIL clad ballooning code of CEGB (unpublished);
- Extensively validated, used in support of SXB safety studies, pre-test and post-test support to experiments such as NRU MT-3, Halden IFA-54X etc.;
- MABEL2 transferred to Imperial College, London, now called MATARE, includes multi-rod coupling.

UKAEA code development on clad ballooning (2/3)

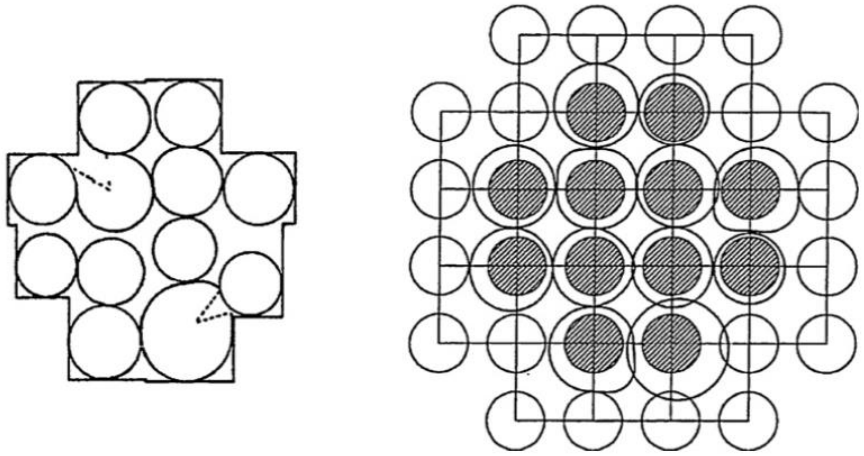
- UK ballooning code development now concentrated in the MATARE code, developed under the leadership of Imperial College, London [Ammirabile and Walker, 2014];
- It is a coupled thermal hydraulic and structural mechanics multi-rod model of reflood and clad ballooning;
- The mutual coupling of (multiple rod) structural mechanics to the transient two-phase thermal hydraulic model is achieved using the MABEL fuel rod analysis code coupled with the RELAP5 2-phase thermal hydraulic systems code (sponsored by the USNRC), with 6 balance equations, the mutual coupling being handled by the TALINK 'communications code';



Schematic of the coupling scheme in MATARE

UKAEA code development on clad ballooning (3/3)

- Code applications can take into account systematic and stochastic differences amongst rods (core-wide flux variation, local variations due to assembly edges or control rods, variations in fill-gas pressure, enrichments, coolant flow variations, manufacturing tolerances….), avoidance of the formation of large coherent blockages;
- Sample benefits from such analysis include: reduction of conservatism implicit in single-rod models which could relax operating envelope constraints, these are particularly important in context of the drive for higher burnups;
  - Better understanding is crucial to permit use of advanced, high ductility clad;
- The code has been used to analyse the in-reactor NRU MT- 3 clad ballooning experiment, with good overall results concerning axial rod profiles, with substantial incoherent ballooning found;
- Calculation of a generic PWR subassembly, with systematic and stochastic differences between rods incorporated, predicts absence of large regions of coherent ballooning,



Observed NRU MT-3 clad geometry, (left) and the MATARE calculation (right)

Use of Karlsruhe LOCA-related data in the UK (1/3)

- Separate-effect tests on creep of Zircaloy in steam by [Hofmann et al., KfK 3168, 1981] along with the NORA model of [Raff, KfK 3184, 1982, “REF 28”) were the basis of the clad deformation and rupture models in CANSWEL2;

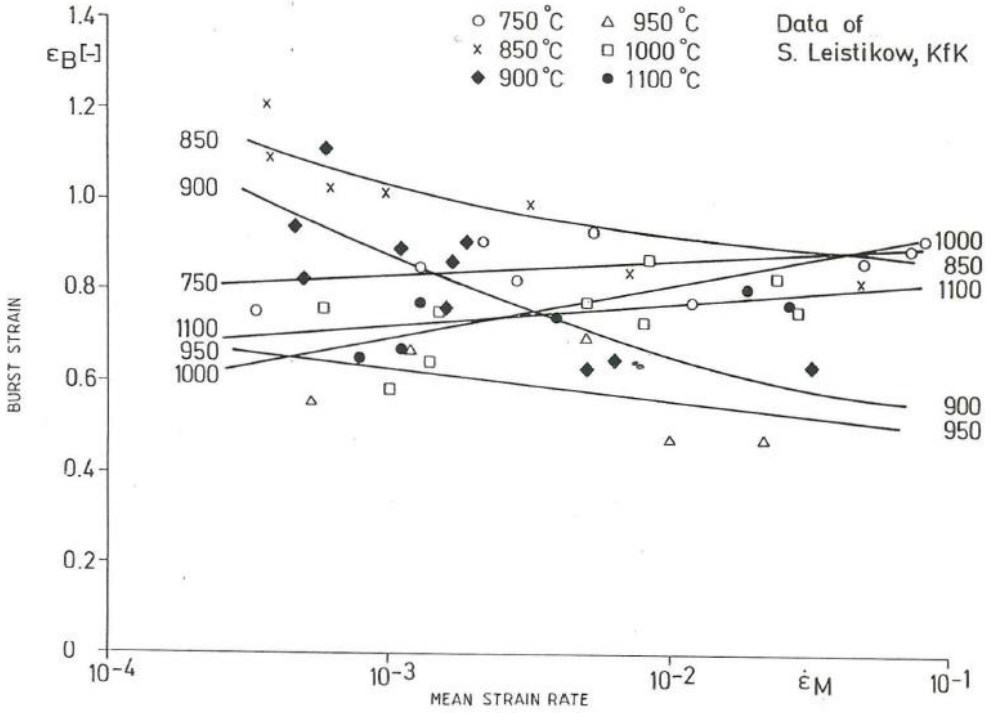


FIG. 5. DEPENDENCE OF CLADDING RUPTURE STRAIN ON STRAIN RATE, KfK INERT ATMOSPHERE TESTS 750 - 1100 °C (REF. 28)

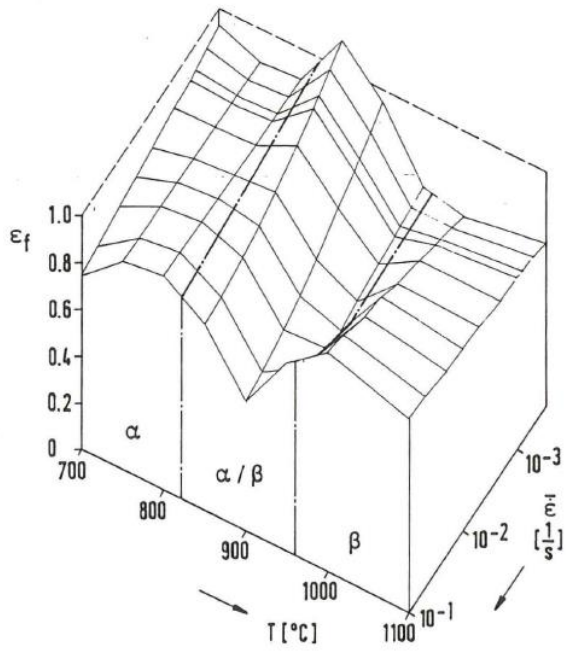
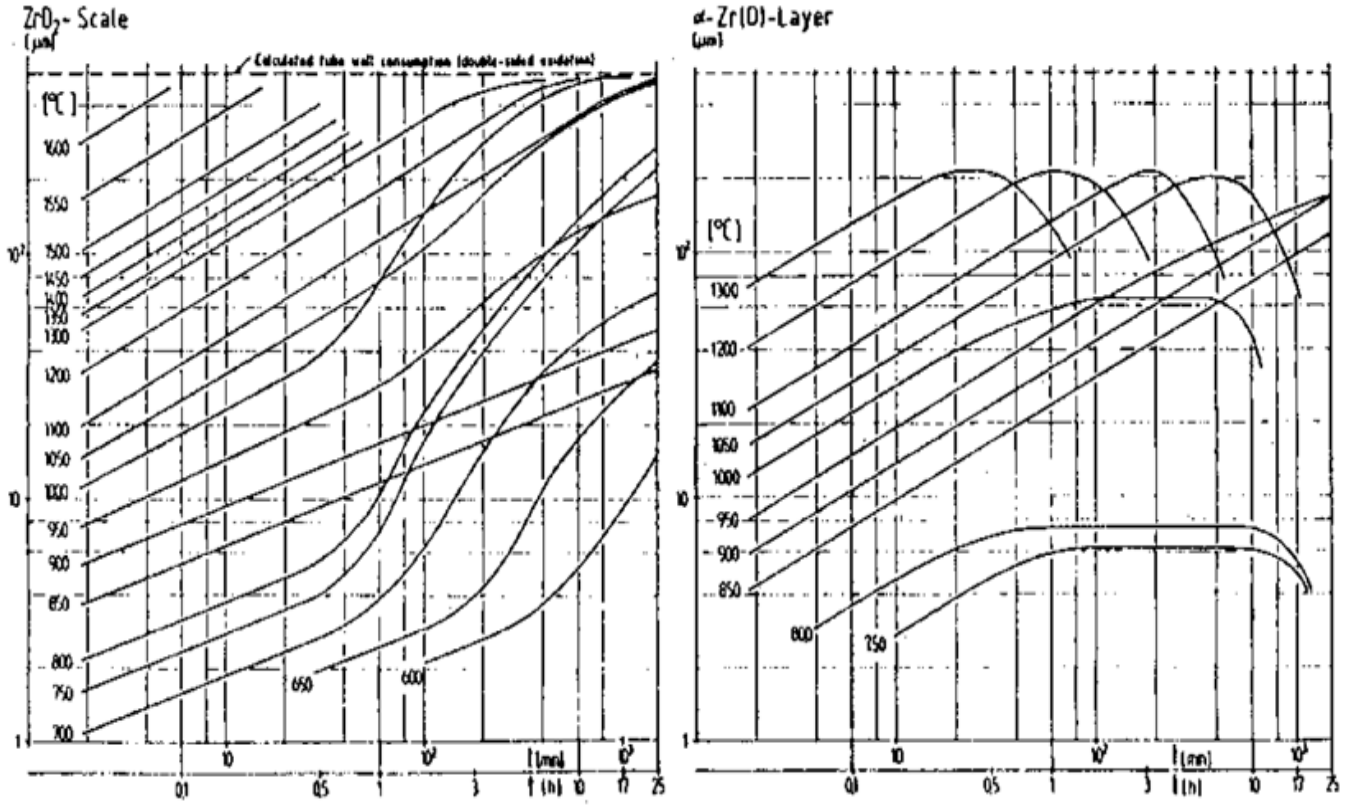


FIG. 6. RUPTURE STRAINS OF ISOTHERMAL TUBE BURST TESTS IN AN INERT ATMOSPHERE (REF. 28)

Use of Karlsruhe LOCA-related data in the UK (2/3)

- Separate-effect tests on oxidation of Zircaloy in steam were the basis of the cubic/parabolic oxidation model in CANSWEL2, notably the classic experiments of [Leistikow et al.. 1978] included in NEA 6846:

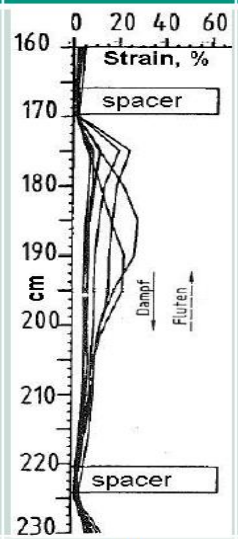


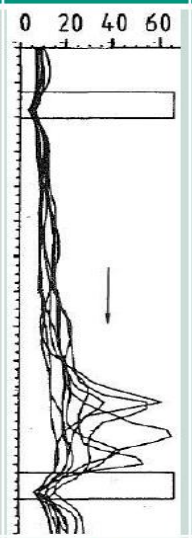
Use of Karlsruhe LOCA-related data in the UK (3/3)

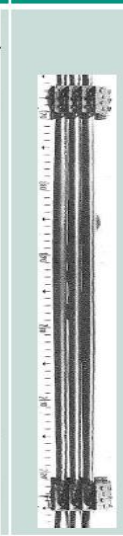
- REBEKA bundle tests were heavily used for MABEL code validation, and also by the industry (BART/TAPSWEL) e.g. REBEKA 6 (OECD/NEA/ISP-14) and REBEKA 7;

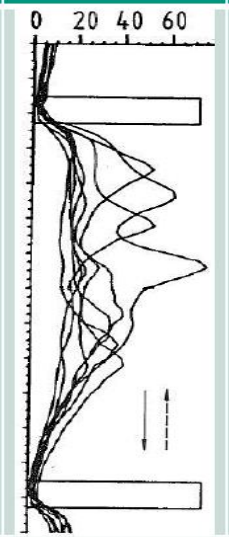
REBEKA test matrix

KIT Karlsruhe Institute of Technology

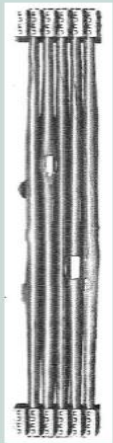


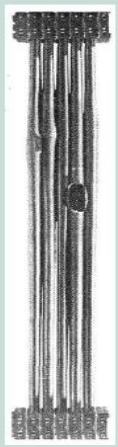


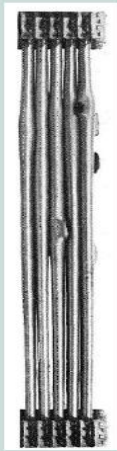












	REBEKA 1	REBEKA 2	REBEKA 3	REBEKA 4	REBEKA M	REBEKA 5	REBEKA 6	REBEKA 7
pressure, bar	60	55	51	53	70	68	62	57
burst T, °C	810	870	830	830	754	800	790	790
strain, %	28	54	44	46	63	49	42	55
blockage, %	25	60	52	55	84	52	60	66
burst region, mm		95	203	242	28	242	140	200

19.11.2013 J. Stuckert – LOCA overview KfK-FZK-KIT QWS-19, Karlsruhe 20 / 24 IAM Institute for Applied Materials

CONCLUSIONS

- This presentation summarises the very extensive research on LOCA carried out in the UK, well supported by the industry and regulator, much of it in support of the safety case for the Sizewell B PWR, a Westinghouse PWR of 4-loop design;
- This involved separate-effect and bundle tests on Zircaloy oxidation, creep, on rod-to-rod mechanical interaction, pellet stack relocation, and reflood tests on bundles with a fixed blockage;
- This knowledge, and also that gained through international collaboration, e.g. with France, Germany & the US, was encapsulated in the 2D (r,theta) CANSWEL/MABEL codes for calculating clad ballooning and channel blockage in postulated large break LOCA conditions, and extensively validated using an international database;
- Examples are given of the importance in this regard of data from KfK/FZK/KIT;
- This research is being continued in development of the MATARE code; with more detailed coupling between clad deformation and thermal hydraulics, which also takes into account the mechanical properties of modern binary alloys;
- New work identified here is being taken into account in an updated CSNI SOAR on fuel behaviour during loss of coolant accidents.

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SOARS

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THE STUDY OF THE CLUSTER OF FUEL-CONTAINING MATERIALS UNDER ROOM NO.305/2 OF "SHELTER" OBJECT

Roman Godun, ISP NPP of NAS of Ukraine, Lysogirska str. 12, Kyiv, Ukraine

Keywords: "Shelter" object, fuel-containing materials, re-criticality, nuclear safety's ensuring, experimental and computational research.

The experimental and computational research of fuel-containing materials inside "Shelter" object were performed by Institute for Safety Problems of NPP of NAS of Ukraine (formal scientific supervisor of works at New Safe Confinement - "Shelter" object during all stages of its transformation).

The temperature fields in under-reactor concrete slab showed existence and allocation of hidden FCM cluster, the direct access to which is absent. After the estimating of fuel's mass and cluster's volume, it was determined that the high mass fraction of nuclear fuel provides the possibility of existence of a critical composition. On June 1990 it was registered the neutron anomaly at the zone of FCM cluster. This anomaly within conservative approach to nuclear safety was interpreted as a re-criticality. The available long-term experimental data (FCM cluster's temperature and heavy precipitation in May-June 1990, dynamics of the NFD and temperature from 1988 till 2017, large share of porosity in structure and a waterproof ceramic crust at the boundary of cluster) indirectly confirm this assumption. Also it was performed a mathematical modeling (by solving of system of differential equations) of NFD dynamics during the neutron incident on June 1990. The received NFD dynamics correlate and practically coincide with experimental and thus the verification of this model was performed. With help of this mathematical model it was get the results and possible NFD dynamics during the "recurrent criticality", i.e. during the reducing the water portion in FCM cluster. A huge amount of calculations was performed by MCNP's code and with spectrum of initial data that does not contradict to available experimental data. In a result, it was obtained the dangerous range of main parameters.

The nuclear danger of this FCM cluster is determined by the probability of critical composition existence in over-wetting condition, which could be achieved within the "re-criticality" in 1990. All other detected FCM clusters inside the NSC-SO are nuclear-safe. The probable (during "recurrent criticality") NFD dynamics might be uncontrolled because of lack of effective monitoring and control of sub-criticality's level and positive temperature coefficients of reactivity that will accelerate the increase of NFD. The power of a neutron flash will depend on the positive reactivity's inputting and the retention time of the breeding system's integrity. Prior to the next stage of NSC-SO transformation (extraction of fissionable nuclear materials or filling of NSC-SO by concrete) the problem of direct access to medium of the FCM cluster under room No.305/2 must be solved. In addition, it is necessary to continue research to obtain the additional data (especially experimental), as well as organization of effective control of the level of sub-criticality in this FCM cluster and parallel development and implementation of means of preventive suppression of conditions that necessary for occurrence/development of self-sustained chain reaction. ISP NPP of NAS of Ukraine propose to specialists (who handle the corresponding/appropriate codes) to involve into this research process and to cooperate for best results' achieving.


Roman Gadun

Institute for Safety Problems of NPP of National Academy of Science of Ukraine

ISP of NPP
of NASU

The study of the cluster of fuel-containing materials (FCM) under room No.305/2 of "Shelter" object



Roman Godun
Head of Department
of Nuclear Materials Control

Karlsruhe, 2017

ISP of NPP of NASU

Institute for Safety Problems of NPP of National Academy of Science of Ukraine

ISP NPP of NASU is a formal scientific supervisor of the works, performed at New Safe Confinement "Arch" + "Shelter" object (NSC-SO) during all stages of its transformation and has all necessary permissions to provide the scientific and technical activity at NSC-SO

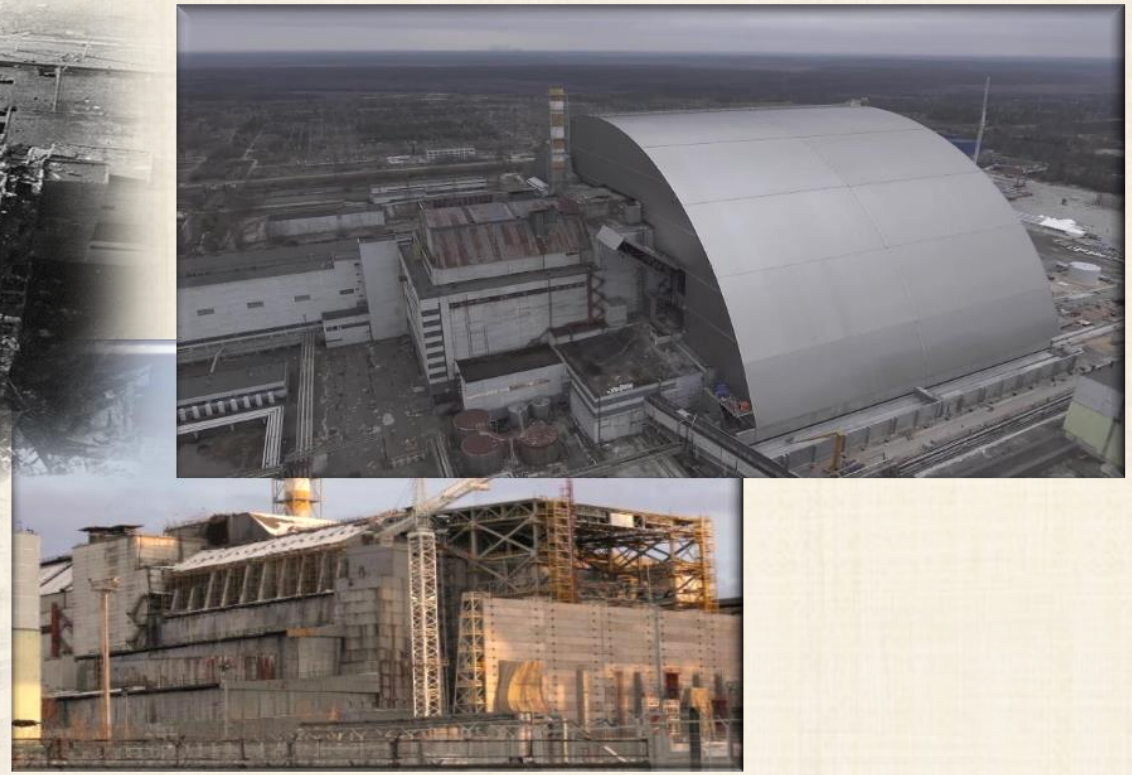
Available experimental facilities:

Laboratories of the 2nd and 3rd class with all necessary equipment (for conducting radiochemical studies, α,γ-spectrometry, mass spectrometry, etc.); systems of nuclear and radiation control, operating in NSC-SO; the llaboratory for manufacturing of the remotely controlled devices (capable to operate under the SO conditions) for scouting, monitoring and sampling of the FCM.

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The explosion at the Chernobyl NPP and the view on the destroyed 4th power unit



"Shelter" object and a New Safe Confinement "Arch"

Remote-controlled devices (developed by ISP of NPP of NASU) for scouting and monitoring of nuclear materials, as well as for the other tasks, performed at "Shelter" object











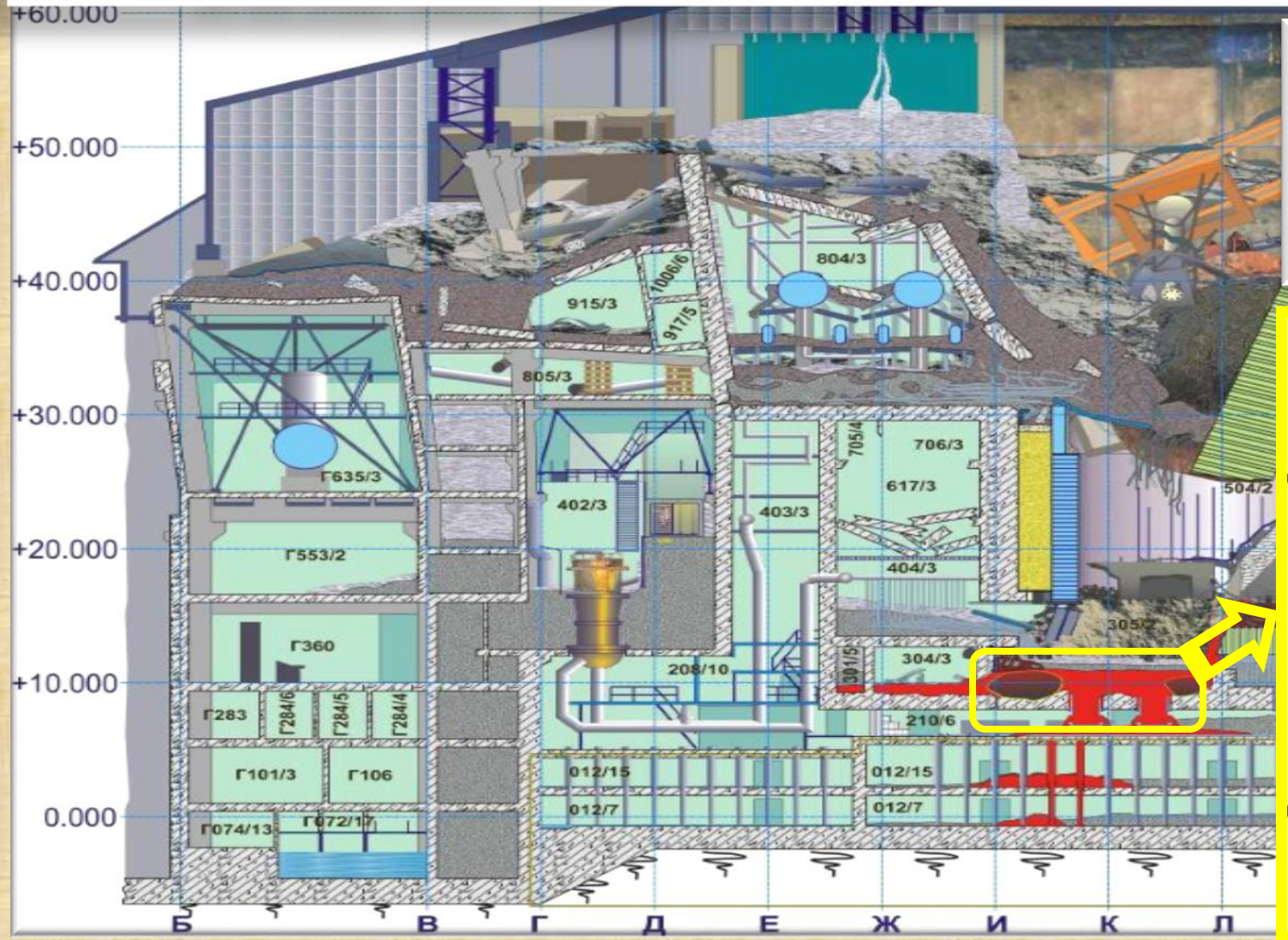




The Department of Nuclear Materials Control

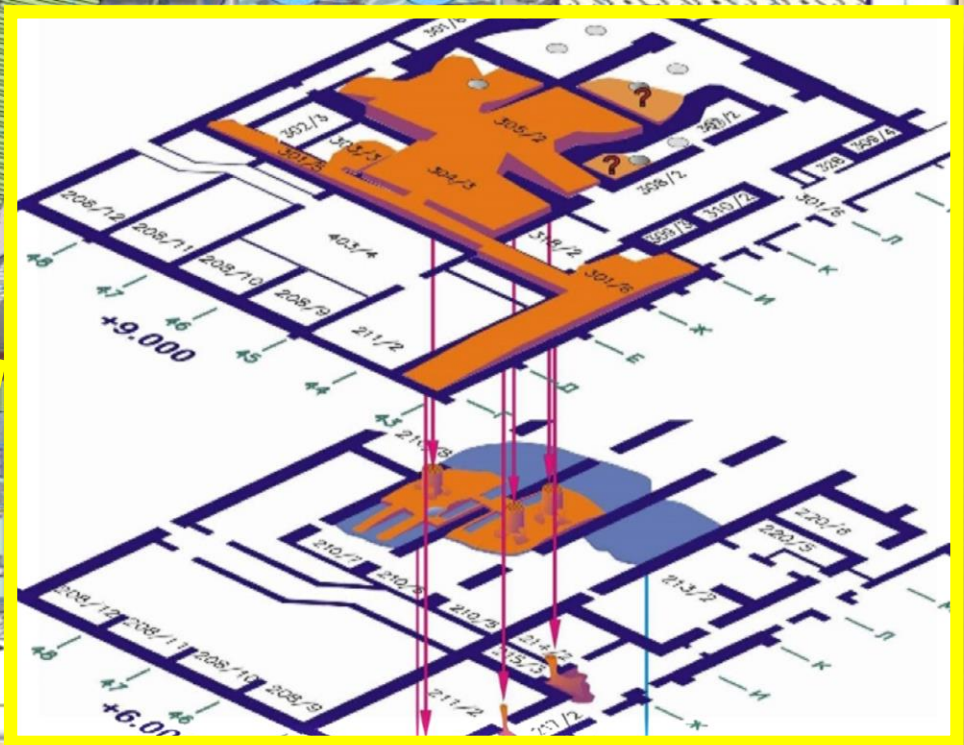
The main objective is the ensuring of nuclear safety and the control of all nuclear materials inside the New Safe Confinement "Arch"

The cross-section of the destroyed ChNPP unit 4 along the axis "46+2500" and the "spreading scheme" (in the under-reactor rooms) of lava-like products of interaction of molten corium with construction materials



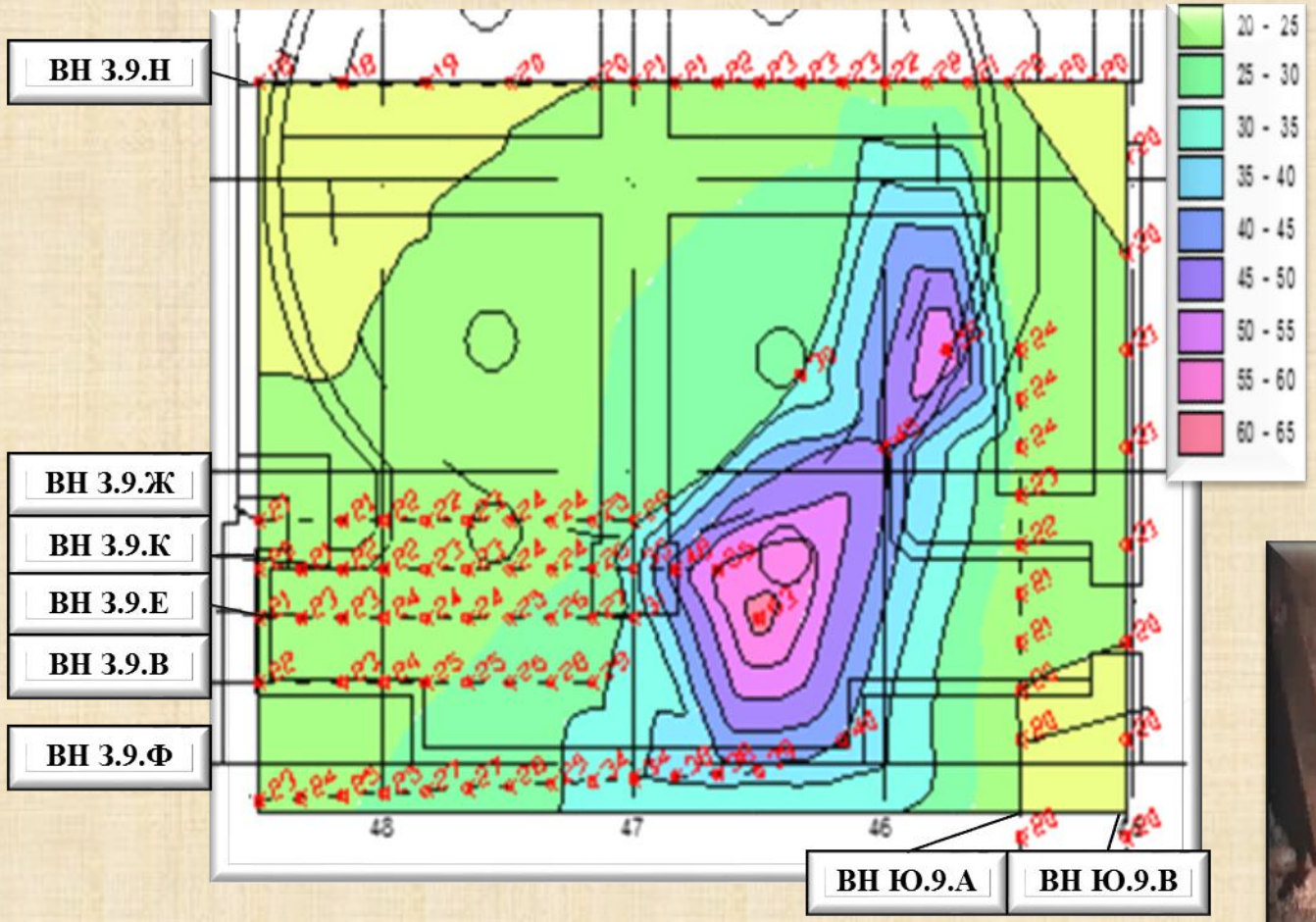
At least 95% of initial reactor loading (about 180 tons of irradiated nuclear fuel) is remain inside the NSC-SO.

77÷25 tons of nuclear fuel are located in room 305/2.



The localization of the FCM cluster (MCCI products) and results of its external observation

Measured temperature fields (November 2008) at the periphery of cluster of FCM and the placing of temperature detectors in under-reactor concrete slab (room 305/2)



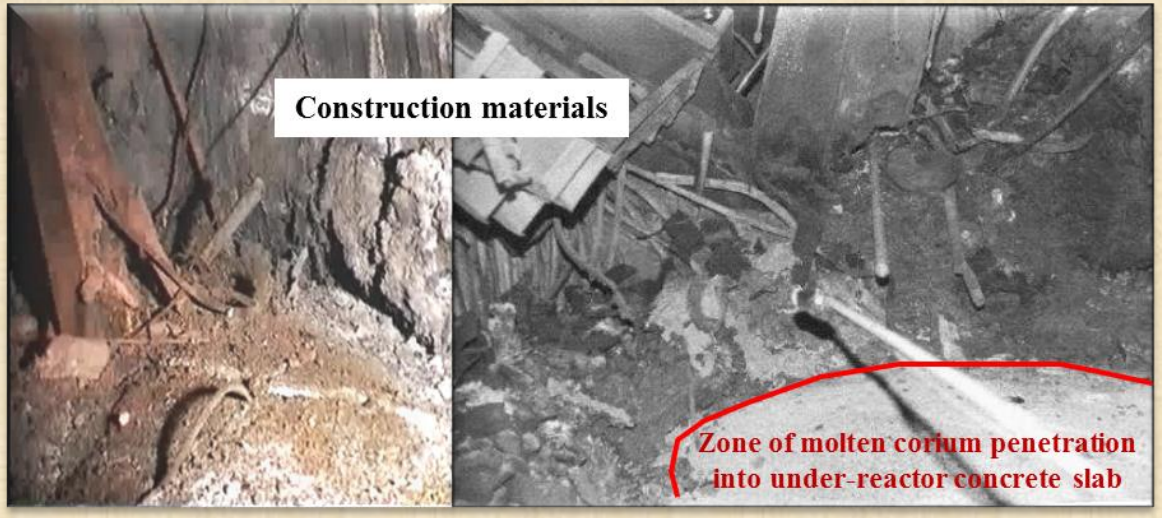
ВН 3.9.Ф – name of research borehole (BH)

– indication of temperature detector in BH

Special robot-scout, moving on rails, entered the room 305/2

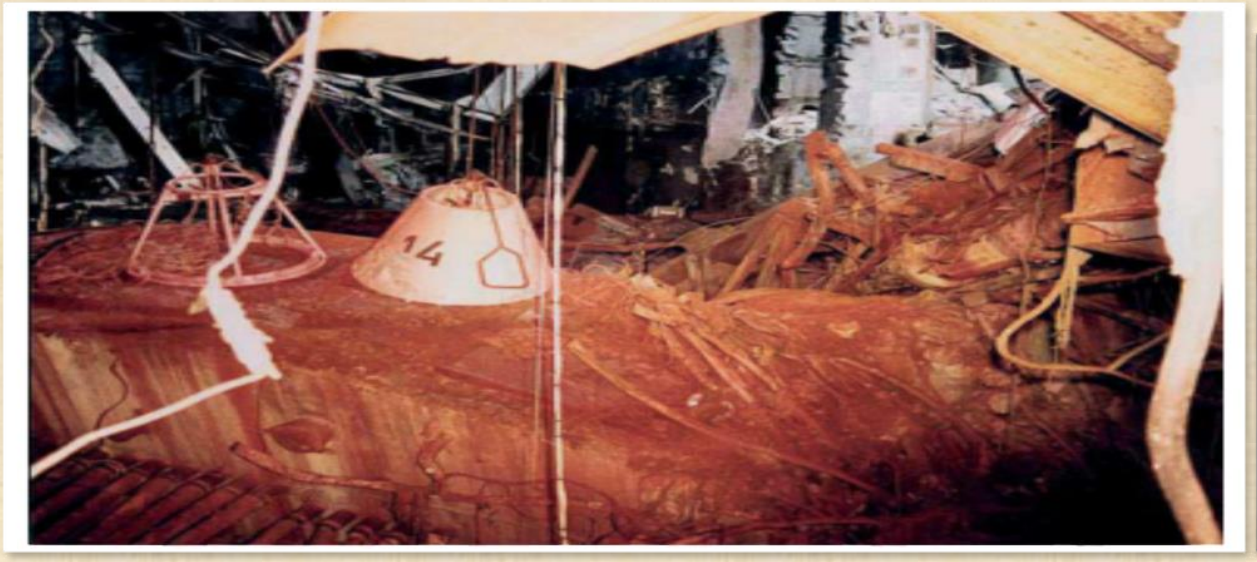


The results of an external observation of zone with MCCI products in concrete slab (bottom of the reactor cavity, room 305/2)

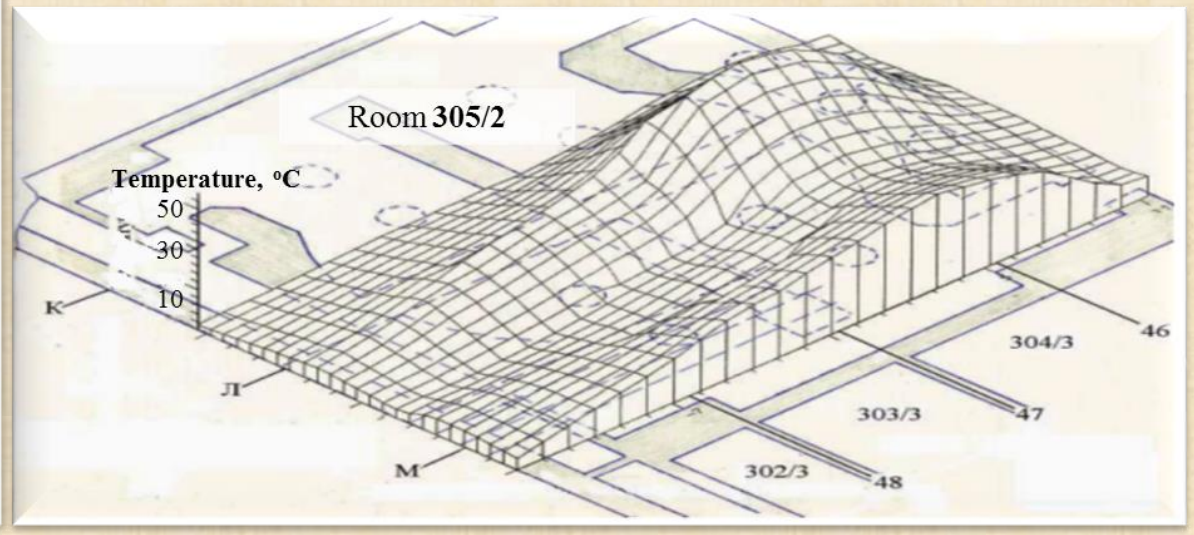


Thermometric measurements and the defining of the mass and volume of FCM cluster

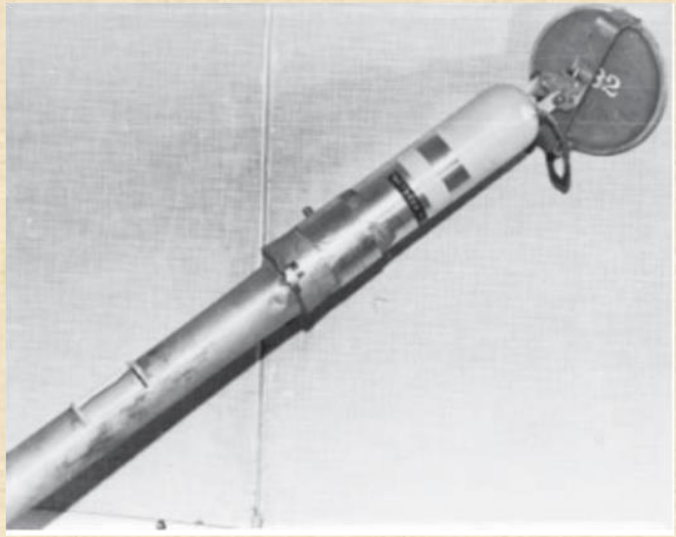
The "Bui“ device, installed in the collapse of the central hall of the ChNPP unit 4



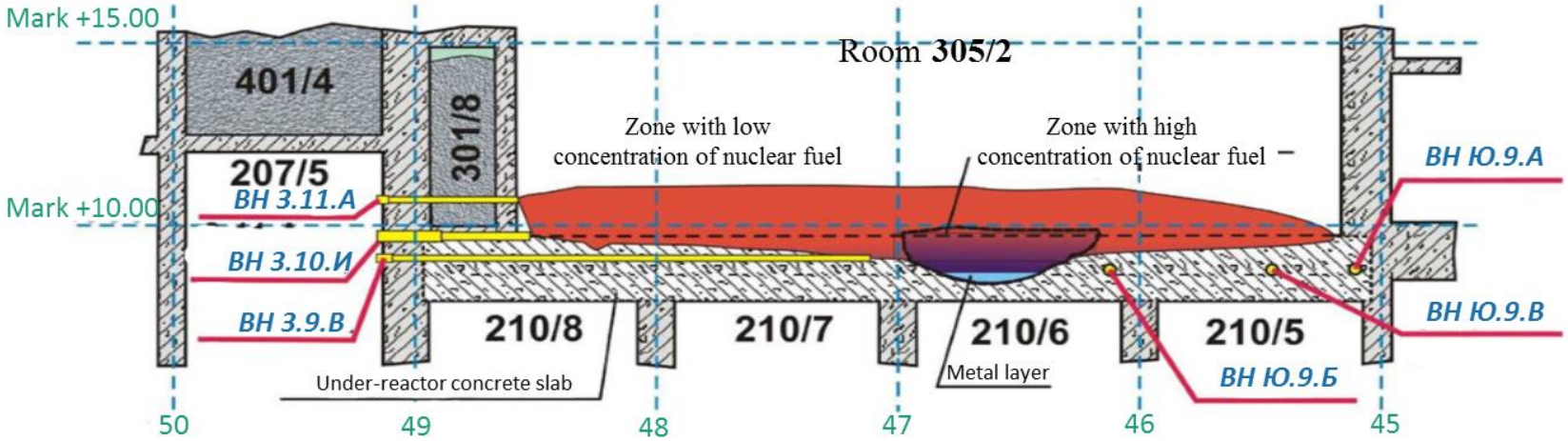
The temperature (results of measurements in research boreholes and by portable instruments) at zone of localization of nuclear dangerous cluster (room 305/2)



The thermometric probe, used in measurements of heat flux and temperature



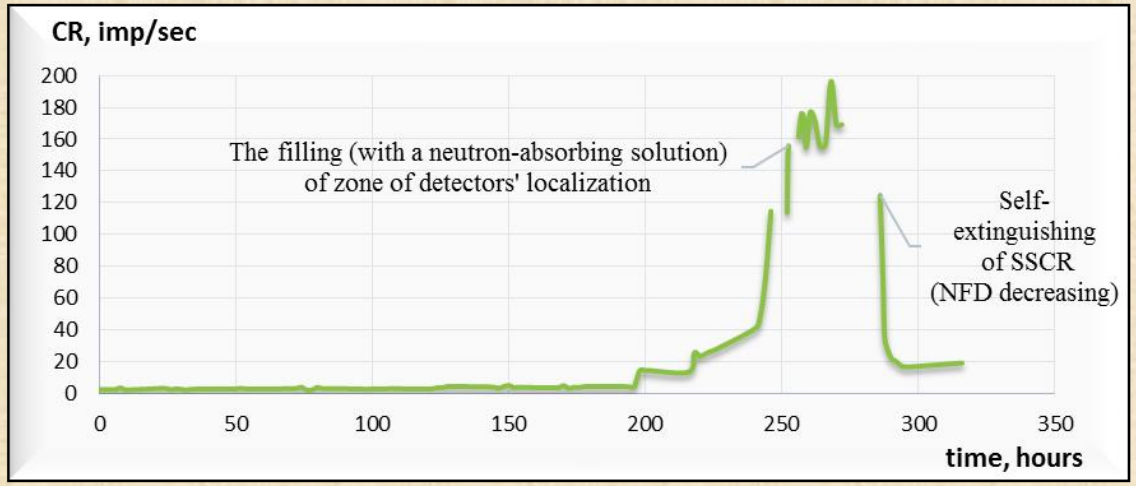
The determination of volume of the zone with high concentration of nuclear fuel, localized in under-reactor slab



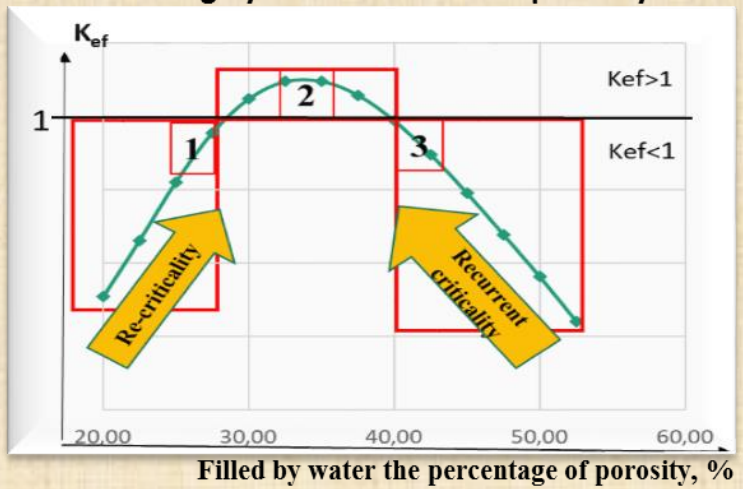
“50” – name of *construction axle* **“210/7”** – *name of room* **ВН** *3.9.В* – name of *research boreholes*

The neutron anomaly, registered at the periphery of zone of the MCCI product's localization

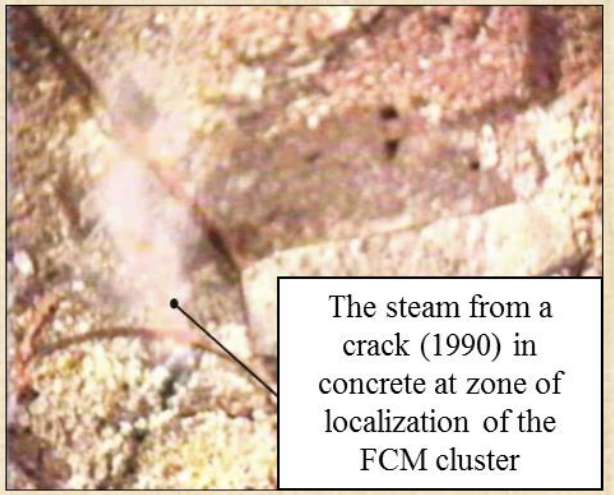
The dynamics of the counting rate (CR, impulses/sec) of neutron detectors during the emergence and self-extinguishing of the neutron anomaly in 1990



The dependence of the effective coefficient (Kef) on filling by water of available porosity



The evidence of a high temperature in 1990 at zone of localization of FCM



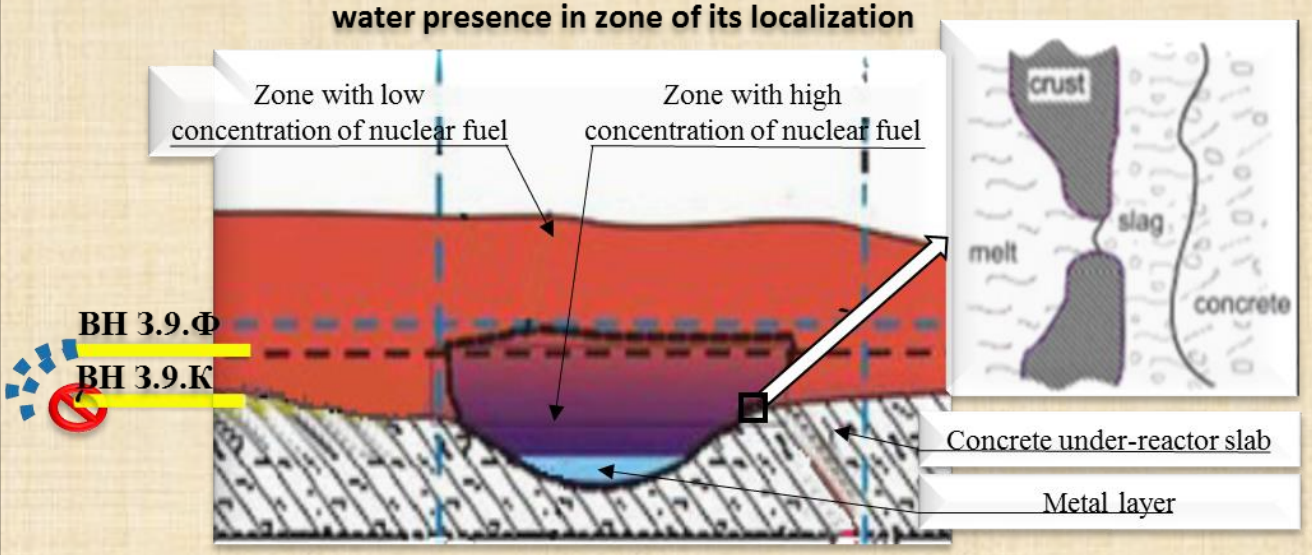
This neutron anomaly can be interpreted in three ways:

1. When water was entering into FCM cluster (which temperature was about 100 °C) the sub-criticality level was gradually decreased (accordingly there was the temperature increasing, steam generation, and pressure/stress increasing), that led to the destruction of system's structure and to eliminating of conditions, that were necessary for maintaining or developing of SSCR.

2. When water was entering into the FCM cluster with high temperature the system achieved the critical state (second zone) and then the self-sustained chain reaction was quenched due to over-moisten of breeding system (it reached the third zone). This processes were indicated by decreasing of count rate of neutron detectors This option of interpretation is conservative approach.

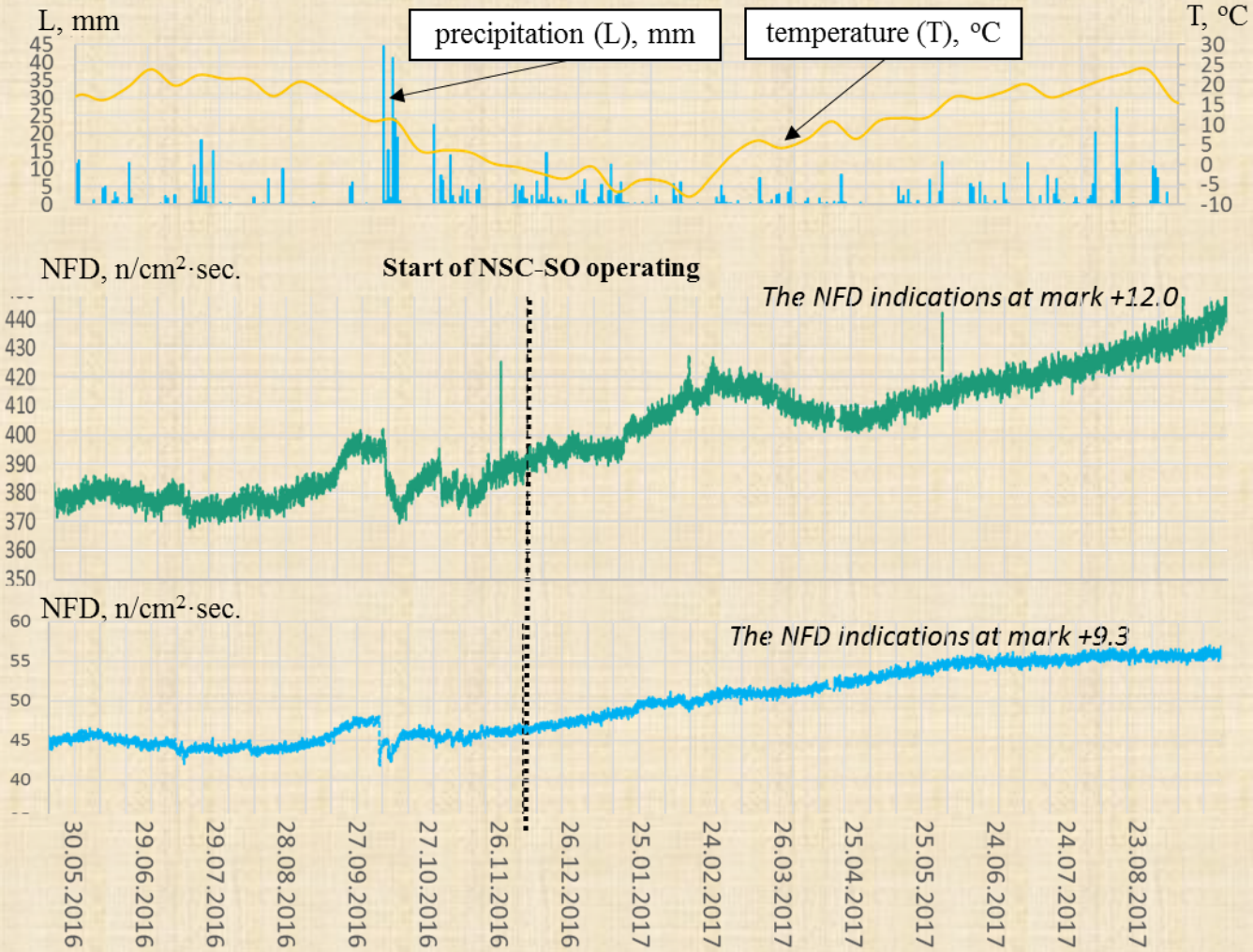
3. The measuring equipment failed, but this option was excluded after careful subsequent rechecking of the equipment.

The intended cross-section of FCM cluster and evidences of water presence in zone of its localization

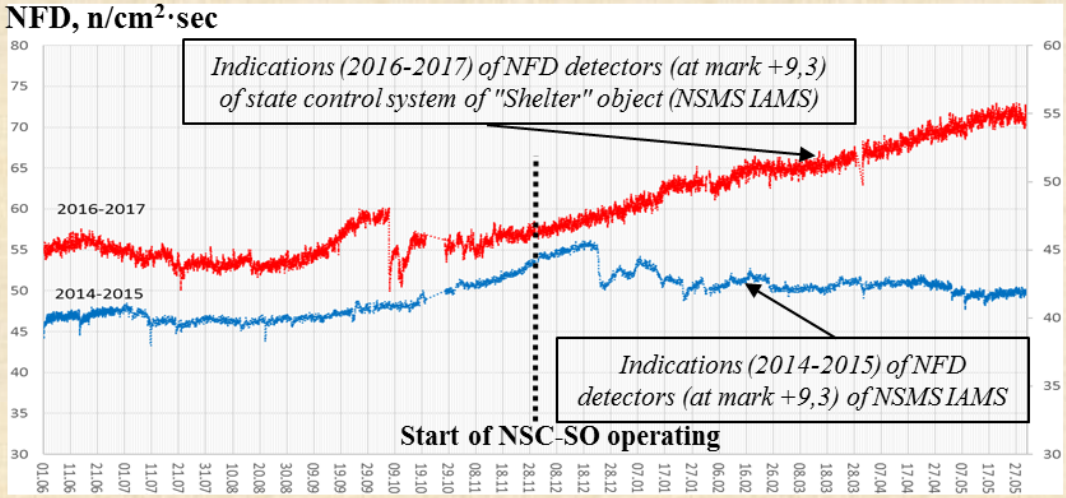


The neutron activity's dependence on water in zone of localization of FCM cluster

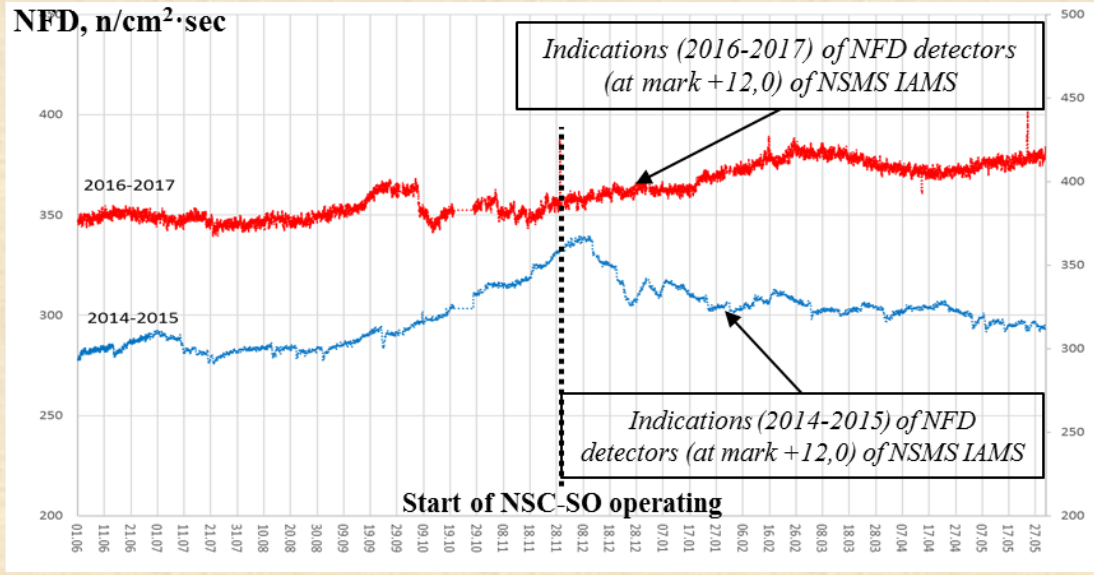
The dependence of neutron activity (at localization zone of FCM) on atmospheric precipitation and temperature inside the NSC-SO



The change (after the beginning of the NSC operation) of the dynamics of neutron activity at the mark. 9.3

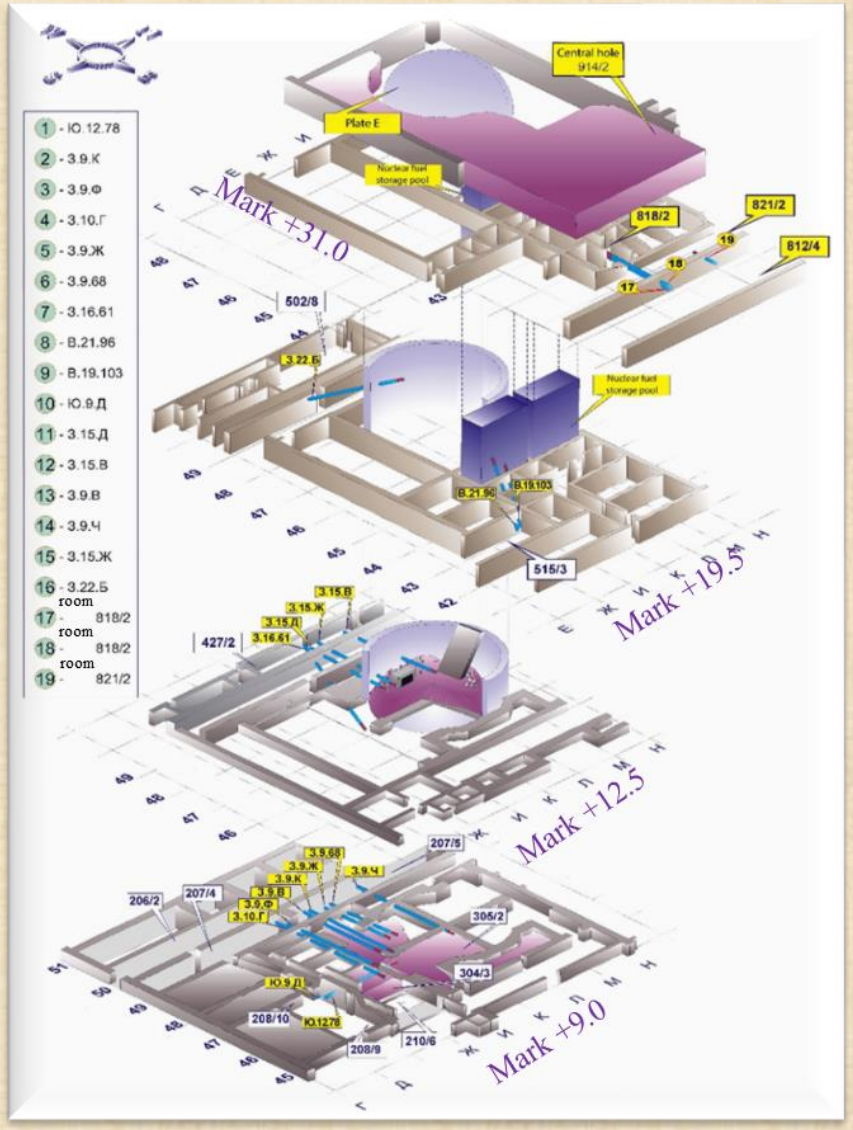


The change (after the beginning of the NSC operation) of the dynamics of neutron activity at the mark. 12.0



The state system (NSMS IAMS) of monitoring of nuclear safety at NSC-SO

Allocation of the research boreholes (RBH) with neutron detectors



– name of RBH (3.9.Ч)

– name of room (305/2)

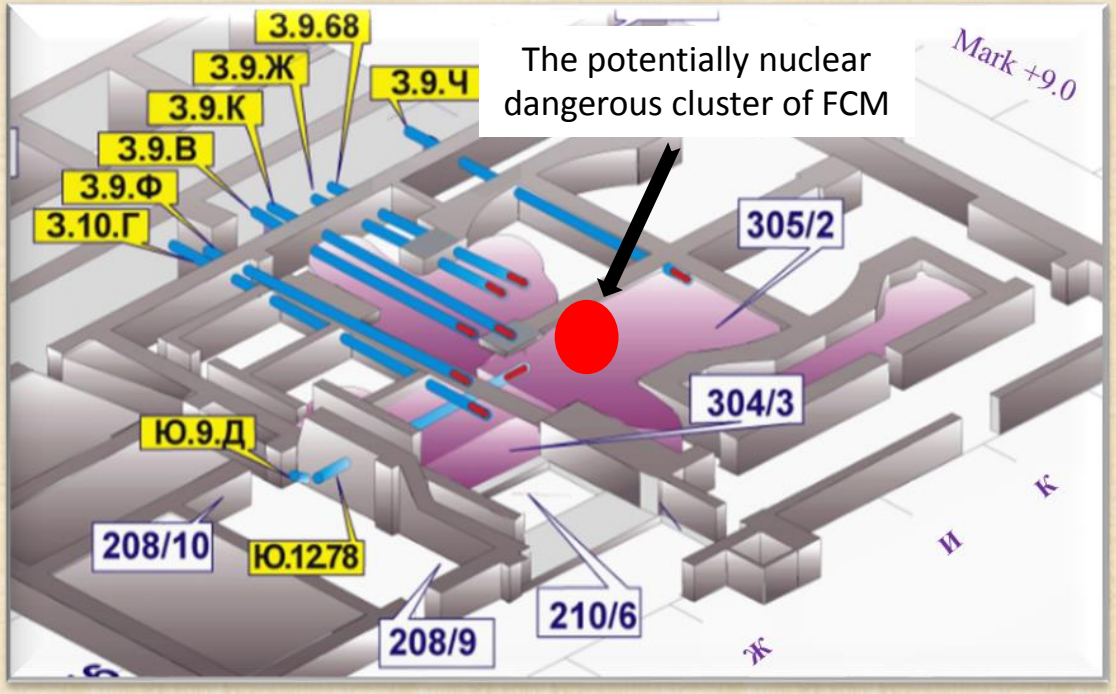
– construction axle "Ж"

– channel of NSMS IAMS

– neutron detectors in RBH

– pipes in RBH

Allocation of research boreholes with neutron detectors at elev. +9.0



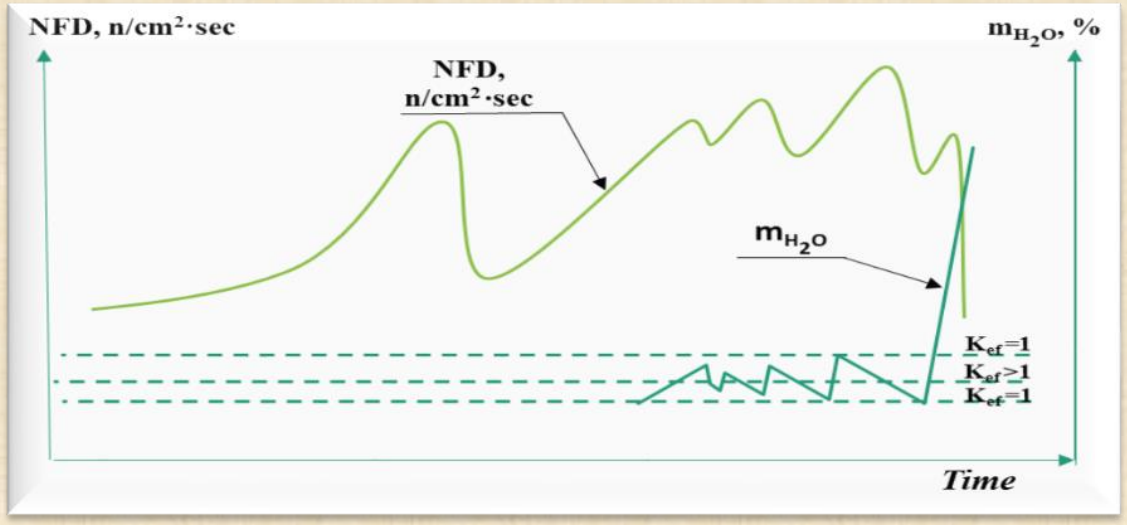
The implementation of procedure of the metrological certification of NSMS IAMS and drilling of RBH



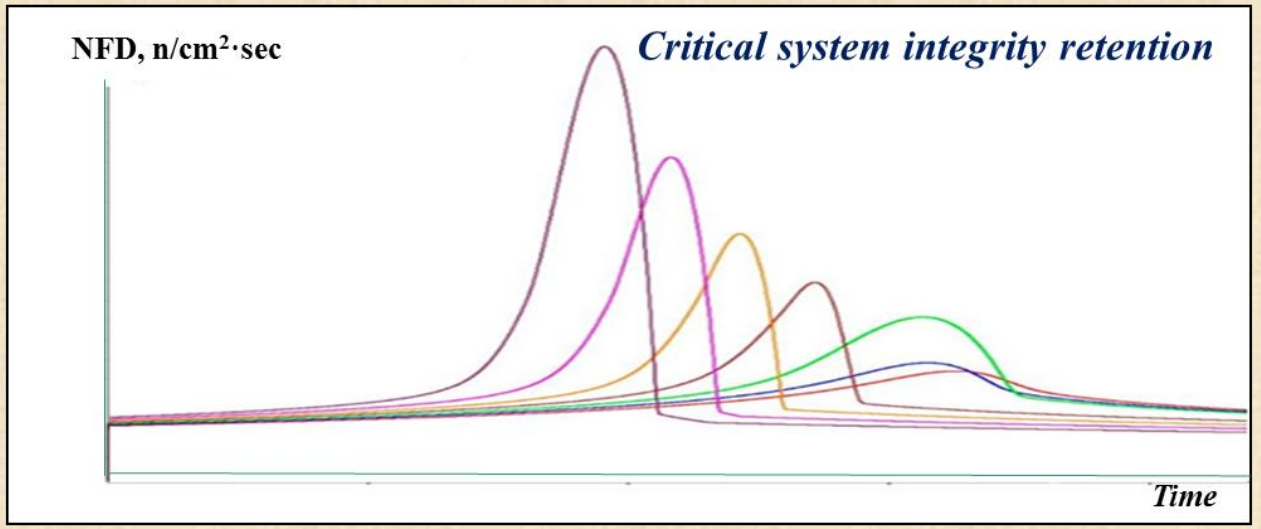


The modeling of NFD dynamics during the neutron incident (1990) and the "recurrent" criticality

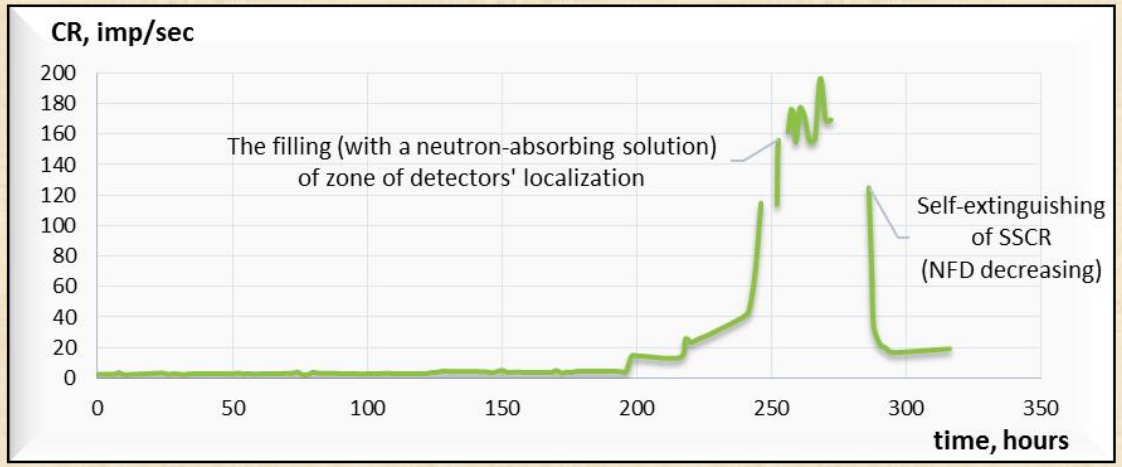
Results of NFD dynamics modeling during the critical composition's filling by water (model was verified by NFD dynamics of neutron anomaly in 1990)



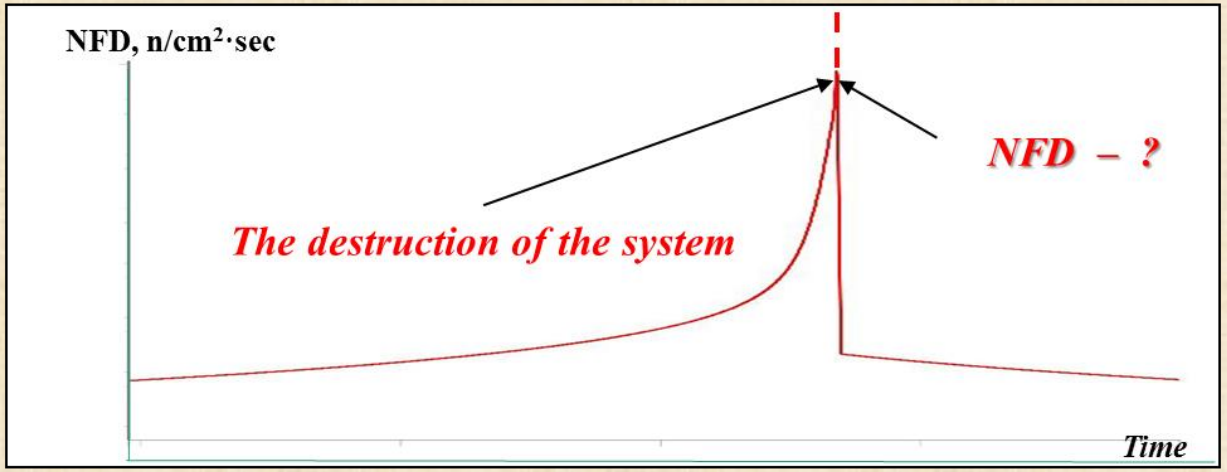
The results of mathematical modeling of NFD dynamics at "recurrent criticality" with condition of the system integrity maintaince



The dynamics of the counting rate (CR, impulses/sec) of neutron detectors during the emergence and self-extinguishing of the neutron anomaly in 1990



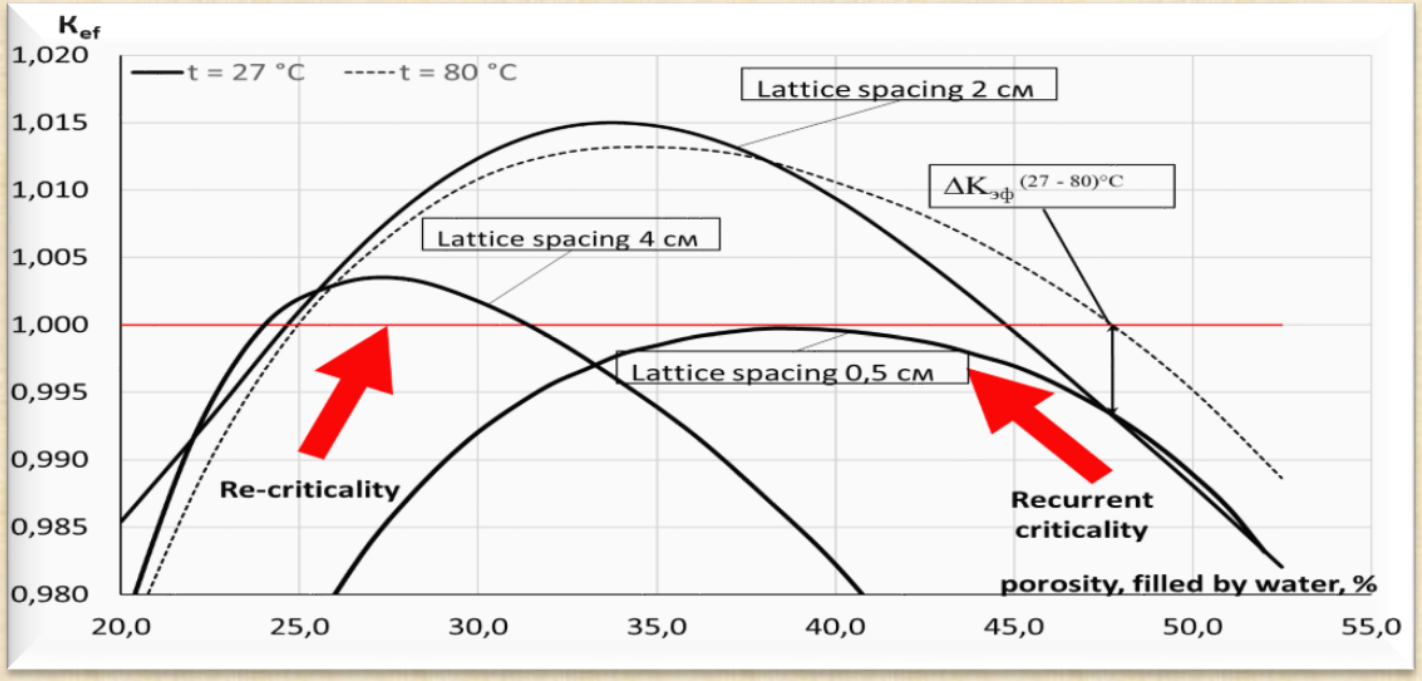
NFD dynamics at "recurrent criticality" and the system destruction



The NFD power during probable "recurrent criticality" will be depended on dynamics of positive reactivity's inputing into the system (mainly depends on the mechanism/dynamics of dehydration of the over-moist critical composition) and time of retention of critical system's integrity, actually sealed inside concrete slab

The parameters that provides the possibility of emergence of "re-" and "recurrent" criticalities (in FCM cluster under room 305/2) and influences on their dynamics

The example of K_{ef} dependence on the level of volumetric water content when the modeling's lattice spacing is 0.5, 2.0, 4.0 cm and FCM cluster's temperature is 27⁰C and 80⁰C



The variational calculations of neutron-physical parameters confirm the probability of existence of nuclear dangerous over-moist critical composition. In this case the stability of its sub-criticality is ensured by the presence of atmospheric, service and condensational water at "Shelter" object + New Safe Confinement.

In case of over-moist FCM cluster (under room 305/2) the main factors that may influence to the dynamics of "recurrent criticality" (the achieving of optimal uranium-water ratio of system during the decrease of water's proportion) are the velocity of water inputing, structure changing (changing the porosity's spectrum/range) and the increasing of the breeding system's temperature.

The calculations were performed by using MNCP code.

The evaluation of parameters of the potentially dangerous FCM cluster was carried out on the basis of computational modeling of critical assemblies with input data, that corresponds to the available experimental data on: the mass and volume of fuel-containing materials, the localization of FCM cluster, the structure of the environment, the neutron anomaly in 1990 and the long-term dynamics of NFD and temperature

The material parameters of presented model

Structure: *the heterogeneous with porosity up to 52% of the cluster volume and density of over-wetted assemblage equal to 4.3 g/cm³*

Composition: *52% U (1.15% ^{235}U enrichment), light nuclei (O, Mg, Al, Na) and other elements that correspond to the selected samples from FCM cluster's periphery*

The parameters range, that provides conditions for emergence of SSCR during the "recurrent criticality"

Parameters	The range of values
Mass of fuel (U), t	15 ÷ 20
Mass fraction of fuel,%	48 ÷ 60
Cavity volume in concrete slab, m³	14 ÷ 18
The maximum volume of FCM cluster, m³	9 ÷ 11,3
Density of FCM, g/cm³	6,3 ÷ 7,9
Density of FCM with considering of porosity, filled by water (up to 52%), g/cm3	3,6 ÷ 4,4
Heterogeneity of breeding environment, cm	0,5 ÷ 5

Current problems of nuclear safety ensuring inside the NSC-SO

The available results of experimental and computational researches show that there is possibility of emergence of self-sustained chain reaction in FCM cluster under room 305/2.

+

The effective monitoring of NFD at zone of localization of potentially nuclear dangerous cluster of FCM (in room 305/2) is provided by only <u>three</u> measuring channels of NSMS IAMS: NFD 01, NFD 03 and NFD 04

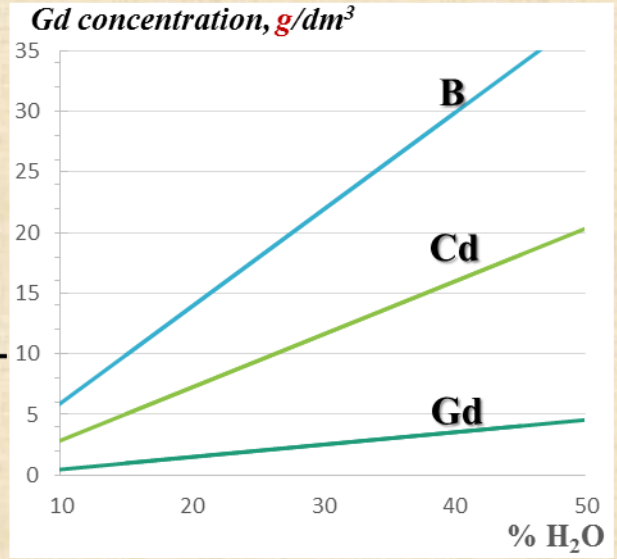
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The nearest (to FCM cluster) detector is located at a distance of about 2.5m (1.5m of which is concrete), therefore the registered NFD's power must be lower by several orders of magnitude then the real one.

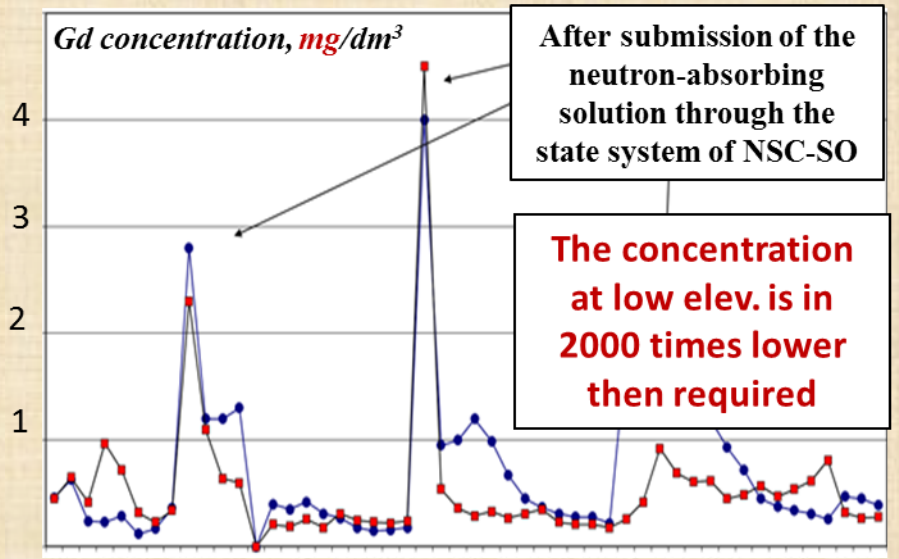
In addition, the knowledge regarding the spectrum of neutron radiation at the control point is insufficient~~ly~~

+

The minimal absorber concentration that required for effectively control of sub-criticality's level of FCM cluster



The concentration of neutron absorber at zone of MCCI products localization after filling it by gadolinium solution through/from dust suppression system of NSC-SO



Thus, there are no guarantees of non-occurence of SSCR at zone of localization of FCM cluster and there is the lack of effective monitoring; + currently at "Shelter" object the means of online active influence on its level of sub-criticality are not effective

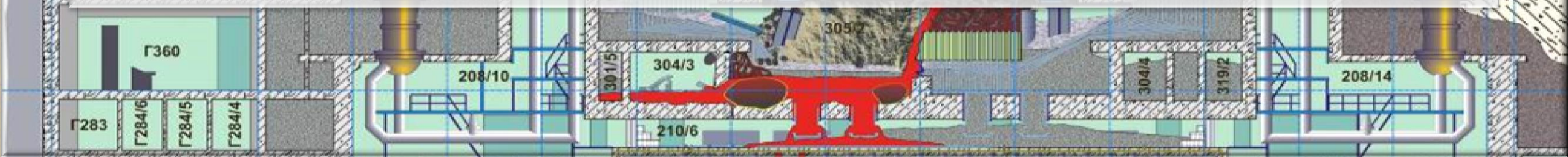
CONCLUSIONS

- ✓ 1. According to the currently available experimental and computational data there is a fact of existence of potentially nuclear-hazardous FCM cluster under NSC-SO's room No.305/2. Nuclear danger of this cluster is determined by the probability of critical composition existence in over-wetting condition, which could be achieved within the "re-criticality" in 1990. All other detected FCM clusters inside the NSC-SO are nuclear-safe.

- ✓ 2. The probable (during "recurrent criticality") NFD dynamics might be uncontrolled because of lack of effective monitoring and control of sub-criticality's level and positive temperature coefficients of reactivity that will accelerate the increase of NFD. The power of a neutron flash will depend on the positive reactivity's inputting and the retention time of the breeding system's integrity.

- ✓ 3. Prior to the next stage of NSC-SO transformation (extraction of fissionable nuclear materials or filling of NSC-SO by concrete) the problem of direct access to medium of the FCM cluster under room No.305/2 must be solved and, accordingly, the level of its nuclear danger will be determined experimentally

- ✓ 4. It is necessary to continue research to obtain the additional data (especially experimental), as well as organization of effective control of the level of sub-criticality in this FCM cluster and parallel development and implementation of means of preventive suppression of conditions that necessary for SSCR's occurrence.



The activities for reducing/elimination the risks, associated with the decreasing of water percentage in FCM cluster under room 305/2

- Reconsideration of effectiveness of the NSC-SO control systems and the development of measures for their modernization, as well as the development of the rapid supply of water or water solutions of neutron absorbers directly to the accessible periphery of the FCM localization's zone.

- Creation of a backup system of monitoring of neutron activity and temperature at zone of the FCM cluster's localization.

- Development of procedures for expert identification of the anomalies of neutron activity and temperature that are associated with dangerous changes of sub-criticality level in FCM cluster under room 305/2 inside NSC-SO.

- The reducing the uncertainties (associated with the risks of the SSCR emergence) by providing of additional computational and experimental researches, namely: the evaluation of strength characteristics and thus the retention's time (prior to destruction) of the breeding system's integrity; the correcting of material model of FCM cluster on the basis of domain version of scenario of the critical composition formation and, accordingly to this model, the revaluation of conditions and consequences of emergency of the self-sustained chain reaction; the modeling of heat-hydraulic processes of the decreasing of mass fraction

New OECD-NEA projects in support of Fukushima Dai-ichi site activities

By T. Washiya, M. Kurata, A. Nakayoshi & D. Bottomley,

JAEA- CLADS Laboratory, Tomioka, Japan

ABSTRACT

6 years after the great eastern Tsunami and the Fukushima F1 reactor damage in March 2011, longer term, planning for decommissioning and research into the accident are starting.

This presentation gives the background to projects being sponsored by JAEA at the Nuclear Energy agency at the OECD. The aim is to research the condition of the units and containment buildings of Fukushima F1, in parallel with the exploratory decommissioning work underway at these sites and to plan the longer-term research into the fuel debris analysis and its extraction.

These are the **Pre-ADES** project (**Preparatory study on Analysis of Fuel DebriS**): proposed to NEA/CSNI from Japan, and the **TCOFF** (**T**hermodynamic **C**haracterization **O**f **F**uel debris and **F**P-behavior). **Pre-ADES** examines the debris analysis and developing a reasonable methodology on defueling of 1F. It will initiate sampling for proposing an international R&D project on fuel debris **TCOFF** aims for enlargement/improvement of thermodynamic databases for core materials under various conditions by a) modelling studies of relevant TD quantities and b) by selected experimental studies of selected nuclear materials.


Karlsruher Institut für

New OECD-NEA projects in support of Fukushima Dai-ichi site activities

T. Washiya, M. Kurata, A. Nakayoshi & D. Bottomley,
JAEA- CLADS Laboratory, Tomioka, Japan

Outline of Presentation

1) Ongoing OECD/NEA Projects, relating to 1F
2) New proposal from Japan, PreADES to NEA/CSNI
3) New proposal from Japan, TCOFF to NEA/NSC
4) Role of JAEA/CLADS
5) Future project outlook & Concluding Remarks

Fukushima Dai-ichi-related NEA/CSNI Projects

CSNI projects (issues directly related to 1F are discussed in CSNI)

for improving safety knowledge and evaluation and achieving safe and timely decommissioning in Fukushima-Daiichi (1F)

- **Pre-ADES** (proposed to NEA/CSNI from Japan)

  Discussion on requirement to debris analysis, international R&D framework on fuel debris of 1F, and reasonable methodology on major issues regarding defueling and sampling

- **ARC-F** (under discussion)

  Discussion and estimation using existing information from examinations at 1F and results from Japanese R&D to analyze accident progression and status inside 1F, identification/prioritization of lacking information, and propose of future projects

- **BSAF** (phase-2 is ongoing)

  Benchmark calculation and discussion to improve severe accident codes and to analyze the accident progression and current status inside 1F

[For reference]

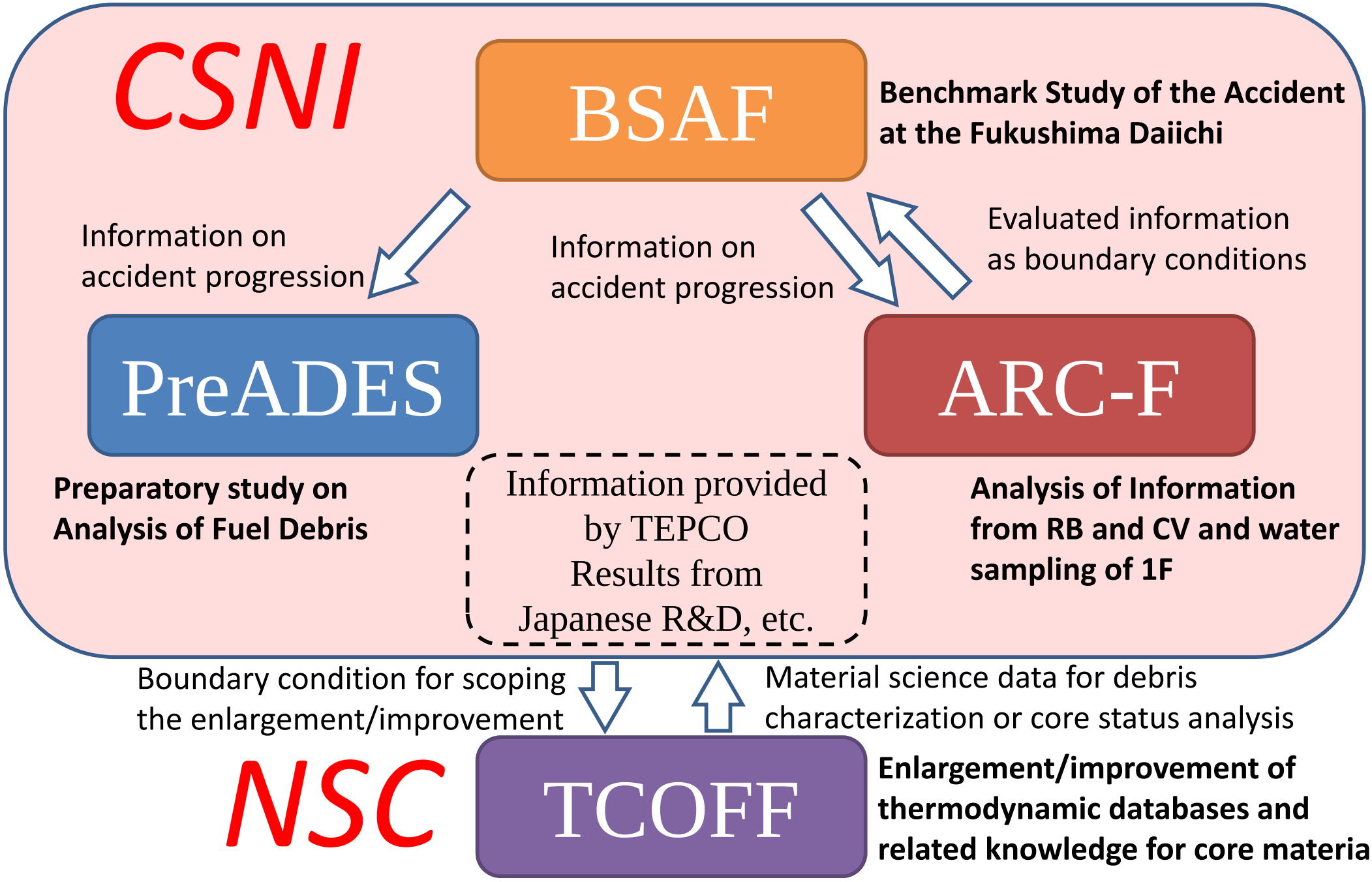
NSC project

for updating fundamental science knowledge. The outputs could be available to evaluation on melt progression, FP-behavior, and debris characterization.

- **TCOFF** (launched June 2017)

  Enlargement/improvement of thermodynamic databases for core materials under various conditions by analytical and experimental studies

Relation among the Projects



OECD-NEA/CSNI International Projects relating to Fukushima Dai-ichi Nuclear Power Station Accident *

1) An international modelling study was first set up

BSAF - Benchmark Study of the Accident at the Fukushima Daiichi Nuclear Power Station Project

Phase 1 (2012/15) Thermal-hydraulics in reactor pressure vessels (RPVs) and primary containment vessels (PCVs) for the first six days of the accident

Phase 2 (2015/18) Fission product behaviour outside PCVs, for upto ~3 weeks after the earthquake

(Op. agent : Inst. App. Ener. (IAE) Japan, supported by JAEA (inc. CLADS), CRIEPI, TEPCO + 11 countries

2) Then an NEA - CSNI committee senior expert group was set-up

SAREF – Senior Expert Group on Severe Accident Opportunities Post-Fukushima Research & Evaluation for Fukushima was set up (Jun 2013) to address safety research knowledge gaps (near–term & far-term) related to the Fukushima Daiichi nuclear accident.

3) From the SAREF deliberations both *near-* & *far-term* projects were proposed (2016-17): Pre-ADES, ARC-F

* Country members for OECD-NEA projects are usually represented by national research institutes

SAREF: Near-term Project Proposals (SEG Report)

Near-term project proposals can start in the *next years (2017 onwards* as a preparatory phase:

- Collect/analyze basic (& existing) information and update "long-term" research activities

- Maintain information channel between CSNI and Japanese organizations, and

- Check/test the feasibility of transportation, analyses, etc.

Proposals

1. Preparatory study on analysis of fuel debris (Pre-ADES)

- Review of fuel debris characterization methods (analytical, experimental)

- Radiation dose & exposure evaluation methods during fuel debris retrieval operations

- Discussion on possible future R&D topics with fuel debris samples

2. Examinations inside RBs and CVs and water sampling (ARC-F)

- Visual inspection inside RBs and CVs

- Water sampling inside TBs, RBs and CVs, and analyses

- Discussion on future long-term research activities

Pre-ADES-Outline-

Preparatory study on Analysis of fuel DEbriS (PreADES)

SAREF Near-term Projects: Program Overview

Collaborative Laboratories for Advanced Decommissioning Science

First Steps/Meetings of Near Term Projects

Preparatory Tech. Meeting 11 January 2017 in Paris

- Scope of work on examinations inside Reactor Basements (RB) and Containment Vessels (CV) and water sampling (**ARC-F**)
- Scope of work on preparatory study on analysis of fuel debris (**PreADES**)

Preliminary Meeting for PreADES

5 July , (Wed) , in Fukushima

-Technical Site Tour for Fukushima Daiichi NPS

6 to 7 July ’17, in Fukushima

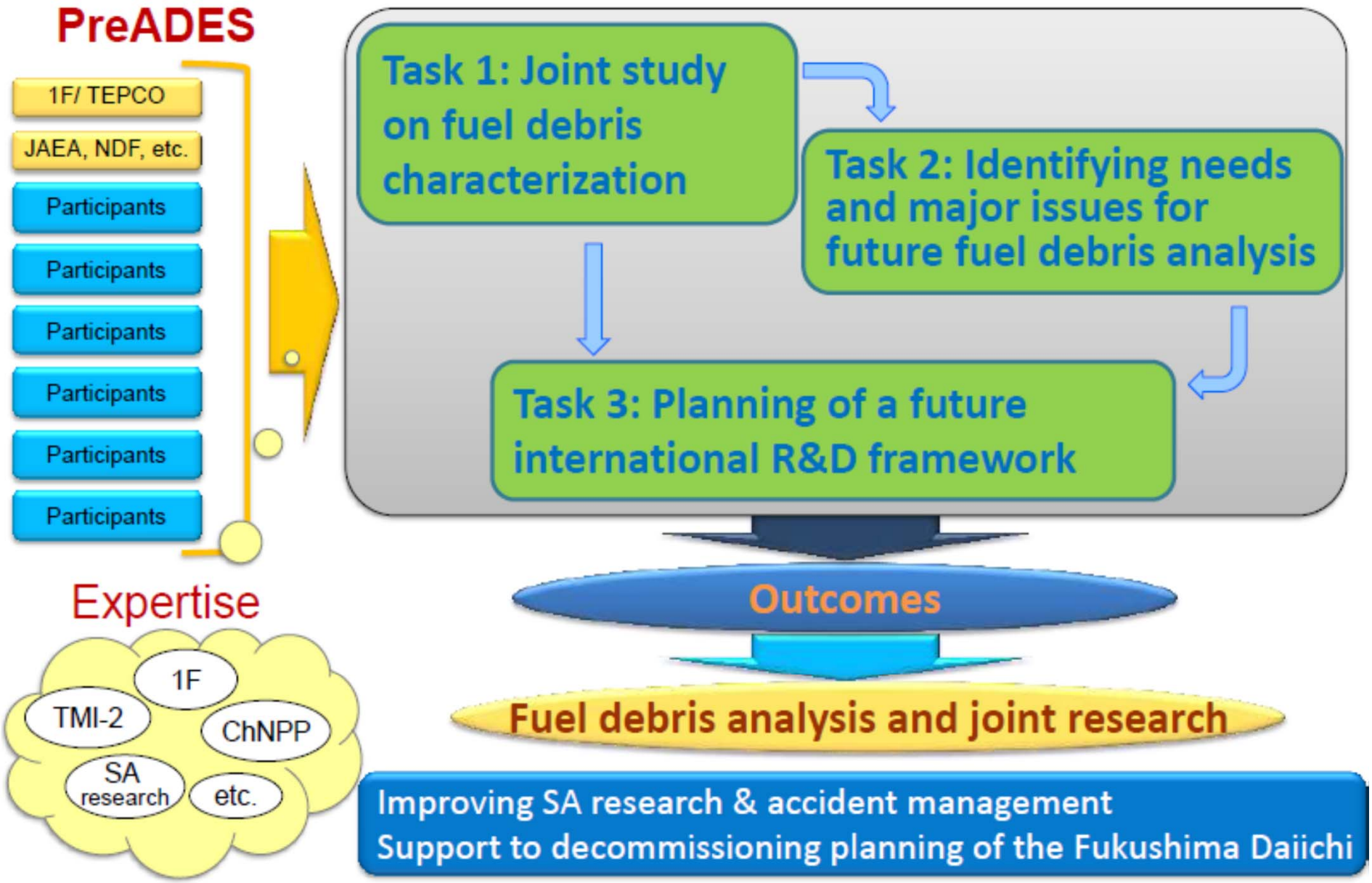
-Preliminary meeting on PreADES, modified proposal was explained by JAEA and discussed;

- Detailed work plan of each Task, and way of proceeding
- member presentations on relevant issues

10 to 11 July ‘17, in Tokyo

- SAREF/PreADES technical meeting and a Joint Session with BSAF-2 Program Review Group
-

Preliminary Meeting for PreADES
: Outcome Summary



Preliminary Meeting for PreADES : Outcome Summary

Collaborative Laboratories for Advanced Decommissioning Science

Task 1: Joint study on fuel debris characterization

- (1-1) Fuel debris characterization methods
- (1-2) Estimated properties of fuel debris

Task 2: Identifying needs and major issues for future fuel debris analysis

- (2-1) Needs for fuel debris analysis
- (2-2) Experimental/analytical techniques and demands in hot-testing facilities
- (2-3) Study for major issues on future debris analysis

Task 3: Planning of a future international R&D framework

- (3-1) Develop a plan of a future R&D framework using actual fuel debris sampled from Fukushima Daiichi NPPs

PreADES: Information flow between organizations - RSAD Table

- During PreADES project, timely interaction and advice are of great help to 1F decommissioning work.
- This project will be an opportunity to discuss with IRID, TEPCO, NDF etc. as subjects/ problems in 1F decommissioning arise

RSAD Table includes detailed work steps for each task and expected outcome as support to organizations above in 1F decommissioning work.

❖ Recommendations of Sampling and Analysis of fuel Debris (RSAD)

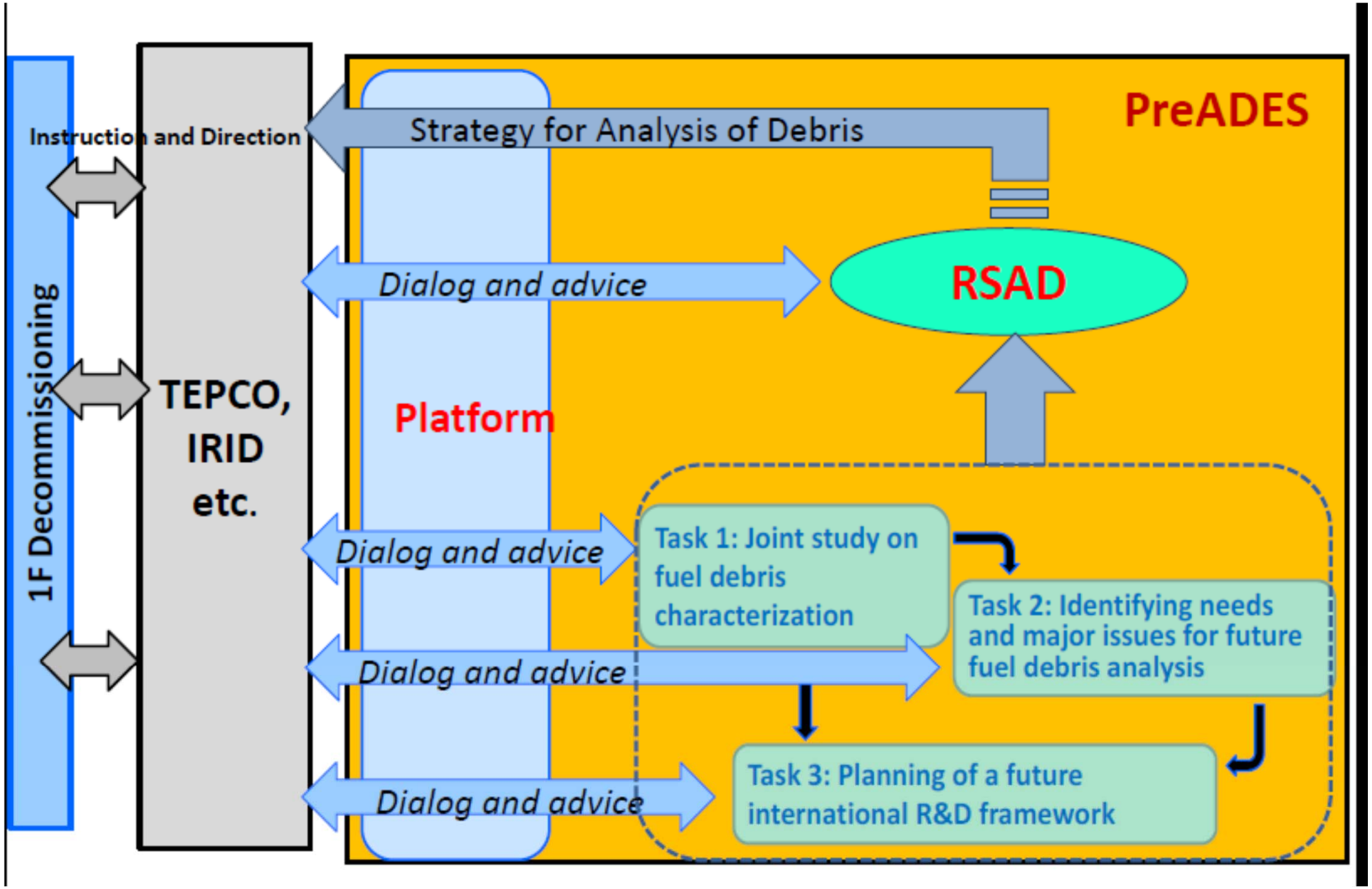
RSAD Table to be established with recommendations/support to current & future 1F debris sampling and analysis in line with the strategy established by TEPCO.

- Identifying types of data that should be obtained through 1F decommissioning, and make a comprehensive properties list correspond to estimated 1F debris location

- The data includes benefits for SA

This Platform provides opportunities for dialogue between PreADES members and TEPCO, IRID etc., and give advice for the meaning of sampling corresponding current works at 1F.

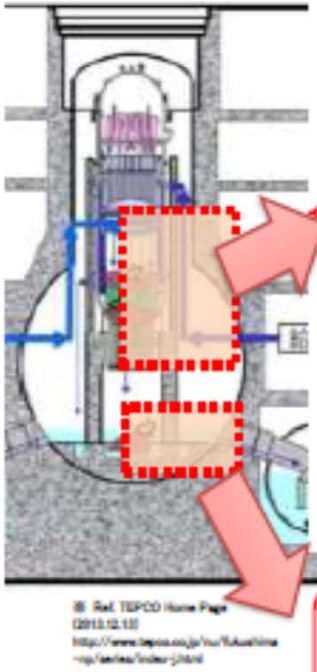
PreADES: Links between Platform and RSAD



PreADES-Output image of Task 1-1

Collaborative Laboratories for Advanced Decommissioning Science

(Ex. of debris characterization by their material properties)



※ Ref. TEPCO Home Page (2013.12.10) http://www.tepco.co.jp/nu/fukushima-np/series/index-j.html

Categories / Material/Phases	Density (g/cm³)	Vickers Hardness (GPa)	Elastic Modulus (MPa)	Fracture Toughness (MPa m$^{1/2}$)	Thermal Conductivity (W/mK)	Specific Heat (J/gK)	Melting Point (°C)	...	Behaviour at high temperature	Stability	Firing information	Danger and characteristics to be noted
RPV												
Oxide								..				
UO_2	11	4-6	190	0-2	10	0.28	2850			UO_2 should be oxidised under oxygen existing, and it causes volume change, makes powders		
ZrO_2-T	6	9-12	200	0-10	1-3	0.61	(tr.)					
$(U,Zr)O_2$-C	6-11	6-14	140-220	0-3	1-10	0.3-0.6	2500-2900					
$(U,Zr)O_2$-T	6-11	6-11	150-200	0-8	1-3	0.5-0.6	(tr.)					
$(U,Zr)O_2$-M	6	3-4	150-200	0-2	1-3	0.6-0.6	(tr.)					
Metallic								..				
Zry-2	7	1-3	60-110	110-150	10--25	0.3	1850				There are explosion cases of Zr fine	
α-Zr(O)	7	1-11	100-180	120-150	23	0.3	1850					
Fe	8	2-4	200	5-200	80	0.4	1075-1535					
$Fe_2(Zr,U)$	7-8	7-9	160-220	1-3	80	0.4	1500					
Other Ceramic												
B_4C	2.5	24	450	0-5	29	2.3	2450					
ZrB_2	6.1	22	440	0-20	24	0.7	3040					
Fe_2B	7.4	16	200	0-20	24	0.6	1389					
Pedestal Floor												
MCCI product								..				
Si rich oxide (mainly SiO2)	3	10-15	100	0-1	1	10	1600					
Si rich oxide (incl. Ca, Cr)	3	4-9	100	0-1	1	10	1600					
U rich oxide	6-11	4-9	140-220	0-3	1-10	0.3-0.6	2400-2850					
Zr rich oxide	6-7	4-9	150-200	0-8	1-3	0.5-0.6	2500-2850					
Cr rich oxide	5	4-9	100-400	0-7	10-33	0.7	2400					
$(U,Zr)SiO_4$	4-9	11	150-180	0-2	6	0.5-0.8	2500					

Mechanical property | Thermal property | Important | Safety information

PreADES-Output image of Task 1-1

Collaborative Laboratories for Advanced Decommissioning Science

(Ex. of debris characterization by appearance and properties)

The images shown are from photographs of MCCI tests, and the actual conditions of Unit 2 may be different. As the internal condition of the reactor vessel become clear, reassessment may be required.

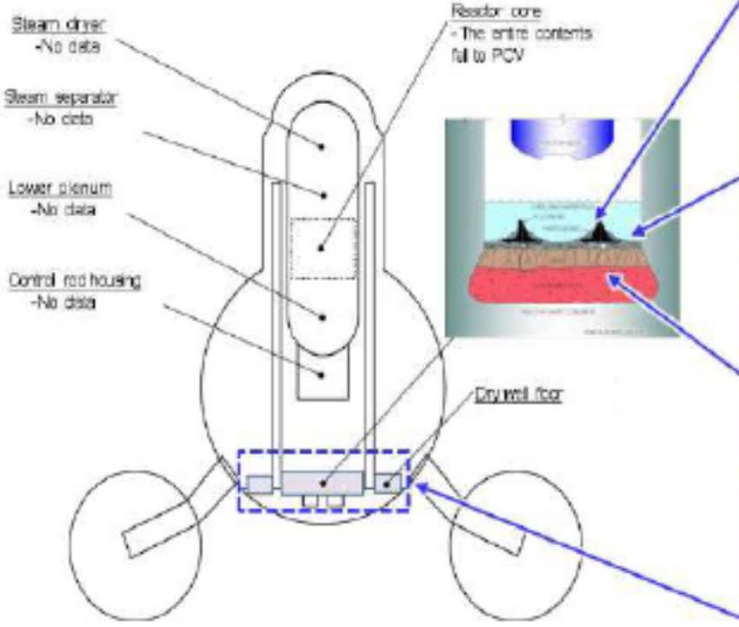
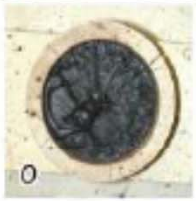


	Image	Characteristics	...
Volcano-like debris generated from MCCI		[Image L: Volcano-like debris generated from MCCI] Debris formed when the molten corium inside streamed out after the solidification of crusts (CEA VULCANO test)	...
Powder-like debris generated from MCCI	No Image M	[Image M: Powder-like debris generated from MCCI] Corium that fell into the water during the initial stage and the molten corium inside streamed out after the solidification of crusts (ANL CCI test)	
Agglomerates generated from MCCI		[Image N: Agglomerates generated from MCCI] Molten corium inside has solidified (ANL CCI test)	...
Debris generated from MCCI (upper part)		[Image O: Debris generated from MCCI (upper part)] Overall image after the test on silica-rich concrete (ANL CCI test)	
Debris generated from MCCI (vertical cross-sectional view)		[Image P: Debris generated from MCCI (vertical cross-sectional view)] Overall image after the test on silica-rich concrete (ANL CCI test) [Main composition: $(U,Zr)SiO_4$ $(U,Zr)O_2$]	

The images are extracted from the following documents with approval obtained by NDF.

L: Reprinted with permission from C. Journeau, P. Piluso, J.-F. Haquet, S. Saretta, E. Boccaccio, J.-M. Bonnet, "Oxide-Metal Corium-Concrete Interaction Test in the VULCANO Facility," Proceedings of ICAPP 2007, Nice, France, May 13-18, 2007, Paper 7328.

N & P: Reprinted with permission from Argonne National Laboratory Source: M. T. Farmer, S. Lomperski, D. J. Kilsdonk, and R. W. Aeschlimann, Argonne National Laboratory, S. Basu, U.S. Nuclear Regulatory Commission Published in: "OECD MCCI Project, 2-D Core Concrete Interaction (CCI) Tests: Final Report," February 28, 2006, OECD/MCCI-2005-TR05, http://www.ipd.anl.gov/anlpubs/2011/05/69907.pdf

O: Reprinted with permission from M. T. Farmer, "Thermite as a Validated Option to Melt Large Corium Masses," Plinius 2 International Seminar, Marseille, France, May 16, 2014.

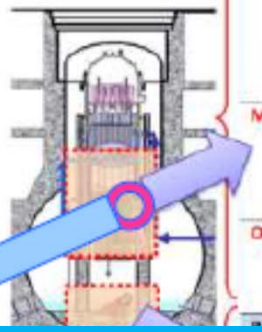
Technical Strategic Plan 2016 for Decommissioning of the Fukushima Daiichi Nuclear Power Station of Tokyo Electric Power Company Holdings, Inc.

PreADES - Output of Task 2-1 & Task 1-2

(Ex. debris characterization by appearance, properties, priority & availibility

Task 2-1, and relation with Task 1-2

(Example)



Categories / Material/Phases	Density (g/cm³)	Vickers Hardness (GPa)	Elastic Modulus (MPa)
RPV			
Oxide			
UO_2	11	6-8	150
ZrO_2-T	6	9-12	200
$(U,Zr)O_2$-C	8-11	8-14	140-220
$(U,Zr)O_2$-T	6-11	6-11	150-200
$(U,Zr)O_2$-M	6	3-4	150-200
Metallic			
Zry-2	7	1-2	60-110
α-Zr(O)	7	1-11	100-180
Fe	8	2-4	200
$Fe_x(Zr,U)$	7-8	7-9	160-220
Other Ceramic			
B_4C	2.5	34	450
ZrB_2	6.1	22	440
Fe_2B	7.4	16	200

(Task 1-2)

- This information will be provided to 1F as RSAD information.

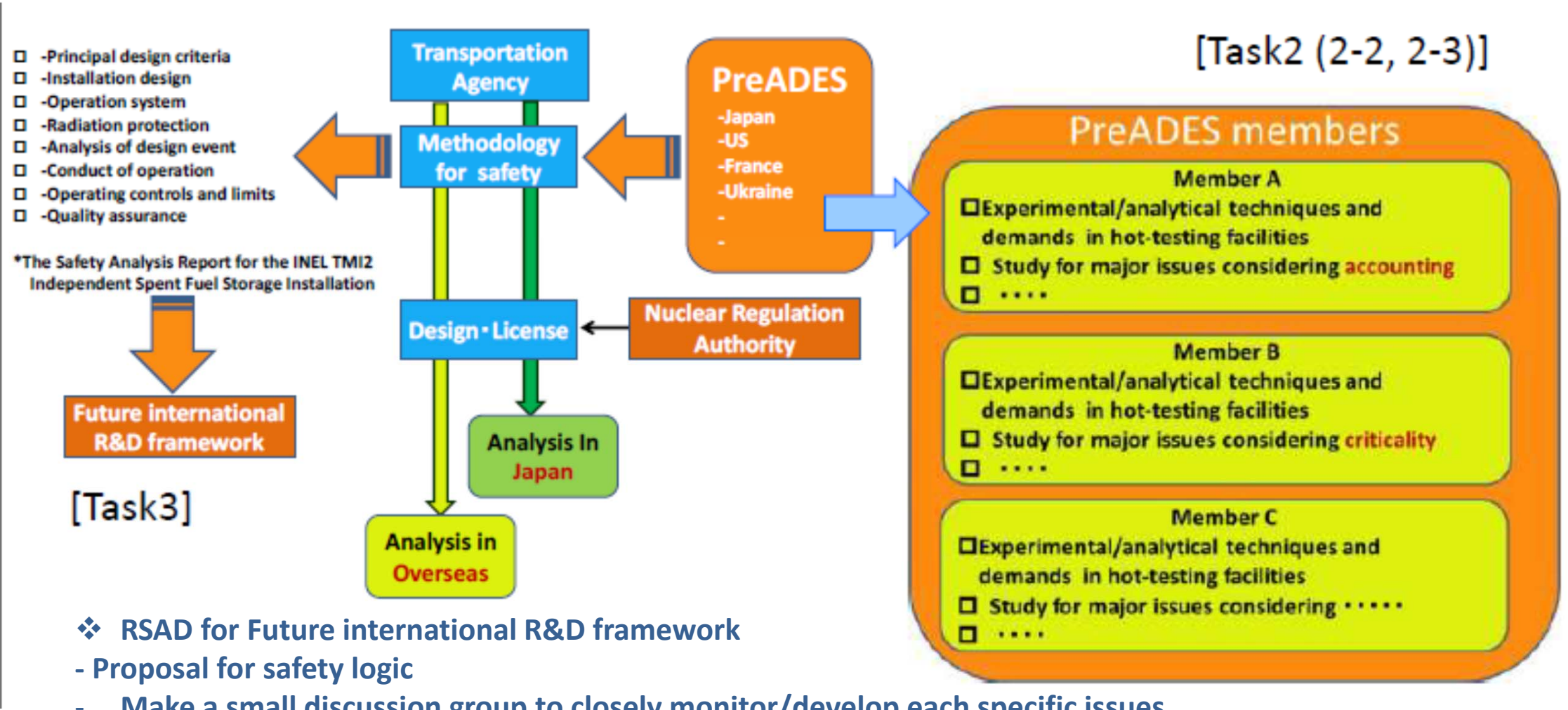
(Task 2-1)

(Example)

	Locations	Samplings	Purpose	Analysis items		Procedures
Early time	PCV	Adhesion material on robot	☐ Debris location and its conditions.	Nuclides, Chemical/mechanical properties		Photos, XRD, ICP-AES, etc.
	PCV	Contaminated water (containing suspension)	☐ Debris location and its condition.	Nuclides, Chemical/mechanical properties		Photos, XRD, ICP-AES, etc.
	• • • •					
	• • • •					
Late time	RPV	Center part of core debris	☐ Criticality, Exposure….	Nuclides, Chemical/mechanical properties		ICP-MS

PreADES -Output of Tasks 2-2, 2-3, & 3

(Proposal for safety logic & Future international R&D framework)



❖ RSAD for Future international R&D framework

- Proposal for safety logic
- Make a small discussion group to closely monitor/develop each specific issues.
- Select a lead country in each group.
- This organizational structure & information leads to future international R&D framework

Preliminary Meeting for PreADES: Outcome Summary

Collaborative Laboratories for Advanced Decommissioning Science

- PreADES

• There were no significant comments for proposed project proposal and agreement. However more detail on filling of RSAD Table will be given. JAEA discussion with OECD-NEA, just completed

• JAEA/OECD-NEA will update project proposals and circulate them together with project agreement by the end ~~September~~ October 2017.

• Participants who wish to join the PreADES will contact NEA and JAEA by end Nov. 17.

• A kick off meeting of PreADES will take place on 18-19 January, 2018.

• Further details of project to be discussed in the meeting.

- ARC-F

• Following conclusions on how to proceed on the project, a revised proposal has been sent. This will also be discussed at Jan'18 set of meetings at OECD, Paris.

TCOFF -Outline-

Thermodynamic Characterization Of Fuel debris and FP-behavior

(based on scenario analysis of Fukushima Dai-ichi Nuclear Power Station (FDNPS) accident)

Background

- Motivation: Internationally recognised need of **continuous updating knowledge of fuel degradation, including thermodynamic databases and material science models**, which applications also include providing inputs to:
  - Improvement of fuel/core degradation models and FP-behaviour models, applied in SA-codes;
  - Severe accident management and fuel debris characterization.
- Important contributions **to enhance fundamental knowledge** in this field derived from post-accident analyses for TMI-2 and Chernobyl, and additional knowledge is expected from those for Fukushima-Daiichi Nuclear Power Station (**FDNPS**)
- To progress along this direction, **MEXT/Japan** is proposing a 3-year joint project (2017-2019) to **NEA Nuclear Science Committee** (in order to maximise synergies with on-going or planned joint projects, TAF-ID (@NSC), SAREF, BSAF-2, etc., (@CSNI))
- In order to accelerate the project implementation, MEXT/Japan will provide the entire financial funding (620 kEuros over 3 years), which accounts for *project management*, *technical staff support*, and an *international call for R&D proposals*.

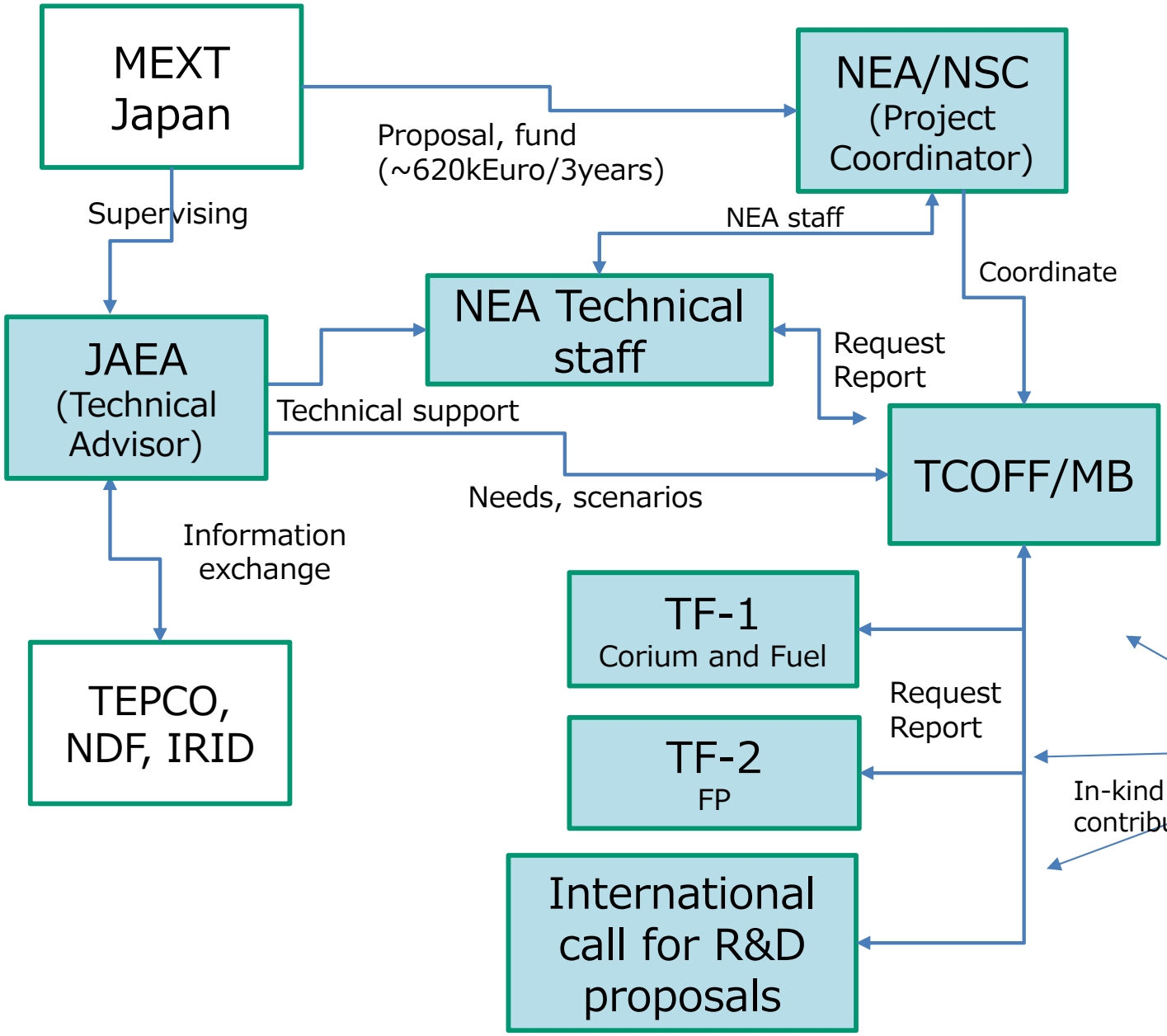
Objectives of TCOFF

1. **Improve quality and/or inventory of thermodynamic databases** which are used for fuel degradation and FP-behaviour modelling, with a reference to the severe accident progression at different units of the FDNPS. Specific technical tasks related to this objective are:
   - **Perform benchmark calculations** by various thermodynamic databases under conditions experienced during the accidents in units 1, 2 and 3 at the FDNPS;
   - **Identify and prioritize material science concerns**, in order to define specific needs of improvement and/or enlargement of current thermodynamic databases;
   - **Improve and/or enlarge thermodynamic databases** through experimental and analytical knowledge, provided by the Parties as in-kind contribution to the Project;
2. **Conduct joint thermodynamic evaluations** of the severe accident progression at in-vessel and ex-vessel phases at units 1, 2 and 3 of the FDNPS, aiming at characterizing:
   - Fuel melting and molten core relocation;
   - Fission product behaviour, including volatilization/deposition/re-vaporization and leaching;
   - Fuel debris chemical and phase composition, including long term degradation;
   - and the thermodynamic evaluation of formation of materials *which may potentially be detected at the FDNPS.* For example, various kinds of Cs-bearing particles detected in the environment, white colored powder precipitated at the floor of PRV, etc.

Method of Work and Structure of TCOFF (1/2)

- ✓ A **Management Board (MB),** composed of representatives of the project signatories, will be established in order to steer TCOFF.
- ✓ The anticipated structure foresees the establishment of **two Task Forces (TFs)** under MB, which will perform the technical evaluation;
  - **TF-1**, thermodynamic characterization of corium and fuel
  - **TF-2**, thermodynamic evaluation of FP-behaviour
- ✓ **JAEA/CLADS will act as Technical Advisor.**
- ✓ The major part of thermodynamic evaluations will be performed by **a NEA technical staff** (hired by TCOFF fund).
- ✓ TCOFF will organize an **international call for R&D proposals to fund research activities in 2018-2019** (the details TBD by MB).
- ✓ The first starting inputs for thermodynamic analyses to be carried out in TCOFF will be **TAF-ID (OECD/NEA), MEPHISTA/NUCLEA (IRSN), HERACLES (PSI) and SGTE (open source)**.

Method of Work and Structure of TCOFF (2/2)



TCOFF members

CVR (Czech Republic)
JRC (EC)
CEA, IRSN (France)
KIT (Germany)
CRIEPI, JAEA, TITECH (Japan)
KAERI (Korea)
Delft Univ. (the Netherlands)
IBRAE, KRI, NITI (Russia)
KTH, SSM (Sweden)
PSI (Switzerland)
NRC, SNL will join in 2018

TCOFF major task (1/2)

TF-1: Corium and fuel

1.Benchmark exercise and sensitivity analysis by referring SA-analysis results of FDNPS, including accident progression of core region, lower head, and MCCI (those chemical conditions are used as initial input of thermodynamic evaluation of NUCLEA/MEPHISTA, TAF-ID, and HERACLES)

2.Re-assessment for key systems, by referring SA-analysis of FDNPS

Ex. Updating U-Zr-O, Fe-B-C, Fuel-concrete systems using recent sim-test data, in-kind contribution data from TCOFF participants and newly obtained data using International Call for Proposals via NEA

3. Updating thermodynamic knowledge for use in modelling/sim-test/database

Ex. Deviation of quasi-equilibrium evaluation results by thermodynamic (material science) approach from phase relations observed in genuine debris or sim-debris/corium

TCOFF major task (2/2)

TF-2: Fission products

1.Updating knowledge of Cs-compounds

ex. To obtain/update thermodynamic data for various complex systems, including interactions between Cs and S.S., gaseous phases (Cs-M-O, Cs-B-O, Cs-Si-O, etc.), Cs-bearing particles, and etc.

2.Leaching behaviour and long term impact of FPs by thermodynamic approach

ex. To improve knowledge on long term leaching by referring recent data on debris or irradiated fuels ⇒ ex. To update knowledge on chemical conditions and the variations of Sr, Ba, Pu, Minor actinides, and so on

Potential Contribution from TCOFF

- **Evaluation of material precipitation in fuel debris:**
  Chemically active materials, neutron absorber material, Pu and Minor actinides, FP-segregation, influence of gradual solidification in MCCI etc.

- **Identification of FDNPS-samples and evaluation of the formation process:**
  Cs-bearing particles, U-bearing particles, powders accumulated in unit-1 pedestal, etc.

- **Further understanding of material interface related phenomena by material science expertise:**
  MCCI interface, crust/corium interface, other solid/liquid interfaces during liquefaction/solidification

- **Identification of potential sub-interaction/reaction along with main process**

Organisation of Workshops

- ✓ TCOFF will also provide a framework for discussion of the relevant scientific knowledge, expected to be issued from the decommissioning process:
  - The first **international workshop** to foster collaboration and disseminate knowledge will be organized in Tomioka, Japan (15 km from FDNPS) jointly with the TCOFF kick-off meeting. (3-4 July, 2017)
  - A **second workshop** aiming at advertising major accomplishments of the project is anticipated for the completion of TCOFF. (Planned in 2020)
  - **NITI and CEA** are kindly proposing to organize related workshops as a part of their in-kind contributions. (Planned in 2018-2019)
  - Relevant international partners and similar initiatives will be invited to attend and contribute to the event, as well as young researchers for training and education purposes.

TCOFF members

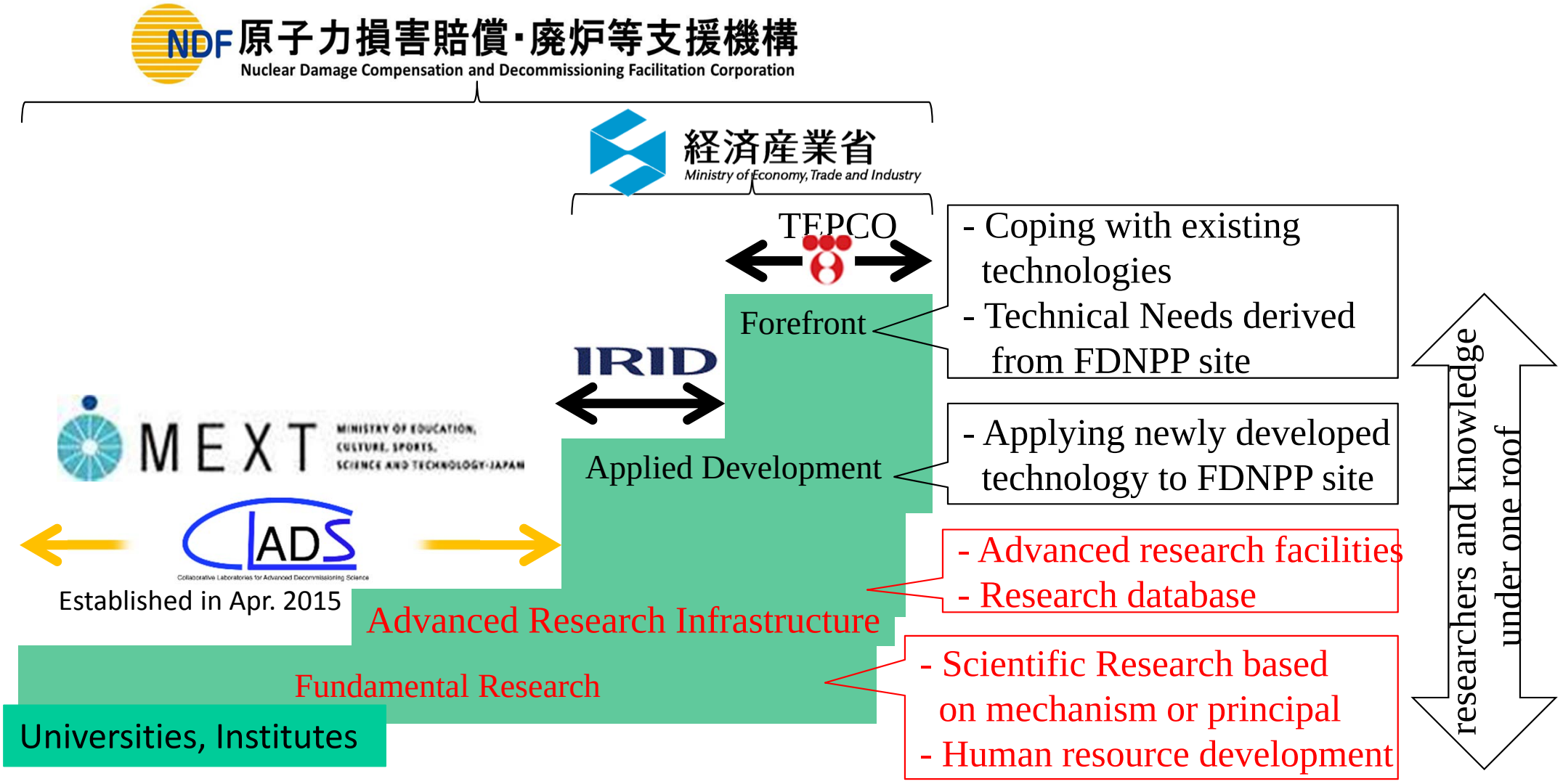
- **TCOFF was activated in June 2017 & Kick-off meeting of MB was held in July 3-4, 2017**
- **Presently 16 organisations from 9 NEA member countries + EC are joining**

TCOFF with propositions of in-kind contributions:

- ✓ Czech Republic: CVR
- ✓ Republic of Korea: KAERI
- ✓ Japan: CRIEPI, JAEA, TITECH
- ✓ France: CEA, IRSN
- ✓ Germany: KIT
- ✓ The Netherlands: Delft University of Technology
- ✓ Russian Federation: IBRAE, KRI, NITI
- ✓ Sweden: KTH, SSM
- ✓ Switzerland: PSI
- ✓ European Commission, JRC-Karlsruhe

* NRC (SNL) will join TCOFF in 2018. Two Russian institutes, Saint Petersburg State University & LOFFE also interested in participation.

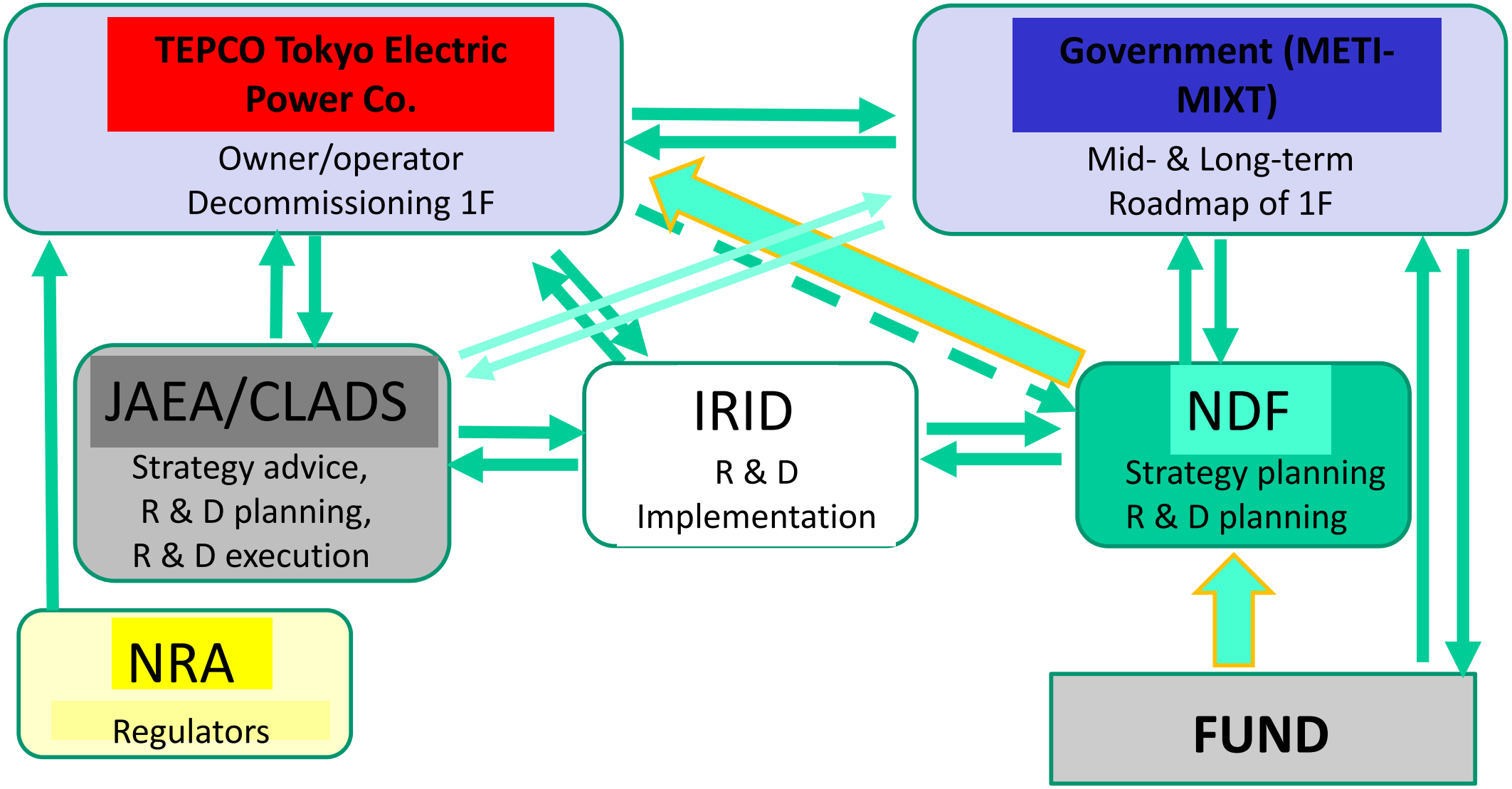
Role of CLADS in Whole Structure of R&D for Decommission of FDNPP



Role of CLADS = Bridging between Advanced R&Ds, including international R&D for improving safety, and “Needs” or “Expectation” of TEPCO, IRID and NDF in near/mid/long-term.

Japan proposes new projects to NSC and CSNI in OECD/NEA, and JAEA/CLADS will act as “technical advisor” or “operating agent” of these projects.

Structure of Fukushima research



Concluding Remarks & Outlook

- The structure of international research & development and technical execution for decommissioning at Fukushima (F1) units has been described.
- These international programs (such as operated by OECD-NEA or IAEA) require time to set up, they enable the wide international experience in severe accidents to be applied to this complex project.
- 2 example projects have been given: **Pre-ADES and TCOFF**.
- These should optimise sampling at 1F (Pre—ADES) and the improvement of thermodynamic data for application to/modelling of fuel degradation mechanisms (TCOFF).
- The priority is to support the immediate decommissioning operations by TEPCO but also to carry out longer term research for the fuel debris treatment, container design and storage.
- The use of existing experience from Chernobyl and TMI-2 is important; but the use of existing severe accident research from EU programs and elswhere in the world. These projects facilitate thisHere the role of such international programs operated by OECD-NEA have an important role.
- Finally for this very long term project, JAEA needs to conserve its own knowledge as well as assimilate it from other sources. It therefore needs to encourage young researchers to work at JAEA-CLADS in this field.

Finally…..

Many thanks for your attention!

- Any questions about these projects please ask
  Tadahiro Washiya (tadahiro.washiya@jaea.co.jp) and,
  Masaki Kurata (masaki.kurata@jaea.co.jp)

- If any young researcher is interested in working at JAEA-CLADS, please contact either the persons above or myself or Dr. Nagae (also present at the QUENCH Workshop).

CLADS test facilities on BWR core material degradation

Yuji Nagae, Hiroki Shibata, Anton Pshenichnikov, Saishun Yamazaki, Masaki Kurata
Japan Atomic Energy Agency (JAEA)
Motooka, Tomioka-machi, Futaba-gun, Fukushima 979-1151, Japan.

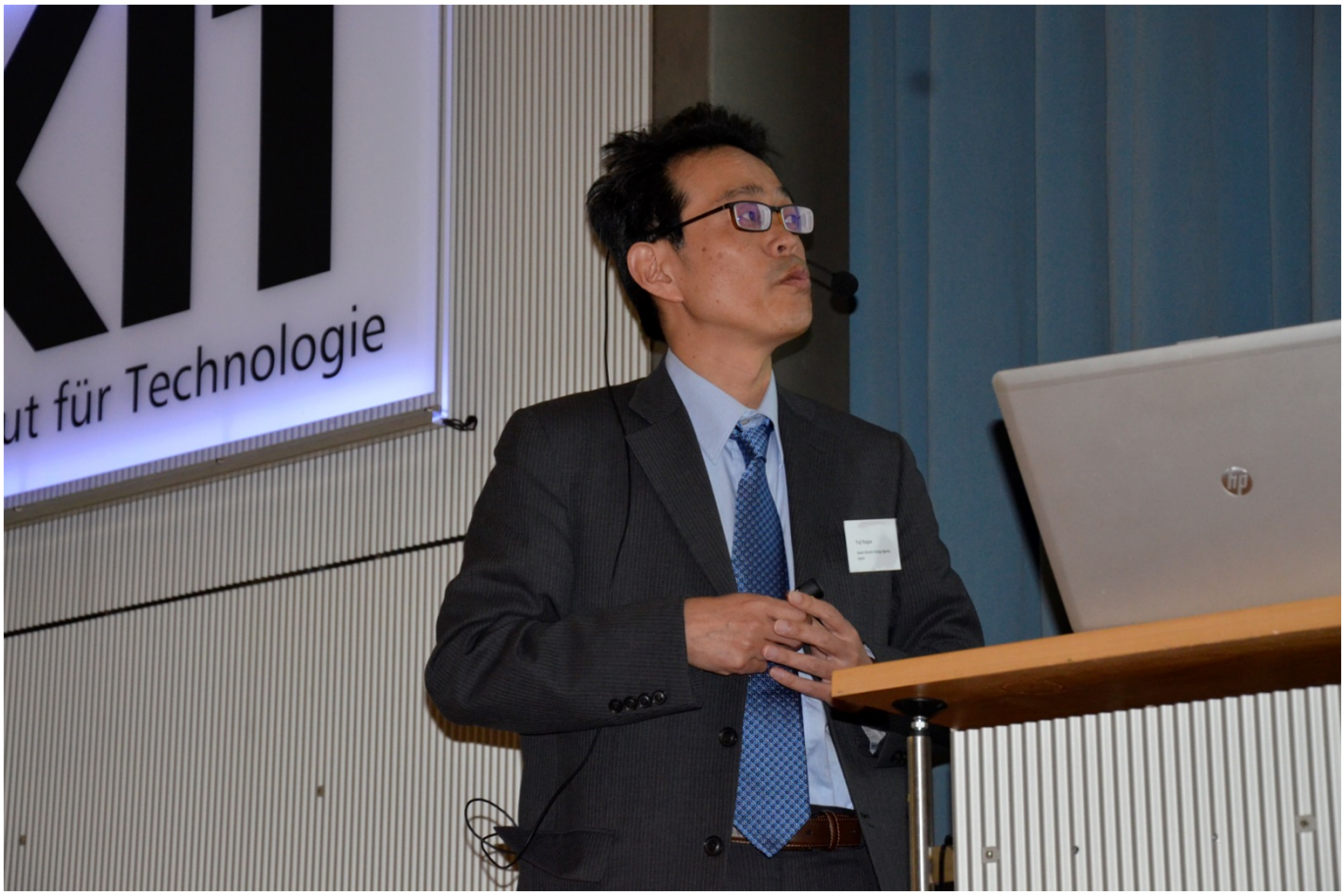
Abstract

Two control blade degradation test facilities which were designed for the investigation of high temperature degradation of the nuclear reactor elements, have been set up in Tomioka site of CLADS in Japan Atomic Energy Agency. One was a test facility for small-scale samples and another one was test facility for large-scale samples. In situ video of the experiment and mass-spectrometric analysis of the output gases (H_2, CO) can be carried out during tests, and heating rate can be controlled up to 1 K/s. The working area of the large-scale facility consists of the isolated heating area 1.2 m height with the water cooled lower part to achieve the natural temperature gradient conditions as during severe accident. Steam rate in large-scale facilities can be controlled in the range of 15 – 100 g/min, and a test piece for the control blade degradation tests can be used to simulate a part of the control blade and fuel assembly with 10 cm×10 cm×1.2m (height).

Chemical reaction between stainless steel/B_4C and Zry is affected by the oxide layer on Zry. The oxidation behavior depends on steel flow rate and tendency of control blade degradation possibly has threshold condition. According to pretest, critical condition for reaction between molten control rod and channel box can be recognized. Further analysis and tests in various steam rate will be conducted to evaluate the critical condition.

Another test facility using laser heating system has also been set in Tomioka site. It was specially designed to achieve very high heating rates and temperature gradient to study high temperature degradation or oxidation in steam. Local temperature reaches very high values so that, for example, Zr metal can be easily melted in a few minutes.

This study is performed in “Advanced Multi-scale Modeling and Experimental Tests on Fuel Degradation in Severe Accident Conditions” supported by Ministry of Economy, Trade and Industry, Japan (METI).


ut für Technologie

23rd QUENCH Workshop at KIT
2017.17 Oct.

CLADS test facilities on BWR core material degradation

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This study is performed in “Advanced Multi-scale Modeling and Experimental Tests on Fuel Degradation in Severe Accident Conditions” supported by Ministry of Economy, Trade and Industry, Japan (METI).

Outline

1. Background
2. Previous tests in other laboratories
3. Introduction of test facilities in Tomioka site of CLADS
4. Results on degradation behavior
5. Summary

Background

SA-experts reached to "General consensus" on fuel degradation progression at severe accident (SA) before FDNPS accident.

- Fuel degradation progresses by overlapping complicated physical-chemical steps under non-equilibrium conditions.
- **"Simplified models for fuel degradation"** are necessary and sufficient for comprehensive understanding of its progression (especially for fuel liquefaction and relocation) **"under prototypic scenarios",** and these models were implemented in SA-analysis codes.
- Priority of the detailed mechanistic modelling was recognized to be low for these comprehensive understanding.

However, several accident steps progressed under "extreme/specific conditions" in each unit of FDNPS (because many desperate efforts were carried out to stop or to mitigate SA-progression)

- Simplified models is often not appropriate for **"extreme/specific conditions"**.
- To bury the gap between **"simplified model"** and **"real fuel degradation in each unit"** or **"as the result, debris characteristics in each unit"**, importance of detailed understanding based on **phenomenology** (mainly by **material science**) is being recognized, especially for **"BWR-fuel assembly".**

Previous tests on degradation behavior in control blade

B_4C tests					
CORA with BWR bundles with B_4C, FZK, Germany	**QUENCH-07, -09 KIT (former FZK) with B_4C, Germany**	**Phebus-FPT3 IRSN, France**	**DF-4 Sandia NL, United States**	**CODEX-B_4C, KFKI, Hungary**	**XR2-1, Sandia NL, United States**
● Out-of-pile ● Heating rate: 1 K/s In CORA-31 and 30 0.3 and 0.2 K/s ● Steam flow rate: 0.111 g/s/rod ● Normally no pre-oxidation ● CORA-28 and 29 clearly pre-oxidized ● CORA-16 Fuel rods: 18 B_4C: 11	● Out-of-pile ● Heating rates: 0.3 – 1 K/s (Mostly 0.3-0.5 K/s) ● Steam flow rate: 0.146 g/s/rod ● Steam starvation QUENCH-09 **0.019 g/s/rod** ● QUENCH-07, -09 with pre-oxidation ● 20 fuel rods	● In-pile test (UO_2 pellets) ● Steam flow rate: **0.025 g/s/rod** ● Geometry similar to QUENCH bundle 5 experiments but only one B_4C test ● 20 fuel rods (2 fresh 2 irradiated)	● In-pile test (UO_2 pellets) ● Steam flow rate: 0.126 g/s/rod ● Pre-oxidation: 700 s at 1000 K 0.44 g/s ● 14 fuel rods ● Geometry like CORA	● Out-of-pile ● Steam flow rate: 0.138 g/s/rod ● Pre-oxidation: 0.027 g/s/rod ● 6 fuel rods with 1 control rod of B_4C ● Russian WWER type fuel with WWER geometry an E110 (Zr-1%Nb) material	● Out-of-pile ● Main focus Melt relocation ● 1st 20 kg of control blade melt ● 2nd 40 kg of Zry

Fundamental research on degradation behavior

【Objective】
Contributing to progress in sever accident analysis and accident management by experimental knowledge and detailed modeling for fundamental degradation behavior based on material science

【Main research theme】
Experimental knowledge on fundamental process in sever accident using developed facilities in corroboration with institutes and universities, and development of detailed mechanistic modeling

Welcome to CLADS test facilities in Tomioka of Fukushima

- **Control blade degradation test facility**
  Temp.(max.): 1700℃, Sample size: 10cm × 10cm × 1.2m (height)
- **Small-scale control blade degradation test facility**
  Temp.(max.): 1700℃, Sample size: 2cm × 2cm × 5cm (height)
- **Laser heating furnace**
  Temp.(max.): 2000℃, Sample size: 2cm × 2cm × 1cm

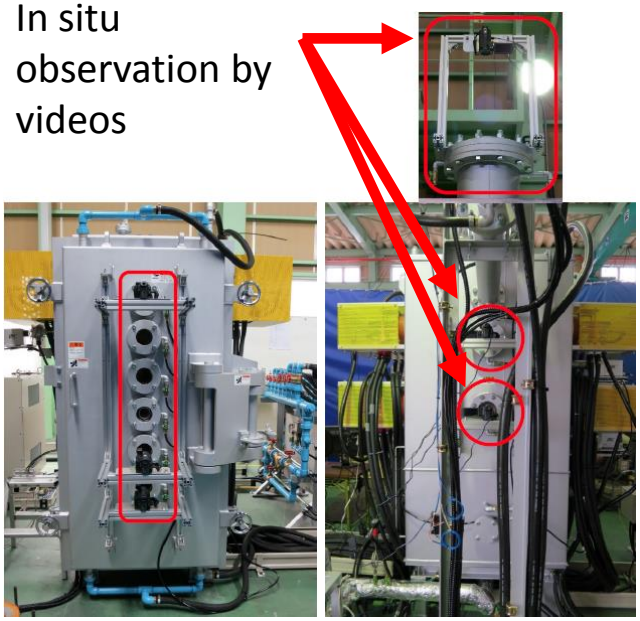


Test facility building

Large-scale control blade degradation test facility

- **JAEA has developed two test facilities for evaluating chemical interaction progression between control blade and Zry-channel box, which is thought to happen in the very early stage of severe accident (SA) progression in BWR.**
- **A large-scale test piece is able to be heated under sim-SA conditions, which include high temperature steam atmosphere, high temperature increase rate, and axial temperature gradient.**
- **Progression of interaction is able to be monitored by three video cameras.**
- **The results will provide useful information on "How the control blade channels were degraded in Fukushima-Daiichi Nuclear Power Station Accident" and also on improving SA-analysis codes.**

In situ observation by videos

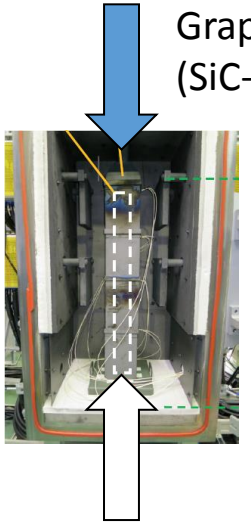


Appearance of test facility



Test piece set in the furnace

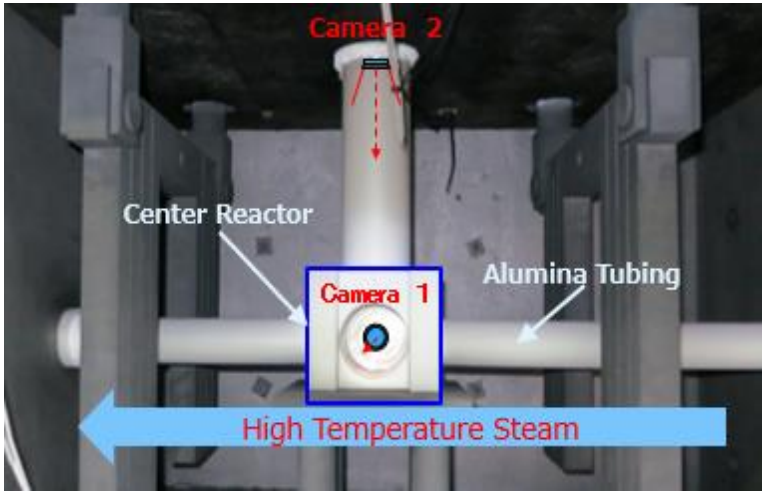
Test piece is set in Graphite container (SiC-coating)



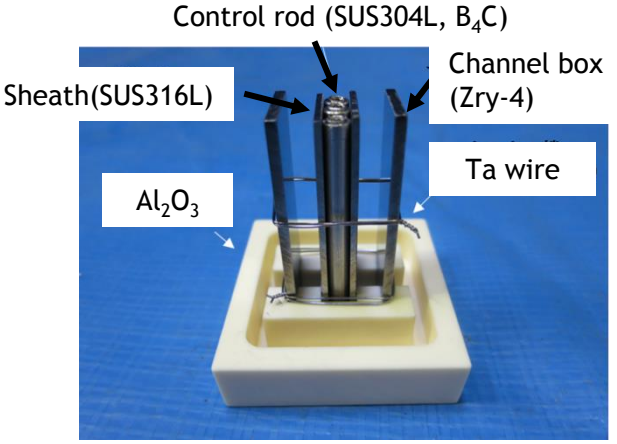
High temperature steam

Small-scale control blade degradation test facility

- A **small-scale test piece** is able to be heated under high temperature steam atmosphere (**homogeneous conditions**).
- Progression of interaction is able to be monitored by two video cameras.
- The results will provide useful information on fundamental behavior of reaction between molten S.S.-B_4C and Zircaloy under various homogeneous conditions.



Technical data	
Maximum temperature	~ 1973 K / 1700 °C
Heating type	Resistive heater
Temperature measurement type	Pyrometric and/or thermocouple
Heating zone, approximately	~ 5 cm long
Heating control / Heating rate	PC-controlled (PID controller) / up to 1 K/s
Working Atmosphere	Argon, Ar + steam mixture
Steam flow rate	Manual control
Size of samples	2 cm×2 cm×5 cm (height)

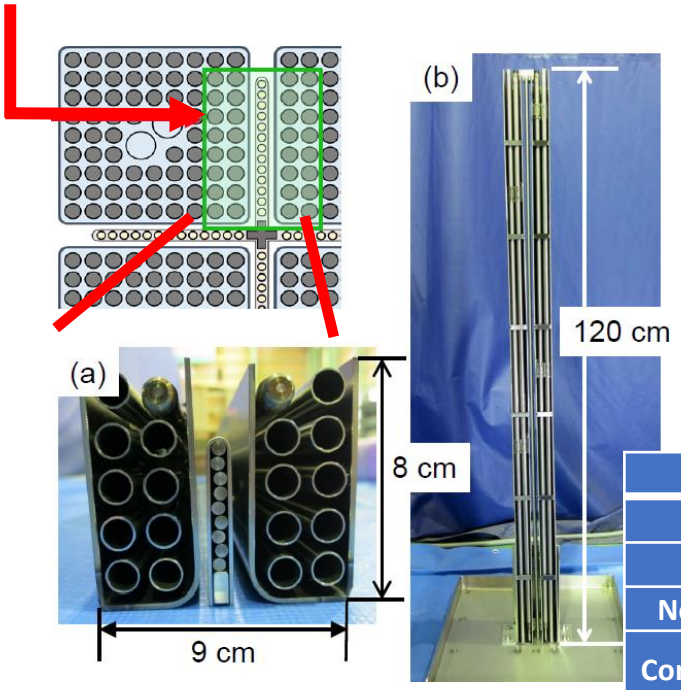


A test piece

Effect of steam flow rate on control blade degradation

- **Visual data of in-situ observation obtained for detailed mechanistic modeling.**
- **Oxidation behavior depends on steel flow rate and tendency of control blade degradation possibly has threshold condition. Critical condition for reaction between molten control rod and channel box can be recognized.**

Cross section of BWR fuel assembly



- These materials are equivalent to those applied for typical BWRs in Japan.
- Thickness of pre-oxidized layer on Zry:
  ～ 0 (fuel rod)
  a few μm (channel box)

Parts	Materials
Fuel cladding	Zry-2
Channel box	Zry-4
Neutron absorber	B_4C
Control rod cladding	S.S304L
Control blade	S.S316L

Appearance of test piece (before)

Control blade and channel box was co-degraded.



0.0125 g/s/rod

Only the control blade was degraded at the upper part.



0.0417 g/s/rod

Steam flow rate

Appearance of test piece (after)

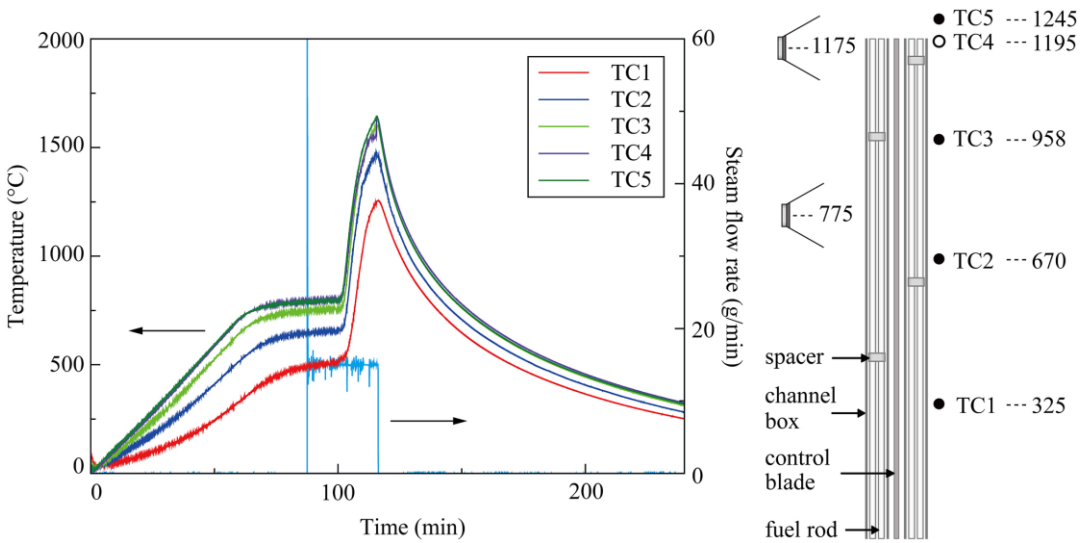
Ref. PhebusFPT-3 test : 0.025g/s/rod, CORA-16 test : 0.111 g/s/rod

Appearance of the degraded test piece (0.0125g/s/rod)

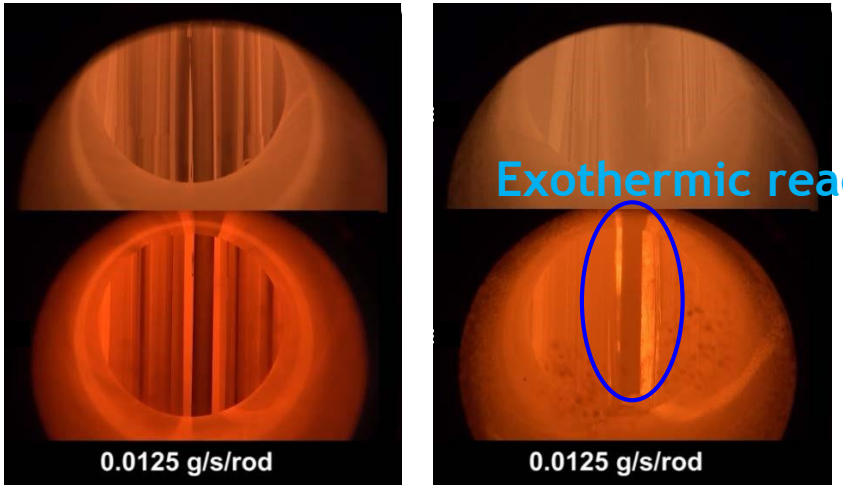
➢ Control blade and channel box were co-degraded.



Droplet-like



Temperature profile

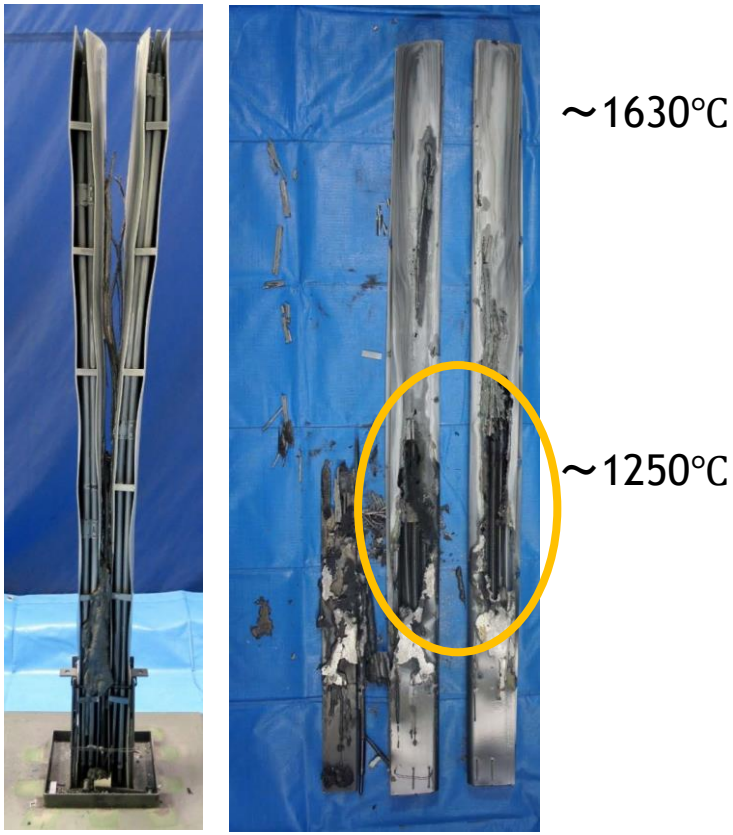


Snapshots

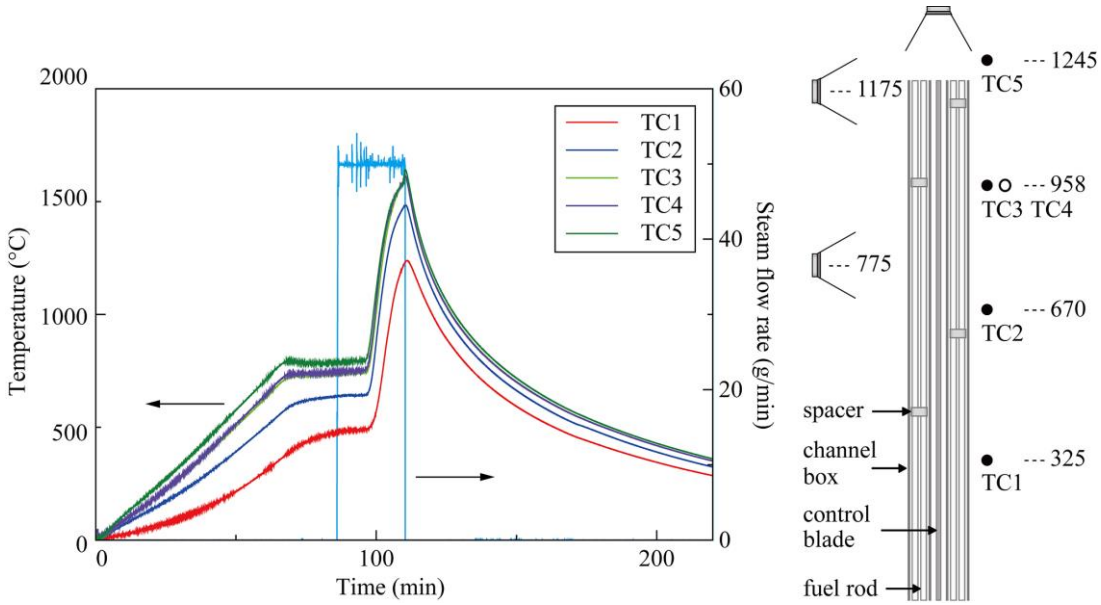
- A reaction between Zr alloy and molten S.S.-B_4C would occur from top to middle part.
- Exothermic reaction was observed.

Appearance of the degraded test piece (0.0417g/s/rod)

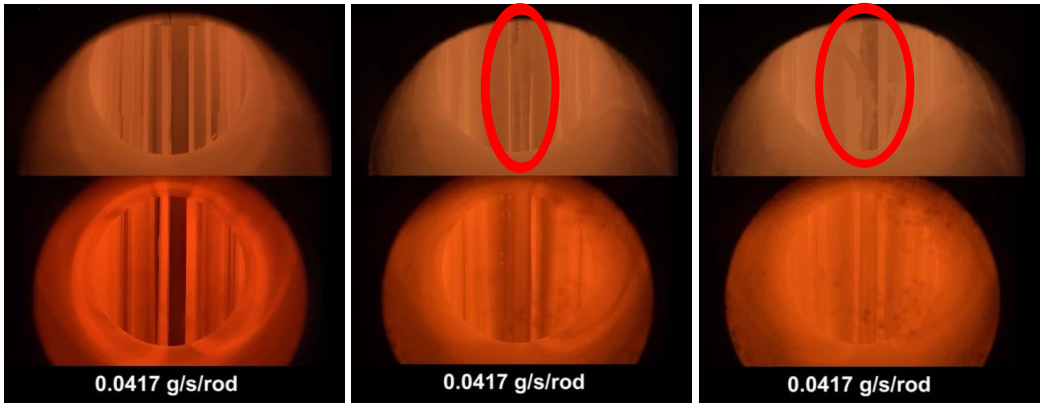
➢ Partial chemical interaction between the S.S./B_4C-melt and Zry was observed only at the lower part.



- Thickness of oxide layer in lower side is probably thinner than in upper side.
- A reaction between Zr alloy and molten S.S.-B_4C would occur in lower part.



Temperature profile

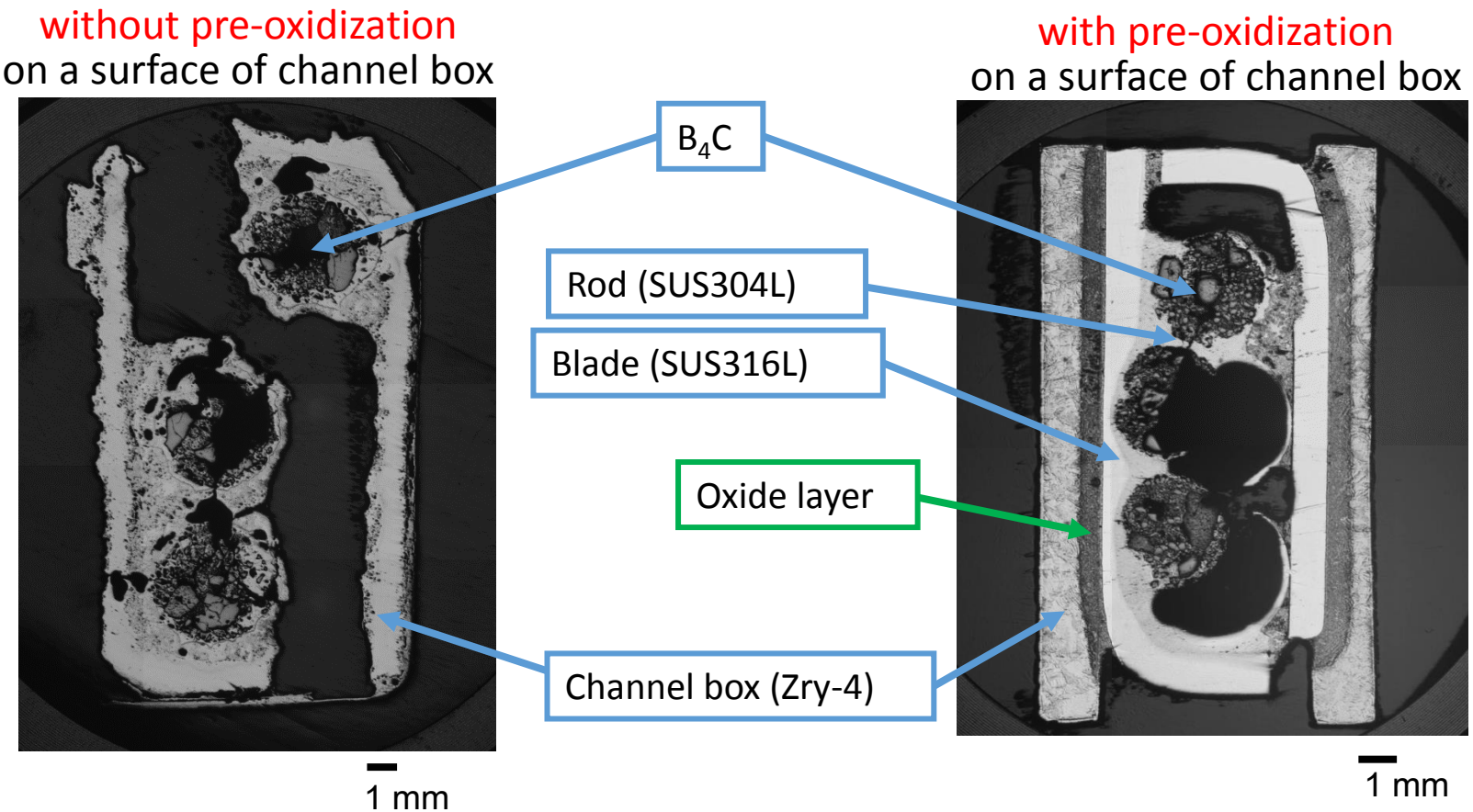


Snapshots

Reaction between molten S.S.-B_4C and Zircaloy

- Test was carried out at 1250℃, at which eutectic reaction between S.S. and B_4C occurs obviously.
- Oxide layer probably protects a reaction between molten S.S.-B_4C and channel box.

Test temperature and hold time : 1250℃×30min



Summary

- Introduction of test facilities in Tomioka site of JAEA
  - Large-scale control blade degradation test facility
  - Small-scale control blade degradation test facility
  - Laser heating furnace

- The results will provide useful information on degradation behavior in Fukushima-Daiichi Nuclear Power Station Accident and also on improving SA-analysis codes.

◆ Future plans
- Threshold conditions will be elucidated by performing parametric tests.
- Apparent Zry oxidation rates, especially in steam starvation condition, will be evaluated by analyzing test pieces.

Main Objectives and Results of the EU SAFEST Project.

A. Miassoedov[a,*], B. Fluhrer[a], C. Journeau[b], S. Bechta[c], Z. Hózer[d], D. Manara[e], P.D. Bottomley[e], M. Kiselova[f], T. Keim[g], G. Langrock[g], F. Belloni[h], M. Schyns[h]

[a] *KIT, Karlsruhe (DE)* [b] *CEA, Cadarache (FR)* [c] *KTH, Stockholm (SE)*
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Abstract

No individual country has sufficient resources to address all severe accident important phenomena within the framework of a national research programme, therefore optimised use of resources and the collaboration at European and international level is very important. Integrating European severe accident research facilities into a pan-European laboratory for severe accident and corium studies and providing resources to other European partners for better understanding of possible accident scenarios and phenomena is necessary in order to improve safety of existing and, in the long-term, of future reactors.

SAFEST (Severe Accident Facilities for European Safety Targets) is a European project networking the European corium experimental laboratories with the objective to establish coordination activities, enabling the development of a common vision and research roadmaps for the next years, and of the management structure to achieve these goals. One of the main objectives is to address and resolve the variety of the remaining severe accident issues related to accident analysis and corium behaviour. The project is a valuable asset for the fulfilment of the severe accident R&D programmes that are being set up after Fukushima and the subsequent European stress tests, addressing both national and European objectives.

Roadmaps on European severe accident experimental research for water reactors and for GenIV technologies will be drafted. Joint R&D is conducted to improve the excellence of the SAFEST facilities: this includes measurement of corium physical properties, improvement of instrumentation, consensus on scaling law rationales and cross comparison of material analyses.

Joint experimental research is a clear objective in the SAFEST project to provide solutions for stabilisation of severe accident and termination of consequences for the current GEN II and III plants. Consequently, the knowledge obtained in SAFEST shall lead to improved severe accident management measures, which are essential for reactor safety- In addition it will offer competitive advantages for the nuclear industry and contribute to the long-term sustainability of nuclear energy.



Main objectives and results of the EU SAFEST project



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F. Belloni, M. Schyns (SCK-CEN)

17-19 October 2017
KIT, Karlsruhe

SAFEST

Severe **A**ccident **F**acilities
for **E**uropean **S**afety **T**argets

EU-supported pan-European Integrated Research Infrastructure Initiative for increased safety of nuclear systems at EU level

Duration: 4 years starting on 1 July 2014

www.safest.eu

Project background

- Severe accidents are the focus of considerable research involving substantial human and financial resources worldwide
  - too many challenging physical phenomena, complicated further by high temperatures and presence of radioactive materials
  - no individual country has sufficient resources (both human and financial) to address all important phenomena in the framework of a national research programme
- Integration of major European severe accident research facilities into a pan-European laboratory:
  - severe accident and corium studies
  - providing resources to other interested European partners for better understanding of possible accident scenarios and phenomena
  - improving safety of existing and, in the long-term, of future reactors

Eight European partners, coordinated by KIT

Karlsruhe Institute of Technology (**KIT**, Germany)

Commissariat à l'Energie Atomique et aux Energies Alternatives (**CEA**, France)

Royal Institute of Technology (**KTH**, Sweden)

Centre for Energy Research, Hungarian Academy of Sciences (**MTA-EK**, Hungary)

Joint Research Centre Karlsruhe (**JRC**, Karlsruhe)

UJV Řež, a.s. (**UJV**, Czech Republic)

AREVA GmbH (**ANP**, Germany)

Belgian Nuclear Research Centre (**SCK CEN**, Belgium)

+

Collaborative Laboratories for Advanced Decommissioning Science (**CLADS**, Japan)

CLADS joins SAFEST as a partner in October 2017

CLADS contribution to the SAFEST activities

- CLADS was established in April 2015 in JAEA as a R&D centre for advanced decommissioning science and is aimed at being an international hub for R&D on decommissioning, and promoting cooperation in R&D and human resource development among government, industry and academia
- Development of joint research roadmap to focus future joint R&D on the stabilisation and termination of severe accidents in PWRs and BWRs
- Exchange in cold crucible simulations with interested SAFEST partners CEA, AREVA, UJV
- BWR-specific issues like control blade behavior and clad oxidation in steam starved conditions
- CLADS operates or plans to build small and medium scale test facilities as a part of CLADS experimental infrastructure. They can be opened for proposals from European organizations for joint experiments
- Participation of SAFEST labs in the analysis of Fukushima samples (organized via SAREF, SAFEST labs will be considered with high priority)
- Involvement in the TCOFF project on fuel debris characterization

Project main goals

- ***Development of research roadmaps*** to focus future European R&D on the stabilisation and termination of severe accidents in PWRs and BWRs
- ***Creation of an integrated pan-European laboratory for severe accident research*** able to address and successfully resolve the wide variety of issues related to severe accident analysis and corium behaviour
- ***Establishing the access to SAFEST research infrastructure*** to investigate all important phenomena from the early core degradation to corium pool formation in the lower head, and ex-vessel melt situations
- ***Continuous improvement and upgrading of the SAFEST infrastructure*** to increase the experimental capabilities and overall quality of R&D to meet current and future challenges
- ***Applications of the results*** of the project to the European light water reactors

SAFEST experimental facilities

- In-vessel and ex-vessel corium and debris behaviour
- Corium properties

QUENCH at KIT



VULCANO and VITI at CEA



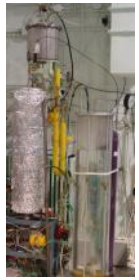


DEFOR and SES at KTH





CODEX at EK



SICOPS at AREVA



MOCKA and LIVE at KIT





COMETA at UJV



KROTOS at CEA, POMECO at KTH CERES at EK







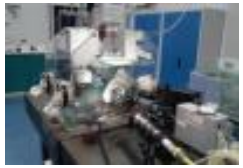
DISCO at KIT, RESCUE at CEA, MISTEE at KTH







FLF at ITU



Experiments in SAFEST facilities

- Two calls for proposals were announced in 2015 and 2016
  - 22 proposals from 10 organisations
  - User Selection Panel with help of independent experts evaluated the submitted proposal and recommend the experiments to be performed in SAFEST
  - Experiments in 16 facilities

- First tests performed in 2016, many tests are scheduled for 2017

Objectives of the SAFEST experiments (1/3)

Test facility	Proposing Organisation	Test objective	Date
MISTEE-AL KTH	IRSN + others	Investigation of the ability of alumina melt to undergo thermal fragmentation as a mechanism for spontaneous steam explosions and determination of the void produced with high temperature drops	09/2015
COMETA UJV	CEA	Quantitative assessment of the kinetics of the reaction between UO_2 and B_4C	03/2016
SES KTH	EDF + others	Investigation of the energetics of spontaneous steam explosion and of the characteristics of the premixing zone in stratified melt under water configuration	04/2016
DEFOR KTH	KIT	Investigation of the influence of control rod and instrumentation guide tubes on melt fragmentation in water for BWRs	07/2016
SICOPS AREVA	SSM	Investigation of MCCI with BWR-specific corium composition (lower U/Zr ratio, lower oxidation fraction of Zr, reduced steel mass) and concrete composition (basaltic in Nordic BWRs)	2016/2017

Objectives of the SAFEST experiments (2/3)

Test facility	Proposing Organisation	Test objective	Date
FLF JRC	SSM	To receive experimental data and to improve understanding of the quasi-binary section of ternary U-Zr-O diagram in the composition domain of BWR specific corium composition, i.e. at U/Zr atomic ratio close to 0.9 and Zr oxidation index close to 20 - 30%	2016/2017
MISTEE-OX KTH	IRSN + others	To investigate the ability of pure zirconium melt to undergo thermal fragmentation while falling into water and thereby undergoing oxidation	2016/2017
CERES MTA EK	Bohunice NPP	Investigation of the effect of the elevated sump level on the in-vessel melt retention for VVER-440 reactors during the melt retention by flooding of the reactor cavity	2017
DISCO KIT	IRSN	Investigation of scaling effect during FCI premixing in the DISCO facility	07/2017
CODEX MTA EK	KIT + others	influence of nitrogen on oxidation and degradation of fuel rod claddings in the temperature range of 800-1200 °C in air-steam atmosphere	2017

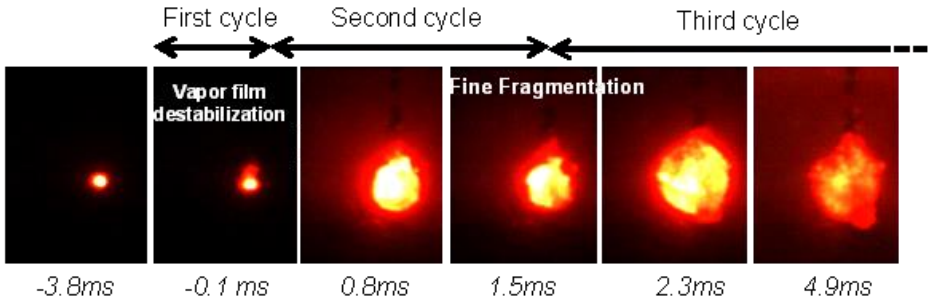
Objectives of the SAFEST experiments (3/3)

Test facility	Proposing Organisation	Test objective	Date
MOCKA KIT	SSM	Study of MCCI in 2D cylindrical crucibles made of concrete typical for Nordic BWRs	07/2017
QUENCH KIT	SSM	Study of BWR-specific important phenomena like influence of control rod blade on bundle degradation	09/2017
LIVE KIT	IRSN	Assessment of the characteristic time of convection development in a homogeneous melt pool in 3D after a rapid change in boundary conditions	07/2017
FLF JRC	EDF + others	Measurements of emissivity of U+Zr+steel alloys in different atmospheres	2017
VULCANO CEA	KIT + GRS	Influence of crusts on the melt-concrete interface during MCCI	2017
VITI CEA	SSM	To obtain thermophysical properties (density, surface tension and, if feasible, viscosity) of (U, Zr, Fe) metallic alloys representative of reactor compositions	2018

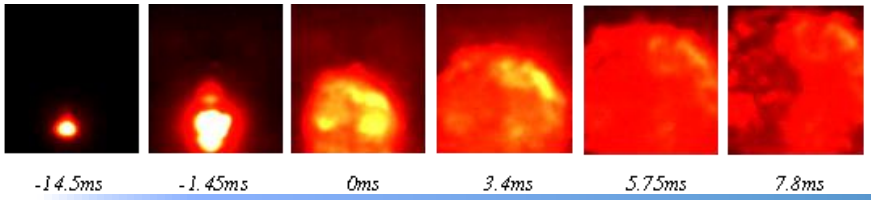
MISTEE-AL experiments on single drop steam explosion

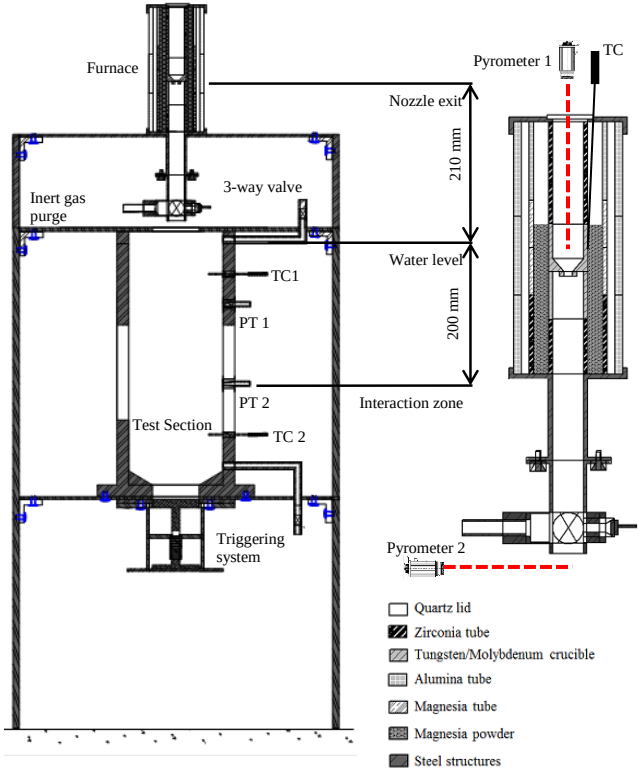
- Facility has been upgraded with new furnace and additional pressure measurements
- 24 tests with Al_2O_3 melt drops FCI are completed
- Only one spontaneous steam explosion happened at very high melt superheat and high water subcooling
- Measured data were used by users for model validation and code development

Triggered SE (Superheat≈ 167K, Subcooling ≈ 82K)



Spontaneous SE (Subcooling ≈85°K, superheat ≈215°K)





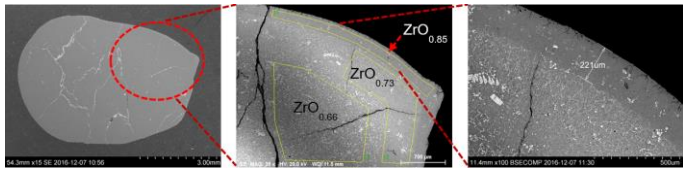
MISTEE-AL experiments on Zr melt fragmentation

- To study oxidation behaviour of Zr melt drops
- Proposed by the user group from IRSN, CEA and JSI
- Facility has been upgraded for metal melts
- The measurements include:
  - High speed visualization of the Zirconium melt droplets and water interaction
  - Estimation of hydrogen production from the size of the bubbles in subcooled water
  - Post-test SEM/EDX analysis and oxygen determination in the quenched drops
- It is complicated to measure melt temperature in water: it will be estimated
- First performed experiments demonstrated:
  - Much larger lifetime of molten Zr drop in water in comparison with other materials which do not undergo oxidation
  - Significant hydrogen formation
  - Large final oxidation of quenched drop
- The tests and post-test examinations will be completed in 2017

Snapshots of Zr drop in water

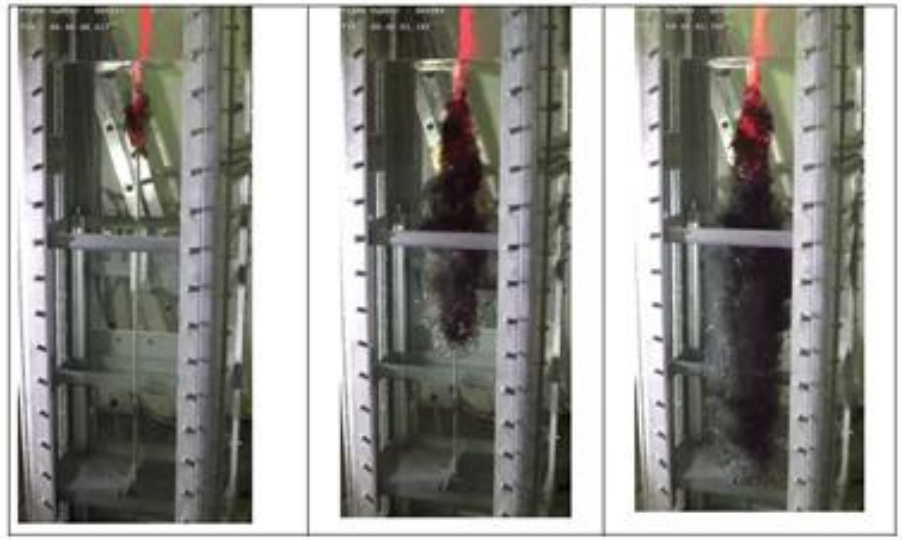


Micro-structure of the quenched drop

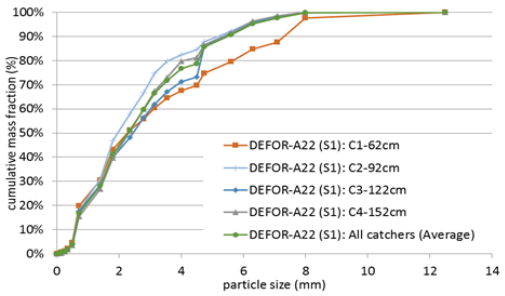


DEFOR experiment at KTH on melt jet fragmentation in water

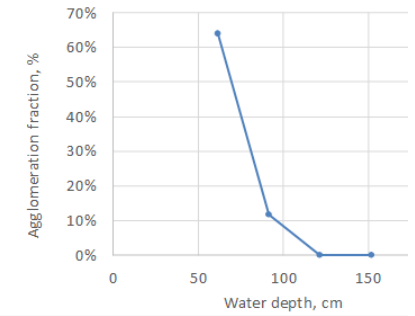
- Test objective was to estimate an influence of control rod and instrumentation guide tubes on melt fragmentation in water and debris bed formation and characteristics
- Eutectic mixture of Bi_2O_3–WO_3 to simulate corium melt
- The main test parameters were similar to the DEFOR-A4 test which was characterised by a relatively low melt temperature of 1221 K (melt superheat of 78 K)
- Although, the measured porosity of debris bed is slightly higher in comparison with the reference tests without the rod, there is general agreement with data obtained in previous DEFOR tests
- Modification of test conditions is recommended for the future tests



Cumulative particle size distribution

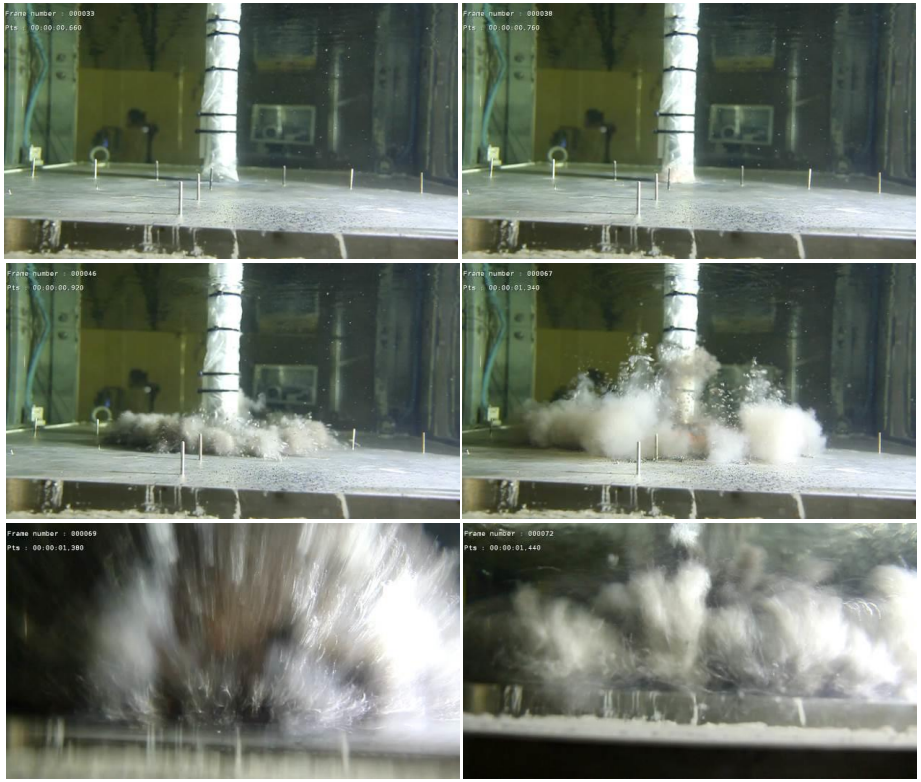


Agglomeration factor



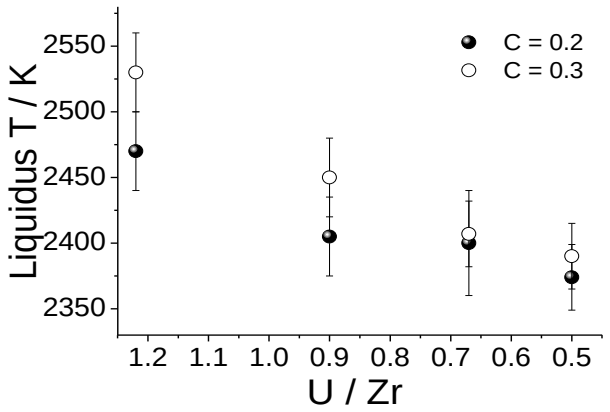
SES experiment on stratified steam explosion

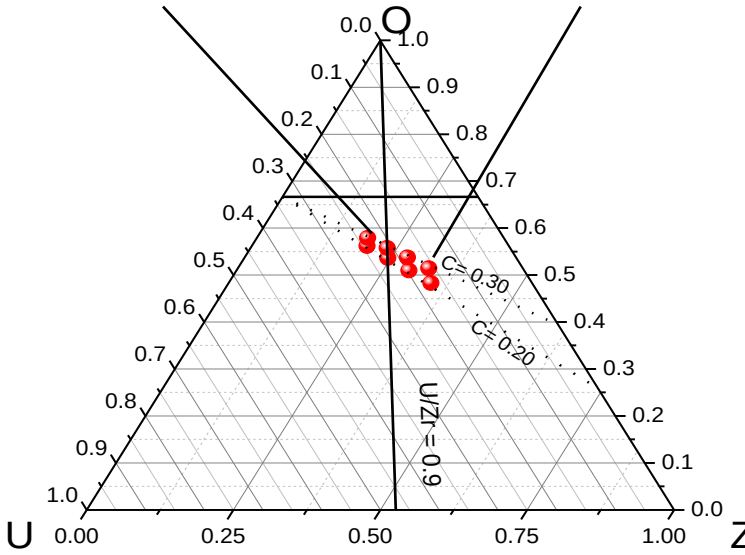
- Test objective was to measure the energetics of spontaneous steam explosion and investigate the characteristics of the premixing zone in stratified melt under water configuration
- The test resulted in a very early spontaneous steam explosion only 0.6 s after the beginning of melt spreading
- The explosion duration was approximately 4 ms, with a peak 175 kN load and a total impulse of 0.42 kN·s, which is comparable, though weaker, than previous explosive PULIMS/SES tests
- The total melt mass released when the explosion occurred was estimated between 0.8 and 1.7 kg
- Preliminary parametric analysis was performed with MC3D for two parameters characterizing the premixed layer - the melt volume fraction and the premixing layer radius
- Best-fit value of a total mass of melt in the premixed layer is around 0.24 kg, which correspond to 17-30 % of the estimated total melt mass released before explosion



FLF experiments on high-temperature behaviour of BWR corium compositions

- The objective is to improve understanding of the quasi-binary section of ternary U-Zr-O diagram in the composition domain of BWR-specific corium composition (U/Zr atomic ratio close to 0.9 and Zr oxidation index close to 20-30%)
- Laser melting/freezing experiments successfully carried out on eight compositions
- Thermal arrests identified
- Spectral emissivity of the surface determined in the visible range, revealing a more metallic behaviour in the liquid and a more oxidic one in the solid surface





U/Zr (mol)	C=ZrO_2/($Zr+ZrO_2$) (mol)	Mol%		
		UO_2	ZrO_2	Zr
0.90	20	47.2	10.6	42.2
0.90	30	47.3	15.7	37.1
1.22	20	55.1	8.9	36.0
1.23	30	55.1	13.6	31.3
0.67	20	40.0	12.0	48.0
0.66	30	39.9	18.1	41.9
0.5	20	33.3	13.3	53.3
0.5	30	33.3	20	46.7

Compositions prepared and analysed in this work

KUBI tests at on assessment of the kinetics of the reaction between UO_2 and B_4C

- Cold crucible load composed of from 2 kg of pellets and powder of depleted UO_2 with 25 g of metallic uranium
- UO_2 melting with a crust on top was achieved under nitrogen flow after less than 20 minutes
- Five B_4C pellets (11 g) were dropped on the pool crust which has then been mechanically broken
- Significant gas release has been observed as interaction started
- After some short generator failures heating was disconnected and corium ingot was extracted from the furnace
- A large porosity was observed and attributed to gaseous releases (mainly CO_2 according to thermodynamic modelling)
- Samples were taken from various ingot locations and prepared for post-test SEM/EDX and XRD analyses

Loaded crucible and molten pool



UO_2 ingot and sampling preparation



MOCKA tests on MCCI with basaltic concrete

- Cylindrical crucibles with 25 cm inner diameter (1D and 2D)
- 12% steel reinforcement
- Melt generation in crucible (thermite burn)
  - metal (Fe)
  - oxide (initially 56 wt% Al_2O_3, 44 wt% CaO)
  - 4 kg Zr at the bottom
- Initial melt temperature ~1840 °C
- Stepwise power input: heating of the oxide phase by addition of thermite briquettes and Zr
- Power deposition into the metallic phase depends on the density ratio, oxide/metal, and the gas release rates from the concrete
- TCs to detect the erosion front

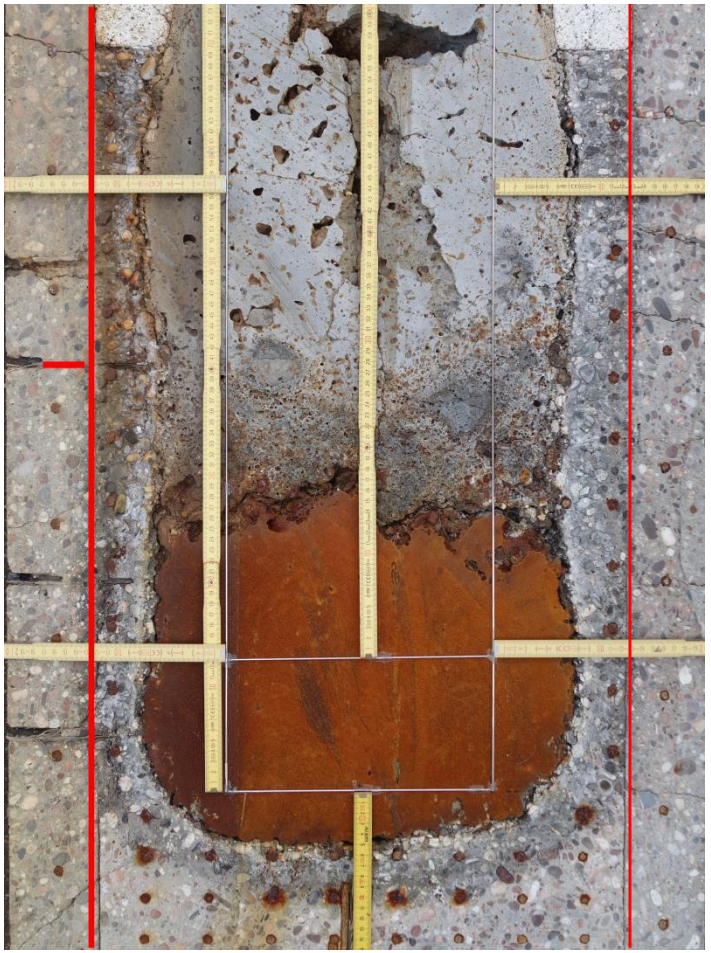


Comparison of downward ablation for siliceous and basaltic concretes with reinforcement



MOCKA SSM
basaltic concrete

MOCKA 5.7
Siliceous concrete



5th NUGENIA-SARNET TA2.1, TA2.2 and SAFEST meetings

April 16-20, 2018



NUGENIA
NUclear GENeration II & III Association

SARnet

Acknowledgments

The authors gratefully acknowledge funding by Euratom to support the work within SAFEST project

Thank you for your attention!

SAFEST ROADMAPS FOR CORIUM RESEARCH

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J. Stuckert[4], F. Belloni[5], P. Vacha[7], Y. Nagae[9], M. Kurata[9], A. Miassoedov[6]
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[8]KRI Saint Petersburg, Russia
[9]JAEA-CLADS, Tomioka, Japan

SAFEST (Severe Accident Facilities for European Safety Targets) is a 48- month EURATOM FP7-supported pan-European Integrated Research Infrastructure Initiative for increased safety of nuclear systems at EU level with 8 partners from 6 European countries. Severe accident with core meltdown is a threat to the containment integrity. Moreover, as Chernobyl and Fukushima accidents have demonstrated, significant release of radioactive products into the environment can cause severe consequences both for people's health and the country's economy. Thus, severe accidents are the focus of considerable research involving substantial human and financial resources worldwide. The research field encompasses too many challenging phenomena, complicated by high temperatures and presence of radioactive materials, so that no individual country has sufficient resources to address all important phenomena within the framework of a national research programme. Therefore it is very important to optimise the use of resources and the collaboration at European and international level. The integration of European severe accident research facilities into a pan-European research infrastructure requires the definition of research roadmaps as guidelines for the common research implementation. The development of such roadmaps has a twofold main goal. First, in the common research framework, investigation priorities and issues should be listed as clearly as possible. Second, shared facilities where studies can be performed addressing precise research priorities and issues should be promptly identified. Conceived in this way, severe accident research roadmaps will steer common research in an efficient way, and they will greatly help the identification of missing research facilities with respect to open scientific questions.

Two such roadmaps have been developed for common Severe Accident Research in the European Union.

The first one, concerning severe accident research applied to existing nuclear power plant systems, has been recently completed and published under the co-ordination of CEA [1]. It helped identify several corium-related high-priority research topics, and also some gaps. For example, it has been concluded that research facilities able to address topics such as core late re-flooding impact on source term and RPV vessel failure mode are still missing in the EU. Another gap lies in the absence of large scale prototypic corium facilities in Europe, although CEA is designing its new large scale facility PLINIUS-2.

The second EU research roadmap, relative to Nuclear Power Plants of the Fourth Generation, is currently being completed under the co-ordination of SCK. It involves the identification of new accidental conditions with respect to more traditional nuclear reactor systems. The objective of the EU Generation IV roadmap consists in assessing the possibility to extend the application field of the SAFEST facilities to the GEN IV reactor technology issues, in particular focusing on the technologies sustained by the Strategic Nuclear Energy Technology Platform (SNETP).

In order to effectively extend the European Severe Accident research approach to international partners, further roadmaps are being developed involving non-EU countries. In collaboration with the Japanese JAEA, KTH is currently completing a EU-Japan severe accident research roadmap. Many common research priorities have already been identified in this initiative, mostly concerning a broad analysis of the 2011 Fukushima Daiichi Nuclear Power Plant accident.
Further international roadmaps are being conceived between the EU and Russia, and between the EU and the USA.

References

1. C. Journeau et al., "safest Roadmap for Corium Experimental Research in Europe", ASCE-ASME Journal of Risk and Uncertainty in Engineering Systems, Part B: Mechanical Engineering 2017, doi: 10.1115/1.4037878



European Commission

European Commission

The European Commission's science and knowledge service

Joint Research Centre



SAFEST ROADMAPS FOR CORIUM RESEARCH

D. Manara[1], D. Bottomley[1], P. Poeml[1], C. Journeau[2], S. Bechta[3], B. Borakov[8], J. Stuckert[4], F. Belloni[5], P. Vacha[7], Y. Nagae[9], M. Kurata[9], A. Miassoedov[6]

[1] JRC Karlsruhe
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[5] SCK-CEN
[6] KIT-IKET
[7] UJV
[8] KRI
[9] JAEA

Joint Research Centre

SAFEST (Severe Accident research Facilities for European Safety Targets) – BACKGROUND

Severe accidents are the focus of considerable research involving substantial human and financial resources worldwide

- too many challenging physical phenomena, complicated further by high temperatures and presence of radioactive materials
- no individual country has sufficient resources (both human and financial) to address all important phenomena in the framework of a national research programme

Requirements for the evaluation of the corresponding risks and update of former evaluations

- uniform use of the best state of knowledge on severe accident phenomenology and qualified computer tools and appropriate methodology
- taking into account notably the inevitable evolutions in reactor operations (new type of fuel higher burn-up extension of plant life new generations of reactors)

SAFEST – BACKGROUND 2

SAFEST (Severe Accident Facilities for European Safety Targets) is a 48 month EURATOM FP7-supported pan-European Integrated Research Infrastructure Initiative for increased safety of nuclear systems at EU level with 8 partners from 6 European countries.

- Development of research roadmaps to focus future European R&D on the stabilisation and termination of severe accidents in PWRs and BWRs
- Establishing the access to SAFEST research infrastructure to investigate all important phenomena from the early core degradation to corium pool formation in the lower head, and ex-vessel melt situations
- Creation of an integrated pan-European laboratory for severe accident research able to address and successfully resolve the wide variety of issues related to severe accident analysis and corium behaviour
- Continuous improvement and upgrading of the SAFEST infrastructure to increase the experimental capabilities and overall quality of R&D to meet current and future challenges
- Applications of the results of the project to the European light water reactors

Eight project partners, coordinated by KIT

Karlsruhe Institute of Technology (KIT, Germany)

Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA, France)

Royal Institute of Technology (KTH, Sweden)

Centre for Energy Research,
Hungarian Academy of Sciences (EK, Hungary)

Institute for Transuranium Elements (ITU, Karlsruhe)

UJV Řež, a.s. (UJV, Czech Republic)

AREVA NP GmbH (ANP, Germany)

Belgian Nuclear Research Centre (SCK, Belgium)

Objectives of the SAFEST SA Research Roadmaps

Producing documents for common severe accident research roadmaps aiming at:

- Establishing coordination activities, enabling the development of a common vision and research roadmaps for the next years.
- Defining research challenges to contribute to further reinforcement of NPP safety.
- Developing a safety research roadmap for next generation plant safety, by taking advantage of the knowledge and expertise obtained for existing reactors as well as specific safety characteristics of considered Gen IV designs (MYRRHA, ALLEGRO, ASTRID).
- Disseminating the European culture of research and innovation as a support to regulatory and industrial decision making in Europe and in other countries.

SAFEST SA Research Roadmaps

CEA (C. Journeau)	**WP2.1** **EUROPEAN CORIUM EXPERIMENTAL RESEARCH ROADMAP**
KTH (S. Bechta, Okamoto, M. Kurata)	**W.P. 2.2** **JOINT POST-FUKUSHIMA EXPERIMENTAL RESEARCH ROADMAP WITH JAPAN**
JRC/KIT (J. Stuckert; A. Miassoedov, D.Bottomley, D. Manara)	**W.P. 2.3** **JOINT EURATOM-ROSATOM EXPERIMENTAL RESEARCH ROADMAP**
SCK/CEA (F. Belloni)	**W.P. 2.4** **EUROPEAN SAFETY RESEARCH ROADMAP FOR NEXT GENERATION PLANTS**
JRC/KIT (A. Miassoedov, D.Bottomley, D. Manara, M. Farmer, M. Corradini)	**EU – USA Roadmap**

WP 2.1 A European corium experimental research roadmap (CEA)

- Objectives:

  Develop a dedicated roadmap for EU experimental R&D on LWR severe accidents issues and corium studies for the next 15 years .
  - based on the research priorities determined by SARNET SARP group as well as those from the newly formed NUGENIA Area 2 on severe accidents.
  - take into account issues identified in the analysis of the European stress tests and from the interpretation of the Fukushima accident.
  - will take advantage of the current and developing European corium infrastructures.
  - define general objectives and specify research needs to reinforce further the safety of NPPs with regard to severe accidents management,
  - detail new R&D challenges and suggest topics to be further developed jointly by the SAFEST consortium, so as to attain a wider common vision.

EU SAR Roadmap: C. Journeau et al.,
ASCE-ASME Journal of Risk and Uncertainty in Engineering Systems, Part B: Mechanical Engineering 2017

http://nuklear-server.nuklear.kit.edu/safest/Dateien/SAFEST-DRR-D2.1_2.pdf

COMMENTS ARE WELCOME!

Ranking of Corium Issues

- **European ranking of Severe Accident Research Priorities**

– SARNET SARP presented at ERMSAR 2013 conference and in Ann. Nucl. Energ., Dec 2014 issue

– SNE-TP Report « Identification of Research Areas in response to Fukushima Daichi Accident »

– NUGENIA TA2 Severe accidents

– In NUGENIA Roadmap 2012 and 2015 editions

- **Selection of high and medium priorities related to Corium issues**
- **Update of the SARP table planned early 2018.**

From SARNET SEVERE ACCIDENT RANKING 2013 (1/2)

High priorities

1) Core and debris coolability and thermal-hydraulics within particulate debris during re-flooding

2) Corium behaviour in a lower head

3) Keeping the RPV integrity by external reactor vessel cooling (ERVC)

4) RPV vessel failure mode and the following corium release from the failed vessel, in the case of vessels with lower head penetrations such as BWRs.

5) Mixing of hydrogen in the containment (and reactor building) atmosphere, possible hydrogen combustion/detonation processes and the influence of countermeasures like Passive Autocatalytic Recombiners (PARs)or the effect of spray systems.

6) Ex-vessel fuel coolant interaction (FCI) including steam explosion (SE)

7) Molten Core Concrete Interaction (oxide-metal configurations; top-flooding)

8) Characterisation and quantification of the fission product (FP) release under oxidation conditions and in particular air ingress for High Burn-Up (HBU) and Mixed Oxide (MOX) fuel.

9) High temperature chemistry in the RCS and its impact on the source term, mainly on iodine species, as well as the chemistry of iodine in the containment and its impact on the source term.

10) Fuel Assembly (FA) behaviour in spent fuel pool severe accident scenarios

11) Development and qualification of specific instrumentation for SA conditions.

Bold characters: *corium related*

From SARNET SEVERE ACCIDENT RANKING (2/2)

Medium priorities

a. Hydrogen generation during re-flooding of strongly degraded cores

b. Integrity of RCS, especially integrity of Steam Generator tubes in High Pressure scenarios

c. Direct Containment Heating (DCH)

d. Long term loss of heat removal from wetwell in a BWR

e. Dry Molten Core Concrete Interaction (with single oxidic phase)

f. Dynamic and static behaviour of containment, crack formation

g. Core re-flooding impact on source term during late phases of core degradation (with highly degraded core loss of geometry)

h. Effect of impurities in water on corium behaviour

i. Data(bases) on corium thermodynamic and thermophysical properties

Bold characters: *corium related*

The 19 CORIUM ISSUES (from these rankings)1/2

European Commission

1) Core and debris coolability and thermal-hydraulics within particulate debris during re-flooding

2) Hydrogen generation during re-flooding of strongly degraded cores

3) Core re-flooding impact on source term during late phases of core degradation (with highly degraded core loss of geometry)

4) Corium behaviour in a lower head

5) Keeping the RPV integrity by external reactor vessel cooling (ERVC)

6) RPV vessel failure mode and the following corium release from the failed vessel, in the case of vessels with lower head penetrations such as BWRs

7) Ex-vessel fuel coolant interaction (FCI) including steam explosion (SE)

8) Direct Containment Heating (DCH)

9) Dry Molten Core Concrete Interaction (with single oxidic phase)

10) Molten Core Concrete Interaction (oxide-metal configurations; top-flooding; liquid source term)

The 19 CORIUM ISSUES (from these rankings)2/2

11) Ex-vessel Core catchers
12) Debris formation and Coolability (in and ex-vessel)
13) Effect of impurities in water on corium behaviour
14) Recriticality in severe accident conditions
15) Spent Fuel Pool scenarios in case of loss of cooling system
16) Development and qualification of specific instrumentation for SA conditions
17) Databases on corium thermodynamic and thermophysical properties
18) Accident Tolerant Fuels
19) Analyses of Fukushima Daiichi corium samples

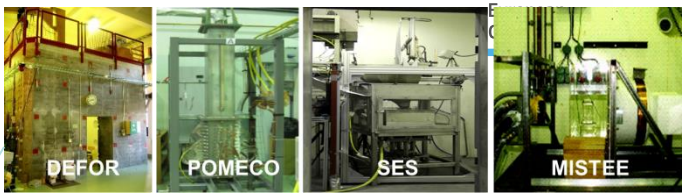
LIST OF Existing/PLANNED facilities

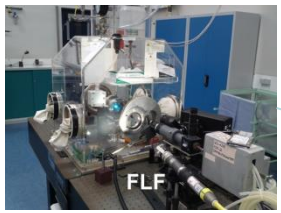
- ***QUENCH***
- ***CODEX***
- ***DEFOR***
- ***PLELUDE***
- ***PEARL***
- ***DEBRIS***
- ***COOLOCE***
- ***POMECO***
- ***LIVE***
- ***RASPLAV***
- ***VITI***
- ***RESCUE***
- ***CERES***
- ***IVR (UJV)***
- ***IVR (CNPRI)***
- ***IVR (SJTU)***
- ***REPEC***
- ***KROTOS***
- ***SES***
- ***DISCO***
- ***VULCANO***
- ***PLINIUS 2***
- ***SICOPS***
- ***HECLA***
- ***CCI - SSWICS***
- ***MOCKA***
- ***CLARA***
- ***BENSON***
- ***COMETA***
- ***ATTILHA***
- ***FLF***
- ***ITU Thermophysical facilities***
- ***LECA-STAR Corium Analysis Lab***
- ***NUCLAB corium Analysis Lab***
- ***LIPC Corium Analysis Lab***

E.U. facilities + International facilities to which European organizations have some access

SAFEST (Severe Accident Facilities for European Safety Targets): the project for a pan-EU SA research laboratory

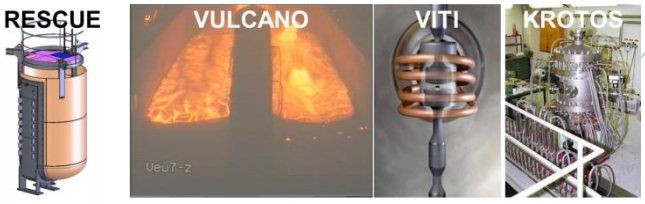
















Synthesis Table (1/2)

Issue Number	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
	Core and debris Coolability	Hydrogen generation during late core re-flooding	Core late re-flooding impact on source term	Corium behaviour in a lower head	External Reactor Vessel Cooling	RPV vessel failure mode and corium release	DCH	Ex-vessel FCI	Dry oxidic-pool MCCI	Steel, Flooding MCCI	Core Catchers	Debris Formation and coolability	Effect of Water Impurities	Recriticality in Severe Accident conditions	SFP scenarios	SA Instrum.	Thermo-dynamic and thermos-physical databases	Accident Tolerant Fuels	Fukushima corium samples
QUENCH	X	X											X		X			X	
CODEX	X	X											X		X			X	
DEFOR								X				X	X	X					
JEBRA								X				X		X					
PRELUDE	X											X	X						
PEARL	X											X	X						
DEBRIS												X	X						
COOLOCE	X											X	X						
POMECO	X											X	X						
LIVE				X		X						X							
RASPLAV3				X															
VITI				X					X	X			X				X	X	

Synthesis Table (2/2)

Issue Number	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
	Core and debris Coolability	Hydrogen generation during late core re-flooding	Core late re-flooding impact on source term	Corium behaviour in a lower head	External Reactor Vessel Cooling	RPV vessel failure mode and corium release	DCH	Ex-vessel FCI	Dry oxidic-pool MCCI	Steel, Flooding MCCI	Core Catchers	Debris Formation and coolability	Effect of Water Impurities	Recriticality in Severe Accident conditions	SFP scenarios	SA Instrum.	Thermo-dynamic and thermos-physical databases	Accident Tolerant Fuels	**Fukushima corium samples**
RESCUE					X								X						
CERES					X								X						
IVR (CNPRI)					X								X						
IVR (UJV)					X								X						
REPEC II					X								X						
KROTOS								X					X				X		
PLINIUS 2								X	X	X	X	X	X			X			**X**
MISTEE								X					X						
SES								X			X		X						
DISCO							X	X											
VULCANO				X					X	X	X					X	X		**X**
SICOPS									X	X	X								
HECLA									X	X									
CCI-SSWICS									X	X									
MOCKA									X	X	X								
CLARA									X	X									
BENSON					X						X								
COMETA									X	X	X		X				X		
ATTILHA				X					X								X		
FLF													X	X			X	X	
ITU analysis lab																	X		**X**
LECA.STAR																			**X**
NUCLAB																	X		**X**
LIPC																	X		
Need for new facility			**O**			**O**													

GAPS - NEEDS

- **2 issues without dedicated experimental facilities**

– **Core late re-flooding impact on source term**

– **RPV vessel failure mode and corium release**

There are needs to prepare experimental programs, either in EU or through experimental collaboration, to provide data to the on-going and future R&D activities.
In some cases, as RPV failure and corium release, there are important experimental and analytical challenges preventing realization of R&D programs tackling the issues.

- **Another gap lies with the absence of large scale prototypic corium facilities in Europe**

– Closure of COMAS, FARO in the late 1990s, early 2000s

– Scaling effect and material effects

– CEA is designing its new large scale facility PLINIUS-2 (next slide)

Summary EU Roadmap

- **Thanks to SAFEST European project a roadmap is being issued that links research priorities to European and international experimental facilities in the corium domain**
- **After Fukushima Daiichi accident, the research priorities have been reevaluated in various European groups.**
- **19 issues of high or medium importance have been listed and described**
- **34 experimental facilities have been considered**
  - 29 in the European Union
  - 5 international facilities
- **Cross analysis between research needs and facilities**
  - 3 domains lacking experimental facilities in Europe
  - Need for a large scale prototypic corium facility in Europe: PLINIUS-2

EU – Japan Roadmap (KTH - JAEA)

The newly founded CLADS research centre (Collaborative Laboratories for Advanced Decommissioning Science, located in Ibaraki and Tomioka, Japan), which will deal with many research issues underpinning the Fukushima-Daiichi site decommissioning, will join SAFEST and its activities in late 2017 or early 2018. Cooperation between SAFEST and CLADS will certainly lead to further improvement of the EURATOM-Japan research roadmap.

Other international (OECD-NEA) projects have been launched in order to perform joint research in support of the Fukushima Daiichi site management, namely TCOFF (Thermodynamic Characterisation Of Fuel debris based on scenario analysis of severe accident progression at Fukushima Daiichi), ARC-F (Analysis of Information from Reactor Building and Containment Vessel and water sampling of Fukushima Daiichi NPS) and Pre-ADES (Preparatory Study on Analysis of Fuel Debris).
These projects are most likely to interact with SAFEST for the joint EURATOM-Japan roadmap conception.

Prof. Okamoto and Dr. Kurata are the JAEA contact people for SAFEST.
Some of the CLADS experimental facilities may be offered in trans-national access to SAFEST participants.

EU – Japan Roadmap (KTH - JAEA)

There are several activities related to the Fukushima accident, prioritized in a the July 2017 CLADS workshop:

- **Study of the simulated and the actual Fukushima debris**, including experimental studies in EU hot cells, **and characterization of corium properties;**
- Testing of various methods of **debris sampling, characterization and reprocessing;**
- Separate-effect experiments to study various l**ocal phenomena during accident progression of Fukushima units #1-3**. (Generally from core degradation to MCCI, including FP release/transport/scrubbing, B_4C effects on core degradation and molten pool behaviour);
- Development and validation of models with the experimental data collected.

EU – RUSSIA roadmap (JRC, KIT)

Some important issues have been encountered in the development of a Joint EURATOM-ROSATOM experimental research roadmap.

Russia carries out much severe-accident related research (cf. for example the ISTC projects).

However the Russian colleagues the Joffe institute together with KTH Sweden made an application to the Ministry of Education and Science of the Russian Federation for a joint performance of a Russian-European Road-map in the area of experimental research of energy –expensive processes at NPP's. This would have been linked to the SAFEST project. Although ideal for our purposes we learnt in October 2016 that the project had not been ranked high enough for funding and so it will be problematic for the Russian colleagues to continue.

Nevertheless a roadmap report might be finalised by February 2018 based on existing material, particularly on the ISTC CEGSAM work carried out in the severe accident field until 2013.


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Severe accident research in the core degradation area: An example of effective international cooperation between the European Union (EU) and the Commonwealth of Independent States (CIS) by the International Science and Technology Center

D. Bottomley[a,*], J. Stuckert[b], P. Hofmann[b], L. Tocheny[c], M. Hugon[d], C. Journeau[e], B. Clément[f], S. Weber[g], S. Guentay[h], Z. Hozer[i], L. Herranz[j], A. Schumm[k], F. Oriolo[l], E. Altstadt[m], M. Krause[n], M. Fischer[o], V.B. Khabensky[p], S.V. Bechta[q,1], M.S. Veshchunov[r], A.V. Palagin[b,2], A.E. Kiselev[s], V.I. Nalivaev[s], A.V. Goryachev[t], V. Zhdanov[u,3], V. Baklanov[u]

Retrospective: strengths of cooperation with Russia in framework of International Science and Technology Center ISTC (1992 – 2012)

- Single purpose international organization on the ground;
- Capacity to bring together best expertise worldwide via its extensive network;
- Capacity to handle effectively multilateral funds for research projects and support programs;
- Capacity to carry out commercialization and innovation projects;
- Capacity to adapt to changing circumstances.

Activities which can be suggested to ROSATOM from EU within the SAFEST project

- Use of SAFEST infrastructure for experimental research of national needs of Russia (VVER, Gen IV systems)
- Join planning for complimentary development of new experimental methodologies
- Experimental verification of severe accident code capabilities
- Analysis of Chernobyl samples (also useful for a deeper understanding of Fukushima).
- Benchmarks

Collaboration JRC- Khlopin Radium Institute (KRI) St. Petersbourg within IAEA agreement (Coordinated Research Project T13015)

European Commission

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Study of a "hot" particle with a matrix of U-bearing metallic Zr: Clue to supercriticality during the Chernobyl nuclear accident

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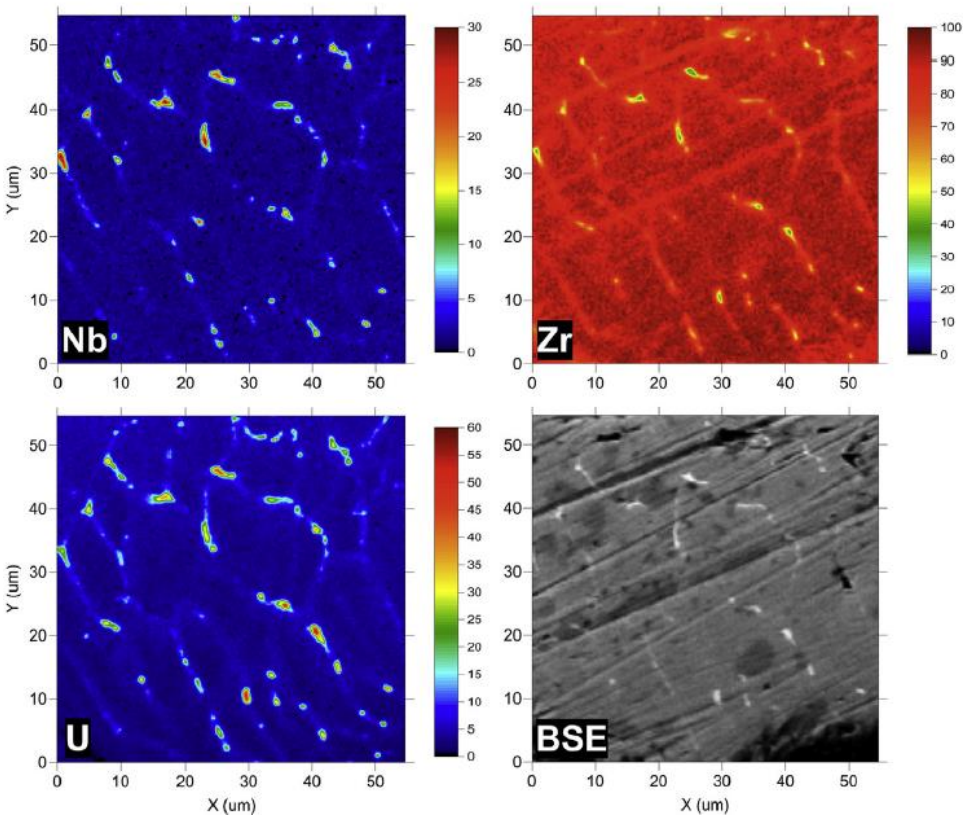
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ABSTRACT

This paper is dedicated to the 30th anniversary of the severe nuclear accident that occurred at the Chernobyl NPP on 26 April 1986. A detailed study on a Chernobyl "hot" particle collected from contaminated soil was performed. Optical and electron microscopy, as well as quantitative x-ray microbeam analysis methods were used to determine the properties of the sample. The results show that the particle (≈240 x 165 μm) consists of a metallic Zr matrix containing 2–3 wt. % U and bearing veins of an U,Nb admixture. The metallic Zr matrix contains two phases with different amounts of O with the atomic proportions $(U,Zr,Nb)_{0.73}O_{0.27}$ and $(U,Zr,Nb)_{0.61}O_{0.39}$. The results confirm the interaction between UO_2 fuel and zircaloy cladding in the reactor core. To explain the process of formation of the particle, its properties are compared to laboratory experiments. Because of the metallic nature of the particle it is concluded that it must have formed during a very high temperature (> 2400 °C) process that lasted for only a very short time (few microseconds or less); otherwise the particle should have been oxidised. Such a rapid very high temperature process indicates that at least part of the reactor core could have been supercritical prior to an explosion as it was previously suggested in the literature.

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P. Pöml – B. Borakov: BSE image of UO_2 Zr Nb hot particle found in contaminated Chernobyl soil. The fact that Zr is hardly oxidised suggests that this particle was formed during early fuel-cladding interaction at high temperature (> 2700 K) before the NPP explosion. This fact could be interpreted in the light of the occurrence of supercriticality in the very early stages of the accident.

EU GEN 4 Roadmap (SCK, CEA)

A European safety research roadmap for next generation plants is currently being developed under the lead of the Belgian SCK, with the co-operation of the Czech UJV and French CEA.

A final report about this roadmap will be delivered at the end of 2017.

The objective of the EU Generation IV consists in assessing the possibility to extend the application field of the SAFEST facilities to the GEN IV reactor technology issues, in particular focusing on the technologies sustained by the Strategic Nuclear Energy Technology Platform (SNETP).

The SAFEST partners expressed their assessment over selected phenomena (which are yet not fully characterized) of the core degradation and post-accident phases of severe accidents.

GENERATION 4 SAFETY

Safety and Reliability of the future generation of reactor designs are met by the achievement of specific goals:

1. **Generation IV nuclear energy systems will excel in operational safety and reliability.** The focus of this goal applies to safety and reliability during normal operation of all facilities employed in the nuclear fuel cycle, and thus, deals with the relatively likely kinds of operational events that set the forced outage rate, determine worker safety, and result in routine emissions that could affect workers or the public.

2. **Generation IV nuclear energy systems will have a very low likelihood and degree of reactor core damage.** This goal calls for design features that create high confidence that the possibility of core damage accidents will be very small for Generation IV reactors. The goal deals with both minimizing the frequency of initiating events, and with provision of design features that ensure that the plants can successfully control and mitigate any initiating events that might occur without causing core damage.

3. **Generation IV nuclear energy systems will eliminate the need for offsite emergency response.** It is desirable that Generation IV systems demonstrate, with high confidence, the capability of the safety architecture to manage and mitigate the consequences of severe plant conditions and that any potential releases of radiation will be small and have only insignificant public health consequences.

→ SA Research in Gen IV addresses issues often different than those studied in earlier NPPs.

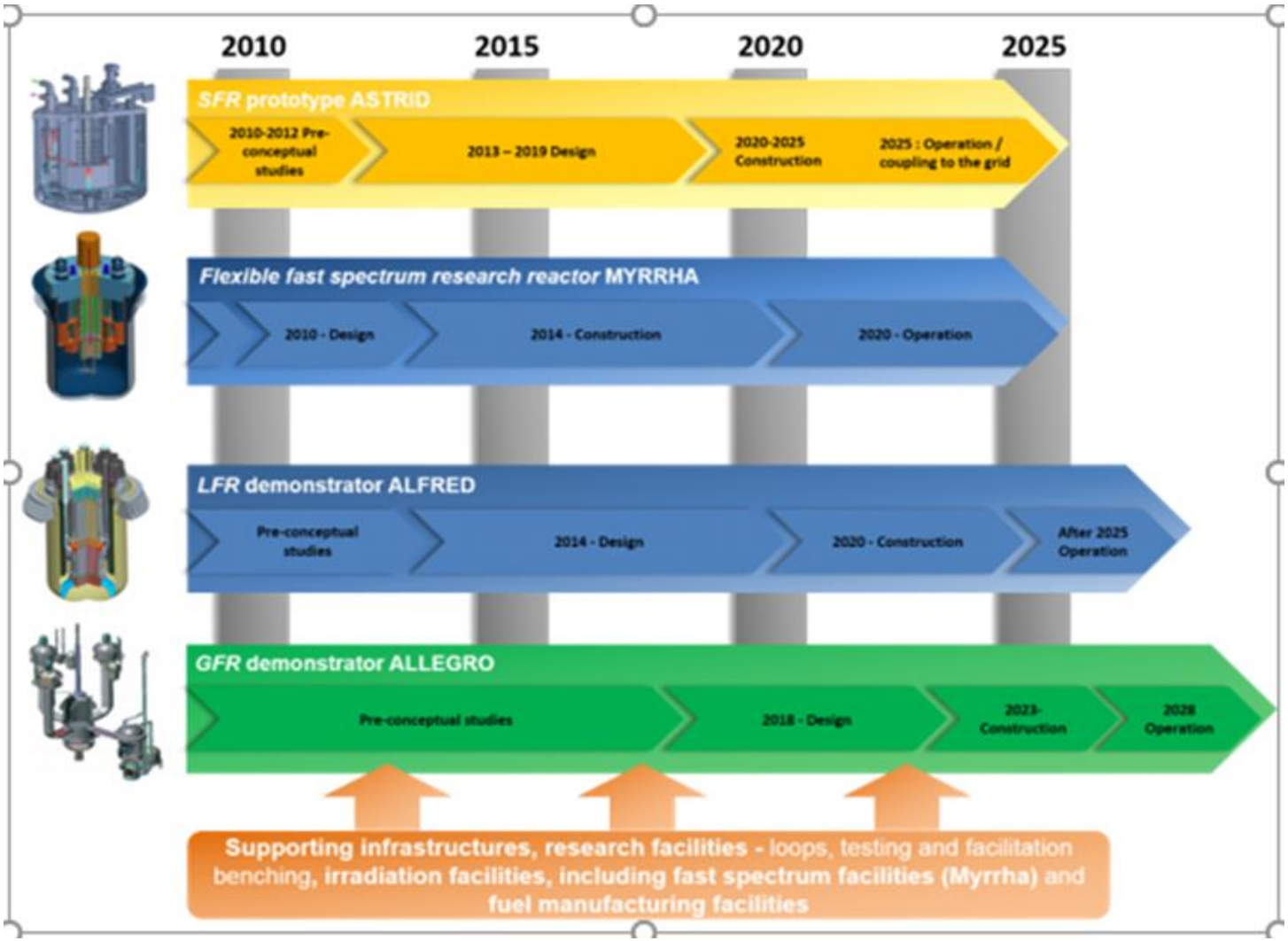
SNETP GENERATION 4 PROJECTS

SNETP has prioritized the different Gen-IV systems and proposed to develop the following projects:

- The **ASTRID** project, a prototype sodium-cooled fast reactor (SFR), that will be built in Marcoule, France.
- The **ALLEGRO** project, a gas-cooled fast reactor (GFR) demonstrator to be constructed in the Czech Republic, Hungary or Slovakia.
- The **ALFRED** project, a lead-cooled fast reactor (LFR) demonstrator that will be built in Pitesti, Romania
- Additionally, the **MYRRHA** research reactor project, a lead-bismuth cooled accelerator-driven fast neutron multi-purpose research reactor, is also being developed. Since MYRRHA is based on the heavy liquid metal technology, it will strongly contribute to the development of LFR technology. MYRRHA will play the role of European technology pilot plant (ETPP) in the roadmap for LFR.

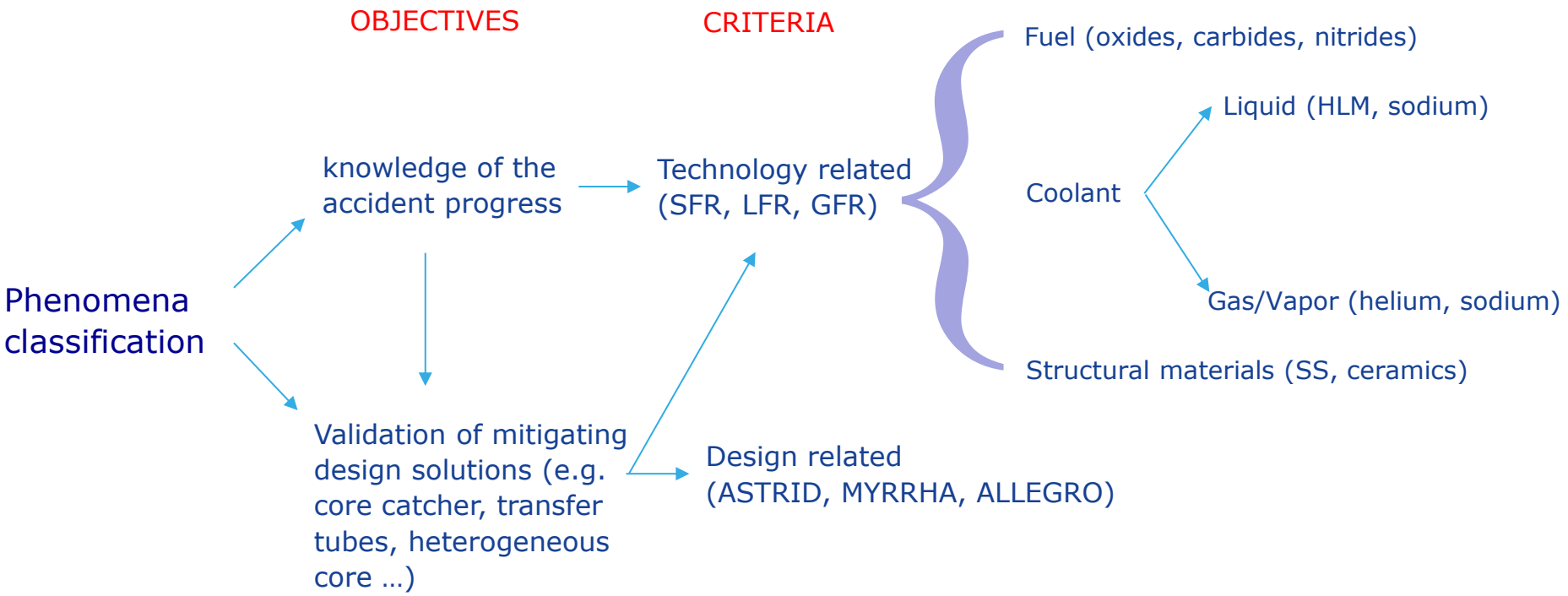
SNETP has set up a Task Force comprising research organizations and industrial partners to develop the European Sustainable Nuclear Industrial Initiative (ESNII)

SNETP GENERATION 4 PROJECTS



GENERATION 4 Severe Accident Research

Phenomena classification: technology and design dependency



The identification of common phenomenology is possible, when one or more features are shared by the different technologies (MOX/SS pin used both for SFRs and LFRs)

Design related phenomena depend not only of the type of technology, but also on the specific reactor design (e.g. ASTRID vs PHÉNIX)

GENERATION 4 Severe Accident Research

Phenomena classification: severe accident phases



Envelope of initiators

Core degradation phase

- Pin failure (overpower, loss of flow)
- Fuel dispersion and relocation inside a Fuel Assembly → Fuel Coolant Interaction, fuel/steel freezing and blockages
- Damage propagation inside a FA → Degraded pool region dynamics (H&M transfer)
- FA wrapper failure → Wrapper melt-through, FCI
- Interassembly propagation → FCI, fuel/steel freezing and blockages, degraded pool region dynamics

Post-accident heat removal phase

- Fuel dispersion outside the core (mitigating devices)
- FCI in the plenum region → Pre-mixing and fragmentation, pressure buildup
- Molten fuel - core catcher interaction (core catcher design)
- Debris bed coolability → HT convection, debris bed levelling
- Decay heat removal (DHR systems design) → HT convection

EU – USA SAR Roadmap

Not foreseen as a SAFEST deliverable → Work is ongoing

Main Contact people in the USA:

M. Corradini - University of Wisconsin, Madison, Wisconsin;
M. Farmer - Argonne National Laboratory, Lemont, Illinois.

Mainly based on recent assessment work

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United States Department of Energy Severe Accident Research Following the Fukushima Daiichi Accidents

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NUCLEAR SCIENCE AND ENGINEERING · VOLUME 184 · 293–304 · NOVEMBER 2016

Reactor Safety Gap Evaluation of Accident-Tolerant Components and Severe Accident Analysis

M. T. Farmer,[a*] R. Bunt,[b] M. Corradini,[c] P. Ellison,[d] M. Francis,[e] J. Gabor,[f] R. Gauntt,[g] C. Henry,[h] R. Linthicum,[i] W. Luangdilok,[h] R. Lutz,[j] C. Paik,[h] M. Plys,[h] C. Rabiti,[k] J. Rempe,[l] K. Robb,[e] and R. Wachowiak[m]

ASCE-ASME Journal of Risk and Uncertainty in Engineering Systems, Part B: Mechanical Engineering | Volume | Issue | research-article

research-article

SAFEST ROADMAP FOR CORIUM EXPERIMENTAL RESEARCH IN EUROPE

Christophe Journeau, Viviane Bouyer, Nathalie Cassiaut-Louis, Pascal Fouquart, Pascal Piluso, Gérard Ducros, Stéphane Gossé, Christine Guéneau, Andrea Quaini, Beatrix Fluhrer, Alexei Miassoedov, Juri Stuckert, Martin Steinbrueck, S Bechta, Pavel Kudinov, Wei Min Ma, Bal Raj Sehgal, Zoltan Hozer, Attila Guba, Dario Manara, David P. Bottomley, Manfred Fischer, Gert Langrock, Holger Schmidt, Monika Kiselova and Jiri Zdarek

CONCLUSIONS - PERSPECTIVES

The conception of joint international SA research roadmaps will disseminate the European culture of research and innovation as a support to regulatory and industrial decision making in Europe and in other countries.

1. A European corium experimental research roadmap has been defined and published. Research priorities and gaps have been clearly identified.

2. A joint Fukushima experimental research roadmap is being developed with Japan. Several joint projects are being carried out in parallel to SAFEST. JAEA-CLADS has recently joined the SAFEST consortium.

3. A Joint EURATOM-ROSATOM experimental research roadmap has been envisaged, although the development of this roadmap seems to be stuck at the moment. Nonetheless, several common SA research initiatives have been jointly carried out in the last few years between EU and Russian institutions.

4. A European safety research roadmap for next generation plants is being developed. Phenomena addressed in this roadmap are often different than those relevant to earlier NPPs.

5. A joint EU-USA SA research roadmap has been envisaged. Fruitful contacts have been created in 2016 and 2017 in order to develop it.

ENEN PLUS:
description and objectives

- **Partly "follow-up" to GENTLE.**
- **Education and Training project accepted and financed within Horizon 2020.**
- **Kick-off meeting October 2nd-3rd 2017, Brussels. Duration: 3 years.**

Main OBJECTIVES

- **Attract** new talents to careers in nuclear.
- **Develop** the attracted talents within and beyond academic curricula.
- Increase the **retention** of attracted talents.
- **Involve** the nuclear stakeholders within EU and beyond.
- **Sustain** the revived interest for nuclear careers.

ENEN PLUS:
Student mobility objectives

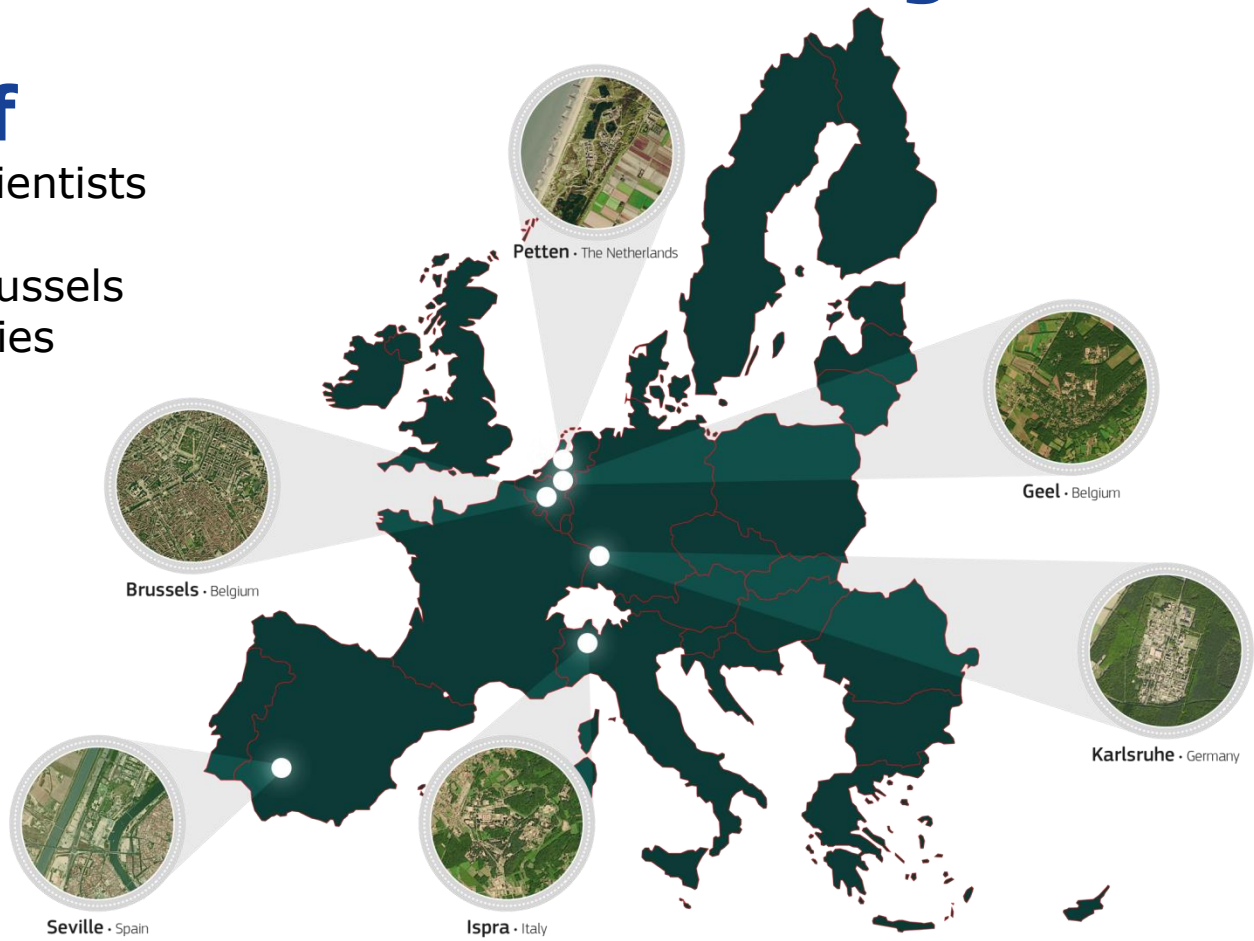
The mobility support will focus on the following target groups and activities:

- M.Sc. students:
  - mid-term internships (several months);
  - presentations at conferences;
  - exchange compatible with the rules of the European Masters of Nuclear Engineering (EMSNE, >=20 ECTS abroad);
- Ph.D. students and post-docs:
  - Internships (up to 24 months);
  - access to research infrastructures;
  - access to EURATOM research projects;
  - ➢ *In principle, all students enrolled in EU universities can participate. Calls are opened also to Russian and Japanese students (ENEN partners).*
  - ➢ *The first call for mobility money is expected to be out early 2018. The first mobility actions could be expected around May 2018.*
  - ➢ *ENEN will manage the contractual part for each student.*
  - ➢ **MORE DETAILS WILL COME WITH CALL FOR APPLICATIONS.**

The Joint Research Centre at a glance

3000 staff

Almost 75% are scientists and researchers.
Headquarters in Brussels and research facilities located in
5 Member States.



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Development of Three-Dimensional Simulation Code for BWR In-vessel Core Degradation

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After the severe accident at the Fukushima Daiichi Nuclear Power Plants, an in-vessel core degradation code of Boiling Water Reactor (BWR) has been developed by Regulatory Standard and Research Department, Secretariat of Nuclear Regulation Authority (S/NRA/R). The code aims at applying to BWR core degradation assessments from temperature rising of the core to molten corium settling on the bottom of the Reactor Pressure Vessel (RPV). The key physical models have were incorporated into the code: (a) multi phases, components and velocity fields, (b) multiple melting points and chemical interactions, (c) candling phenomenon, (d) melt blockage on the grid spacers, and (e) three-dimensional geometry in the vessel. The code consists of the neutronic diffusion calculation module (based on SKETCH), the thermal-hydraulic calculation module and the fuel pin behavior calculation module (based on FEMAXI-6). The three modules communicate with each other through a "control module" and data are exchanged among the three modules in optimized time steps. The modules have been assessed in simplified geometries and calculation conditions. The physical models of in-vessel core degradation phenomena have been assessed against the DF-4 at Sandia National Laboratories (SNL) and the QUNECH-06 at Karlsruhe Institute of Technology (KIT) in terms of estimating the cladding temperature escalation and hydrogen generation, and the CORA-16 and 18 at KIT in terms of estimating axial- and lateral-directional motion of molten corium. The models of metallic melt relocation around the core support plate, and molten corium breakup in the lower head have been validated by the XR-2 at SNL, and the FARO L-14, -19 and -20 and the KROTOS K-37 and -45 at Joint Research Centre (JRC) facilities. Furthermore, exploratory analyses have been carried out for a typical middle-power-level BWR with initial core conditions as variable parameters: initial water level, decay heat, radial power shape, and fuel cladding condition. The present paper describes the introduction of the code system, the progress of verification and validation, and the result of exploratory analyses.

Keywords: Core degradation, BWR, Simulation code, Three-dimensional geometry

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Development of Three-Dimensional Simulation Code for BWR In-vessel Core Degradation



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Contents

1. Background
2. Objective and Targets
3. Outline and Verification of Code System
4. Validation Progress
5. Simulation Geometries and Exploratory Analyses
6. Summary

1. Background

- Core degradations at TEPCO's Fukushima Daiichi Nuclear Power Station Units 1–3 in March 11, 2011

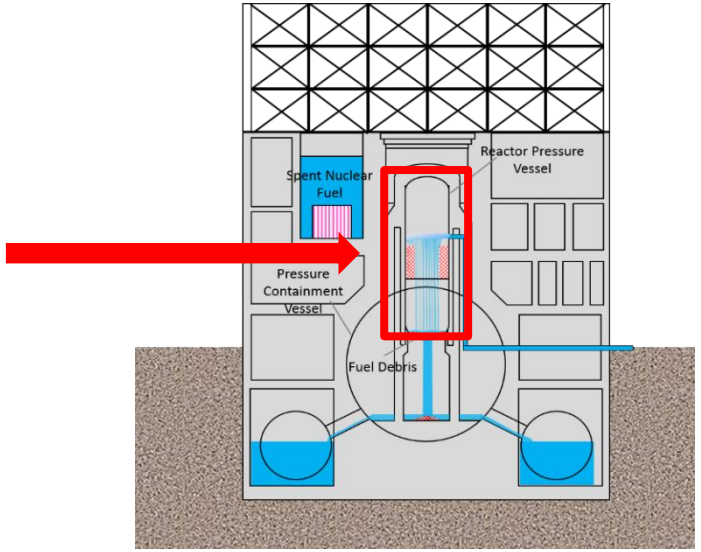
- Taking into account the lessons learned from the severe accidents, S/NRA/R has started development of a detailed code of in-vessel core degradation for BWRs as fundamental safety research.

2. Objective & Targets

- To simulate and understand phenomena of BWR in-vessel core degradation with using the NRA detailed code.
- Calculation Zone:
  i. Core
  ii. Around core support plate
  iii. Lower (& upper) plenum.
- Calculation Stage:

  From normal operation

  to severe accident conditions.

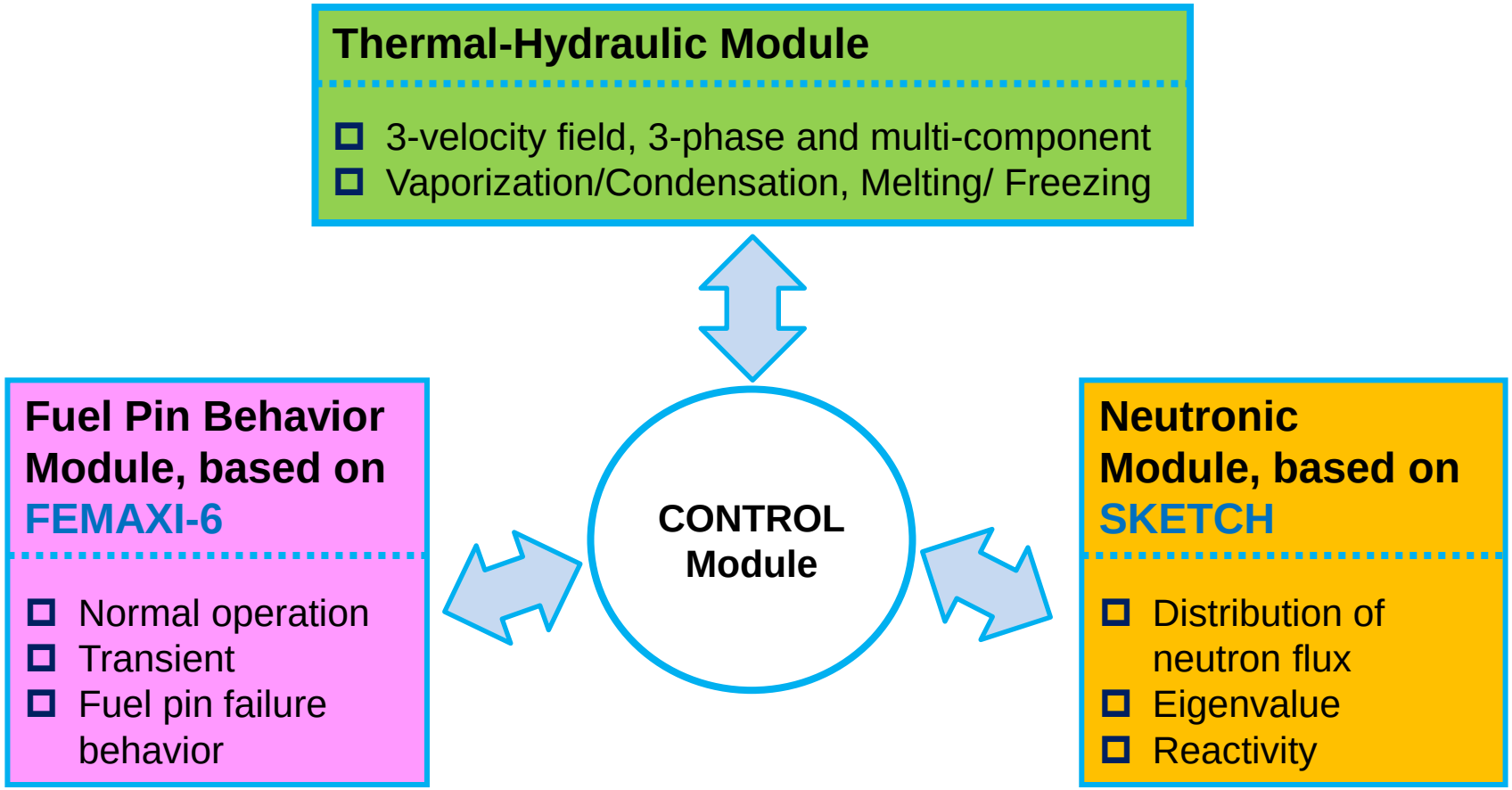


- Required Functions:
  i. Tight coupling calculation of the neutronic, thermal-hydraulic and fuel pin behaviors
  ii. Three-dimensional geometry
  iii. Multiple melting and chemical interactions.

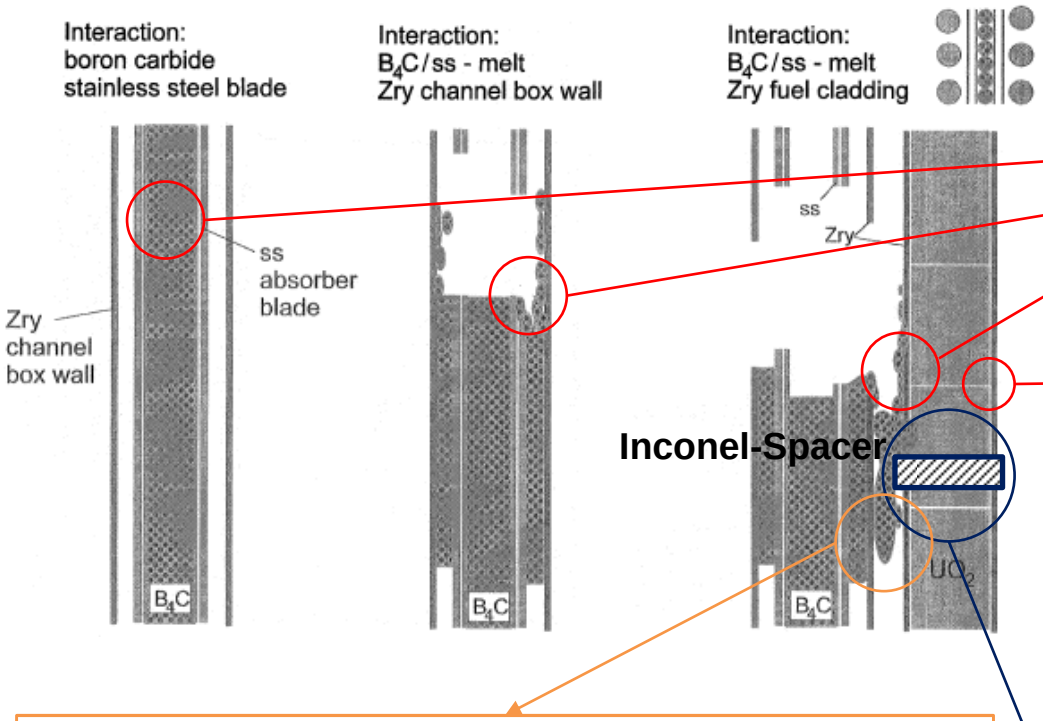
3. Outline and Verification of Code System

Outline of Code System

- The Code consists of the "Thermal-Hydraulic", the "Fuel Pin Behavior" and the "Neutronic" Modules.
- Three modules communicate with each other through the "CONTROL" Module.



Important Physical Models of Core Degradation Phenomena



Reaction Zone Growth Rate Model

The following chemical reactions are incorporated in the "Thermal-Hydraulic Module":

- **B_4C-SUS eutectic**
- **B_4C-SUS-Zry melt formation**
- **B_4C-SUS-Zr-UO_2 melt formation.**

The UO_2-Zry melt formation is incorporated in the "Fuel Pin Behavior Module":

- **UO_2-Zry melt formation.**

In calculation, heaviest materials will represent these complicated molten materials in a calculation cell, due to the limited material data.

Candling Model

The Candling model is incorporated in the "Thermal-Hydraulic Module":

- For increasing heat transfer coefficient and friction effect between rod and liquid metal, **Binary contact area is adjusted** in order to enhance the interaction between rod and liquid metal.

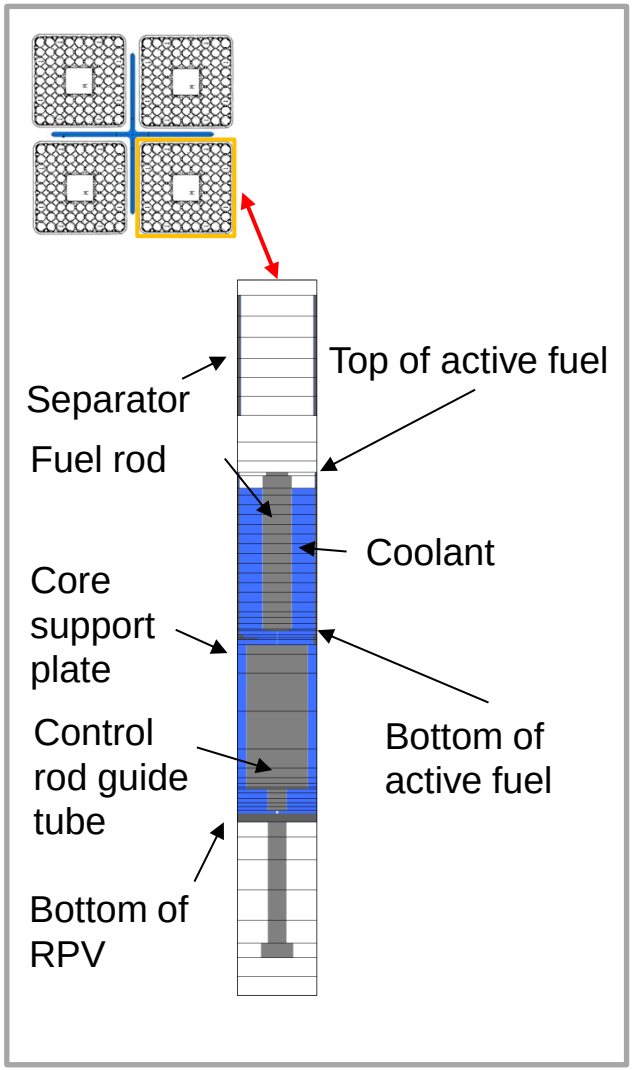
Grid Spacer Model

The reaction between grid spacer spring and Zry cladding is incorporated in the "Thermal-Hydraulic Module":

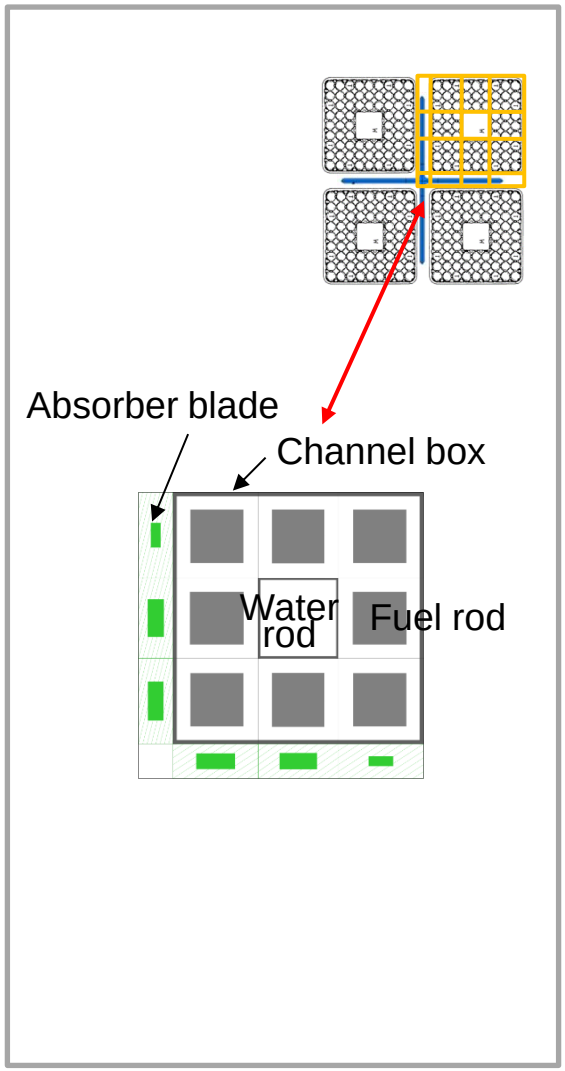
- **Zry-Inconel eutectic.**

Ref) S. Hagen et al, Impact of Absorber Rod Material on Bundle Degradation Seen in CORA Experiments, FZKA-5680 (1996)

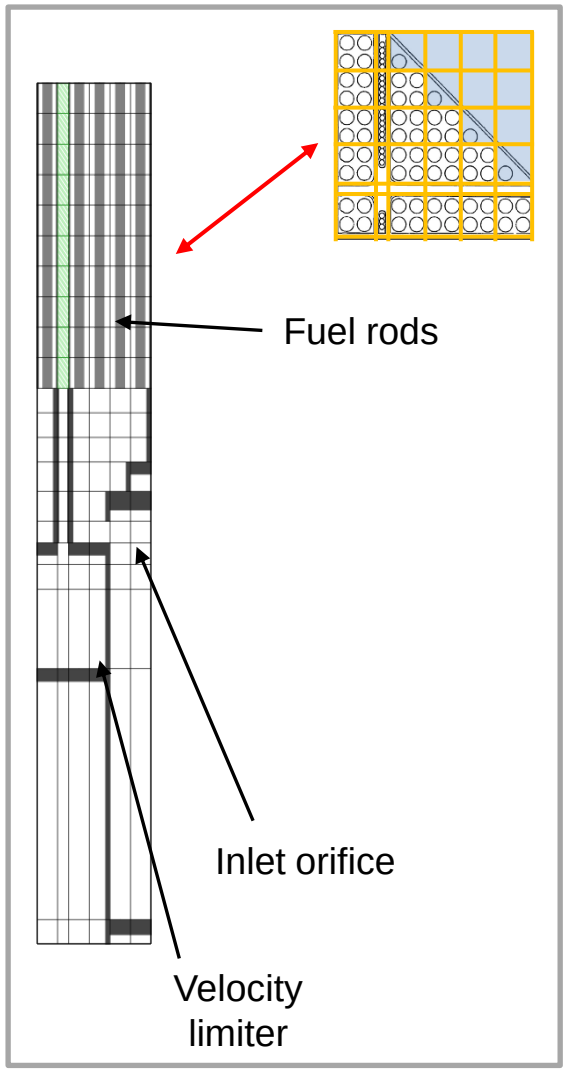
Outline of Three Simulation Geometries



Single BWR Fuel Assembly Equivalent Geometry

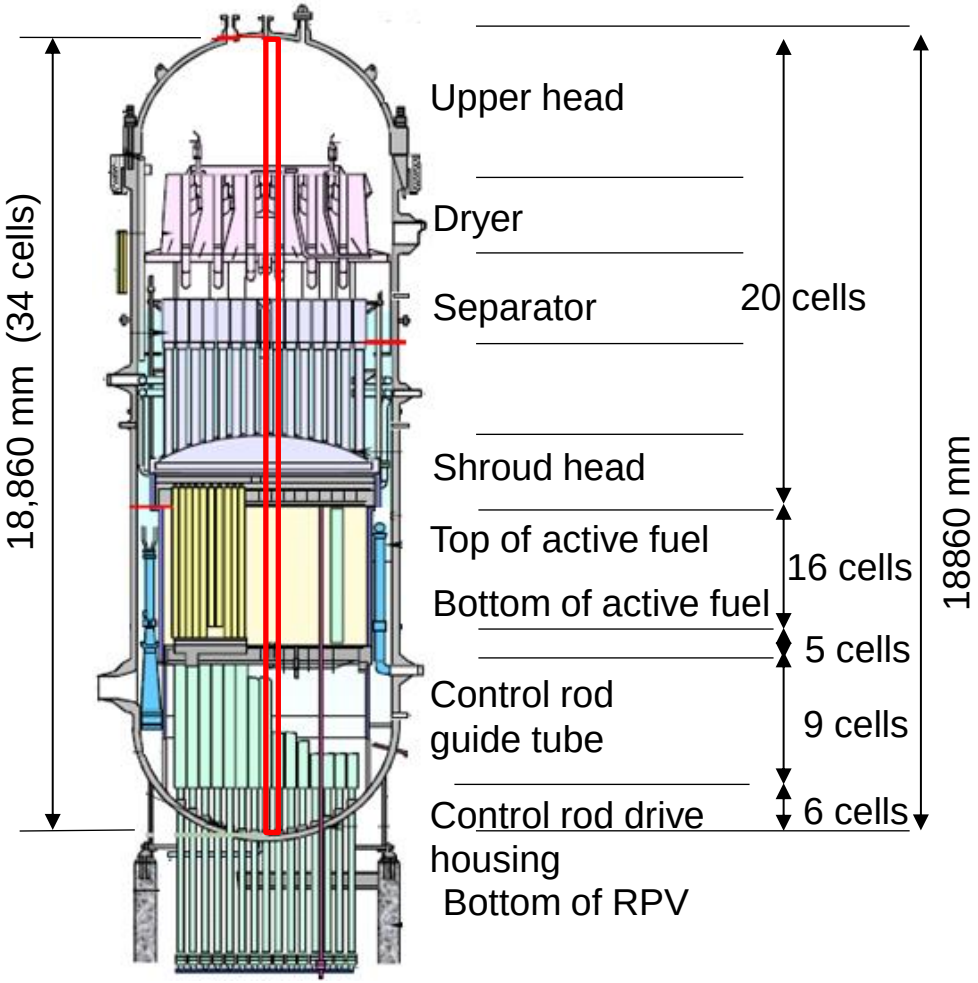


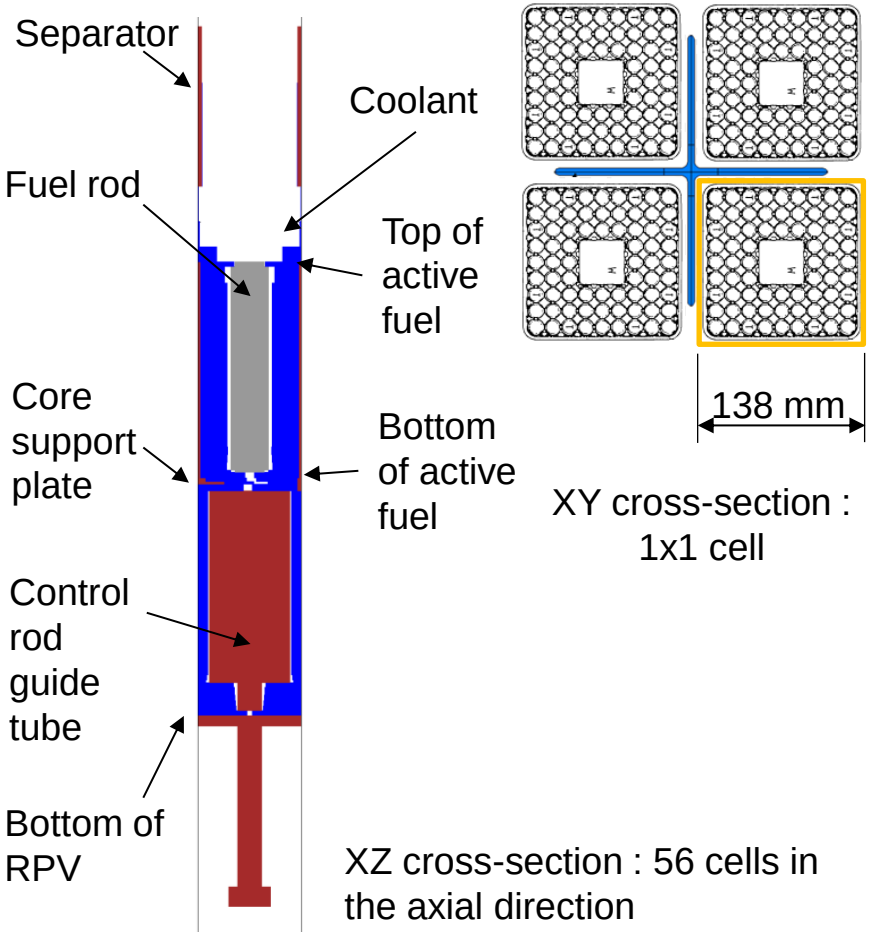
Fine 9x9 Fuel Pins Assembly Geometry



Core Support Geometry

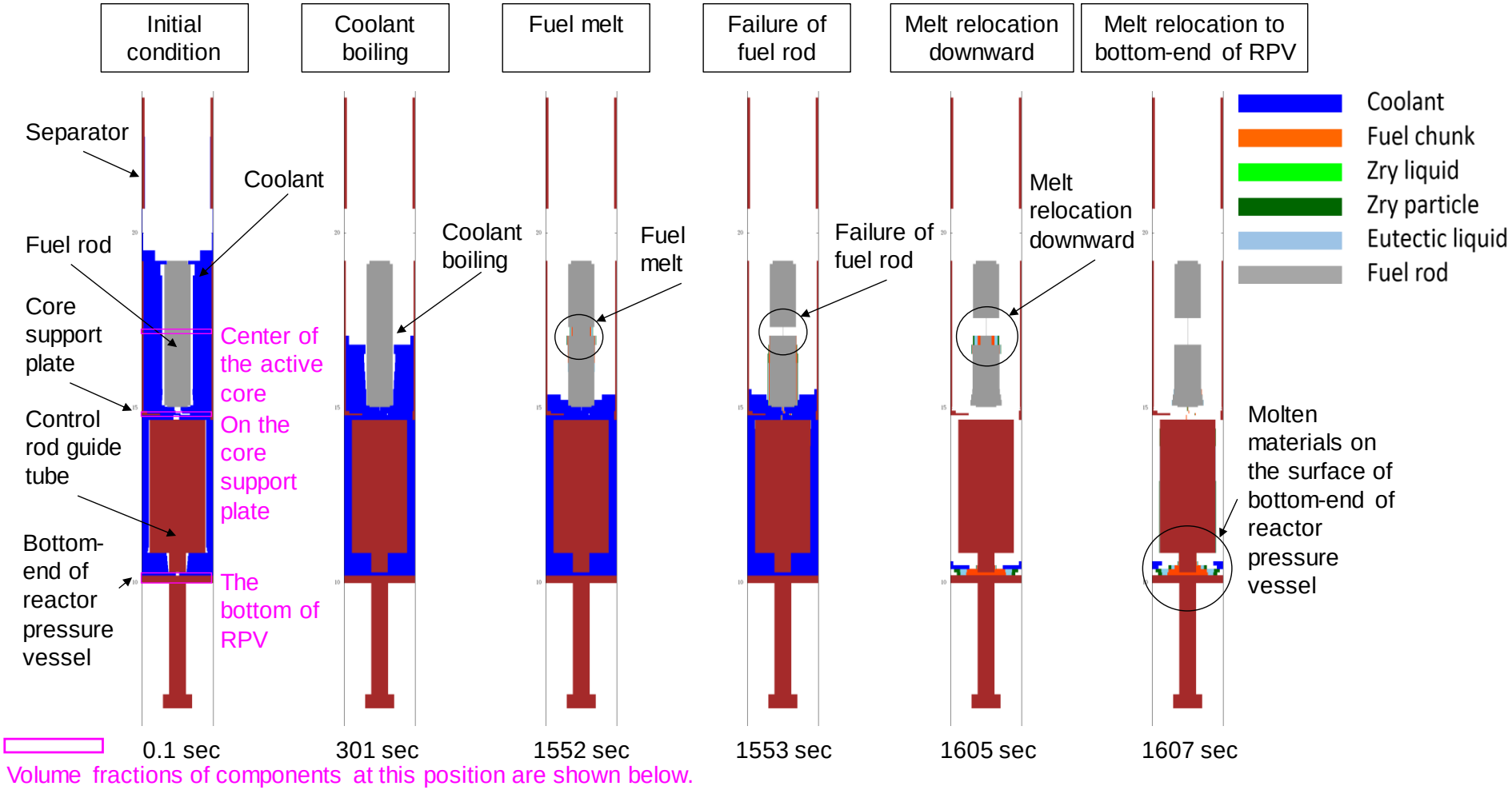
Verification of Tight Coupling Simulation for Single BWR Fuel Assembly: Calc. Geometry





Verification of Tight Coupling Simulation for Single BWR Fuel Assembly: Calc. Result (cont'd)

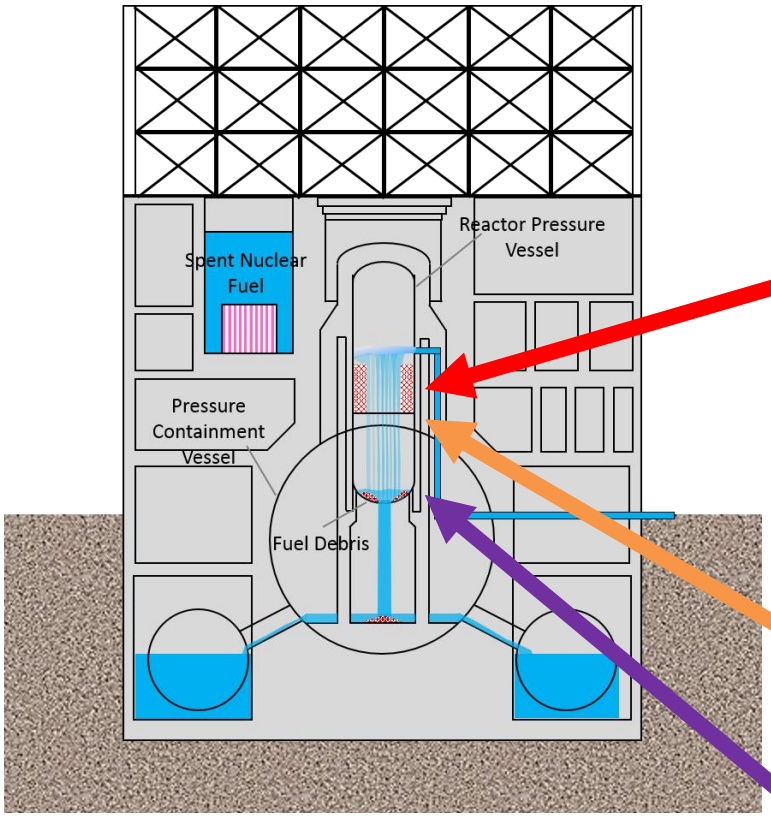
*The reactor was shut down at the starting time of this calculation. Hence, the decay heat is very high.



4. Validation Progress:

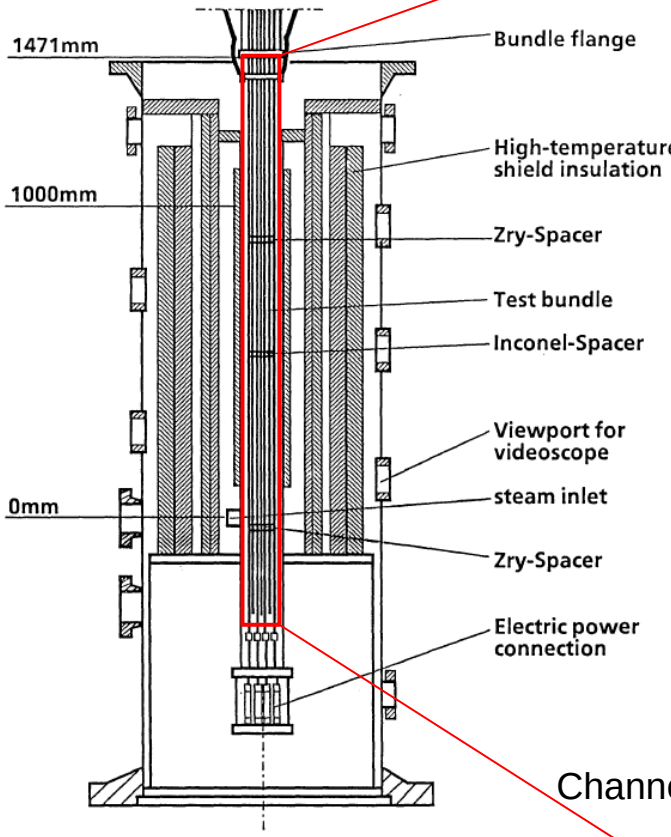
Validation for Axial- and Lateral-directional motions of molten corium of CORA-18

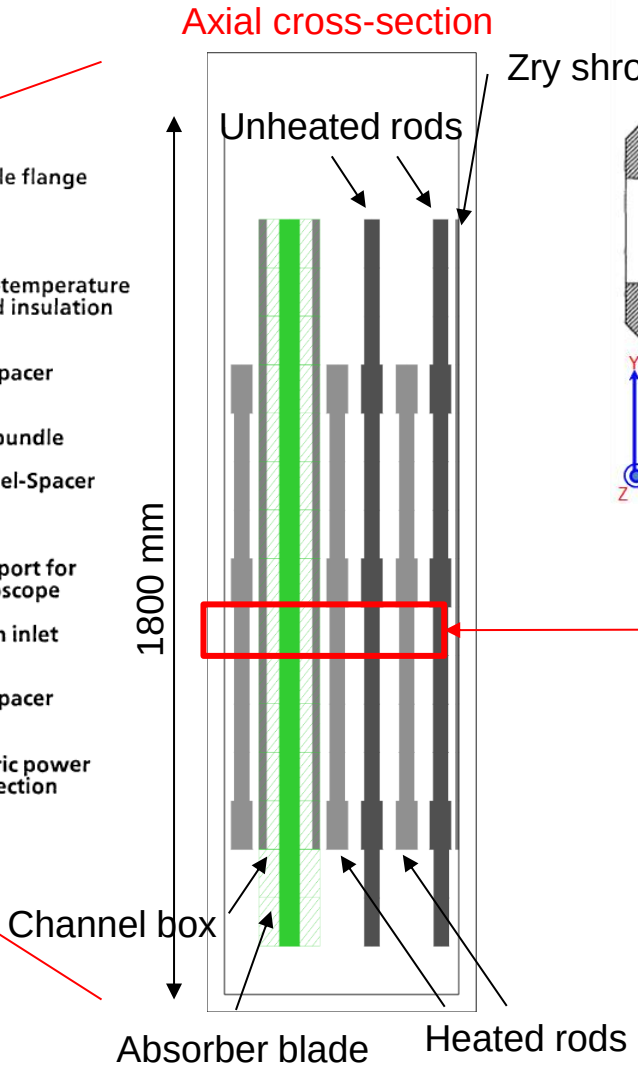
Validation for Existing Severe Accident Experiments

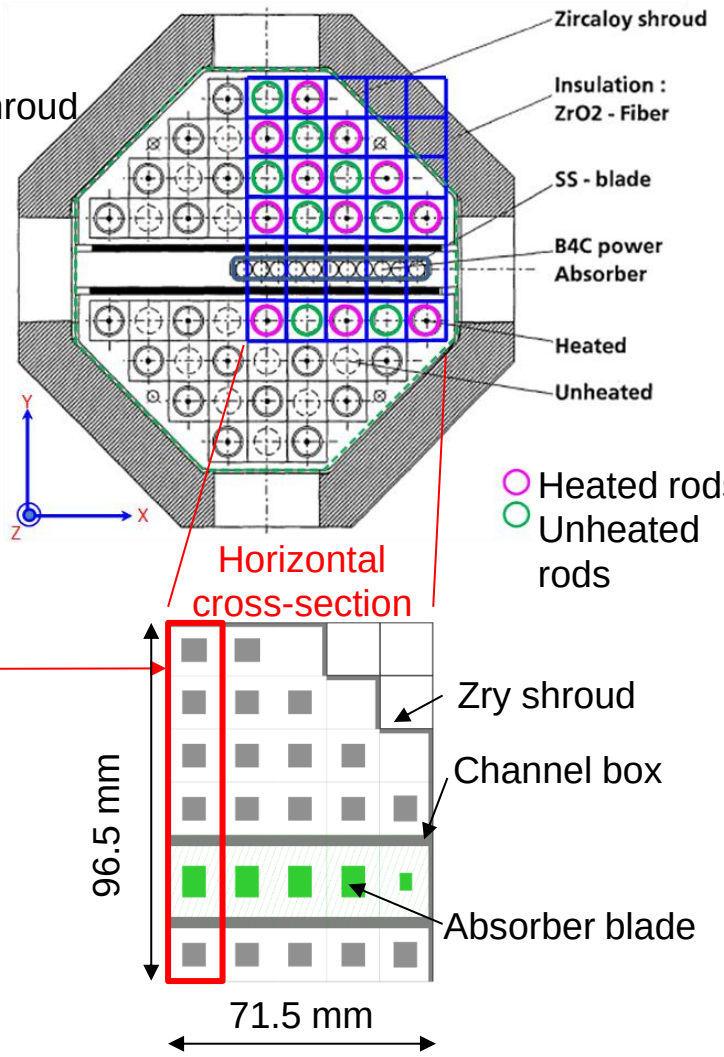


Phase	Validation Target	Program	Lab.
Core Degradation	Cladding temperature escalation & hydrogen generation	DF-4	SNL
		QUENCH-06	KIT
	Axial- and lateral-directional motion of molten corium	CORA-16 CORA-18	FZK
Melt flow around core support plate		XR-2	SNL
Molten corium interaction with water pool in RPV lower head		KROTOS K-37 & -45	JRC
		FARO L-14, -19 & -20	JRC

Validation for CORA-18: Calc. Geometry

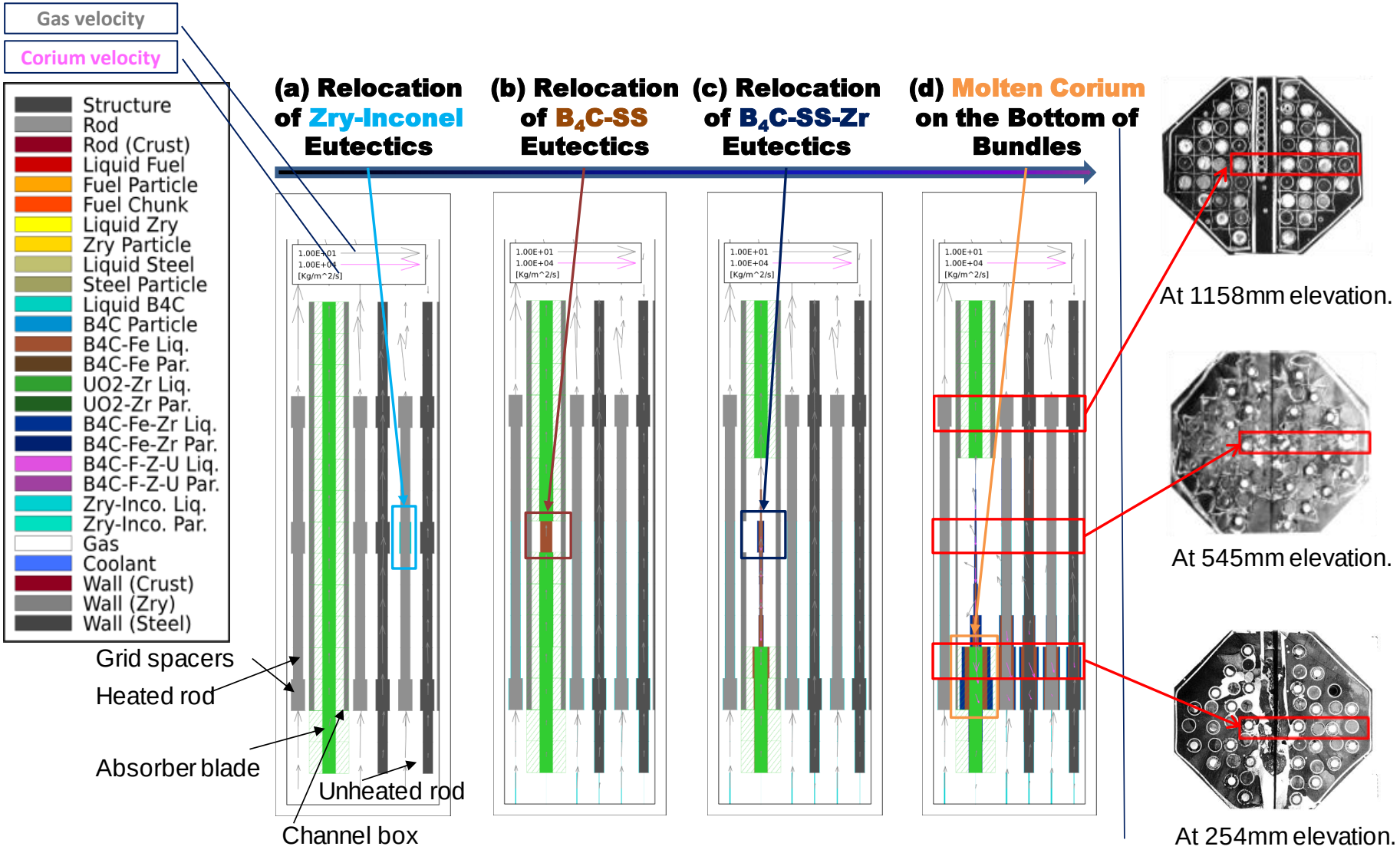






CORA Bundle Arrangement **Axial Cross-Section: 18 Cells** **Horizontal Cross-Section: 5x6 Cells**

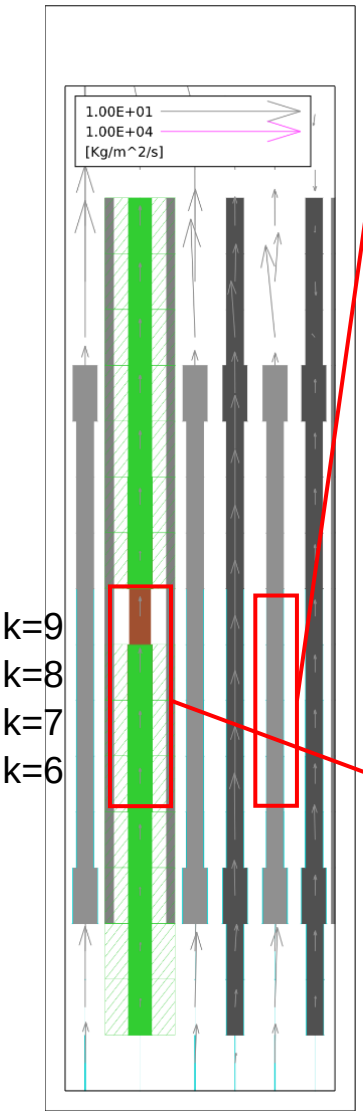
Validation for CORA-18: Axial Relocation



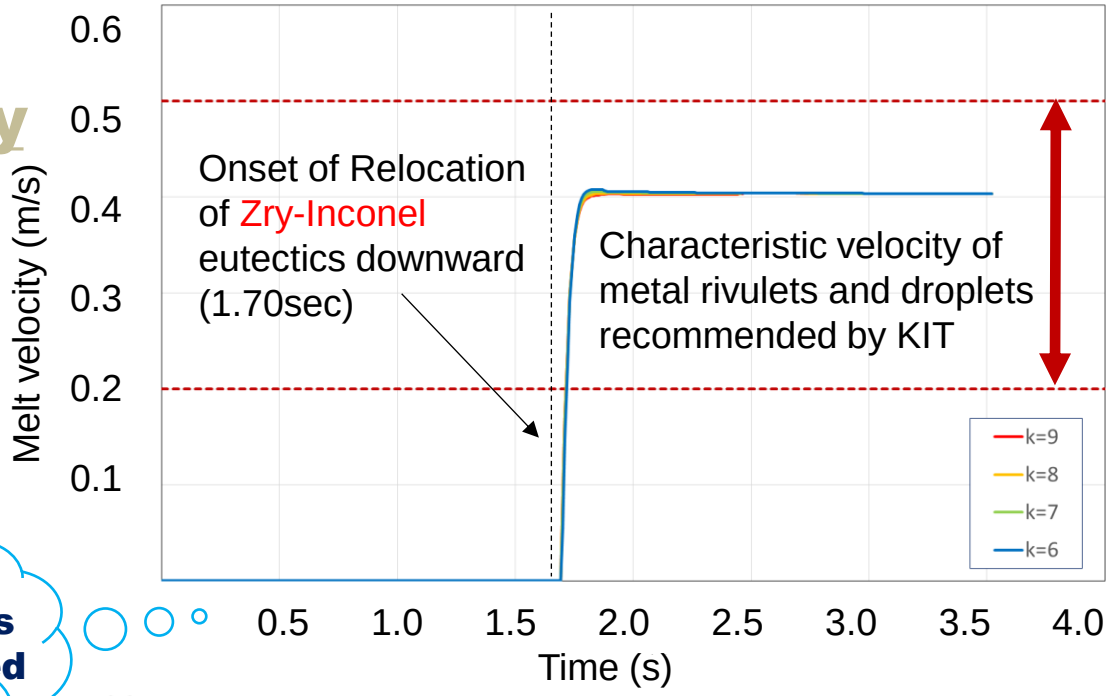
Calculation results for axial relocation in CORA-18 experiment.

Horizontal cross-section of experimental results

Validation for CORA-18: Axial Relocation Velocity

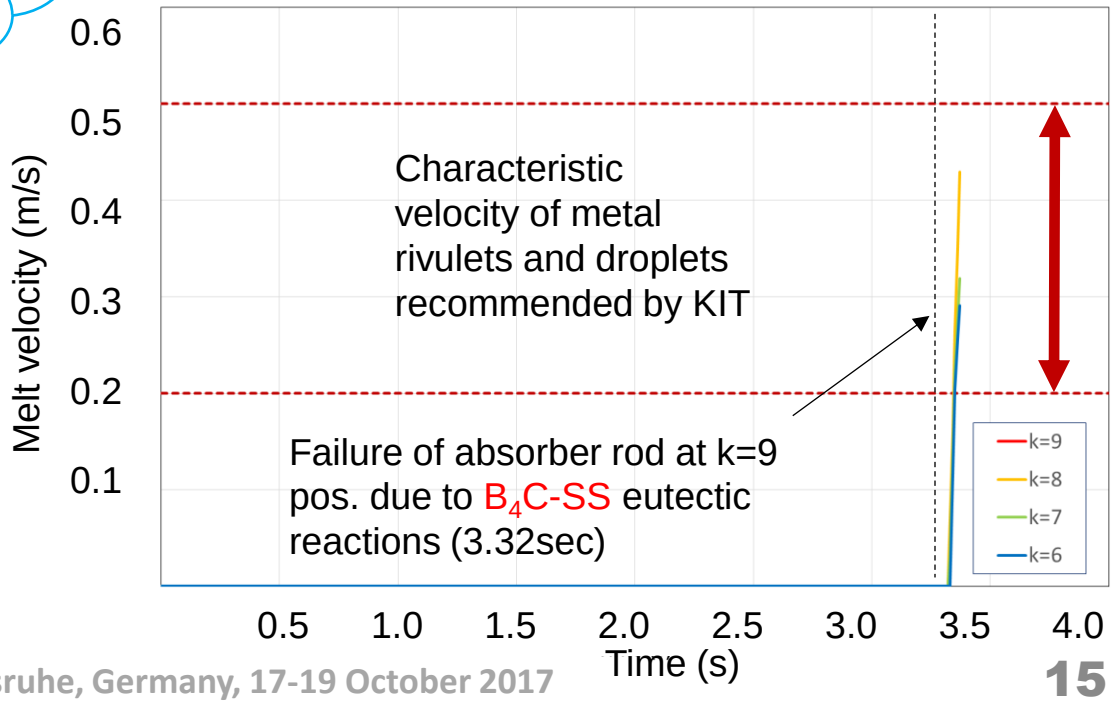


Melt Velocity Downward along the Heated Rod



0.2 m/s - 0.5 m/s is recommended by KIT.

Melt Velocity Downward along the Absorber Rod



5. Exploratory Analyses

For applications to simulation of core degradation in the future,

To access sensitivities of core degradation to various initial conditions.

Exploratory Analyses: Calculation Conditions & Initial Parameters

☐ Calculation Conditions:

i. Geometry: ¼ Sector-Core
ii. Simulation time: 250s
iii. Decay heat level: 10% of thermal output.

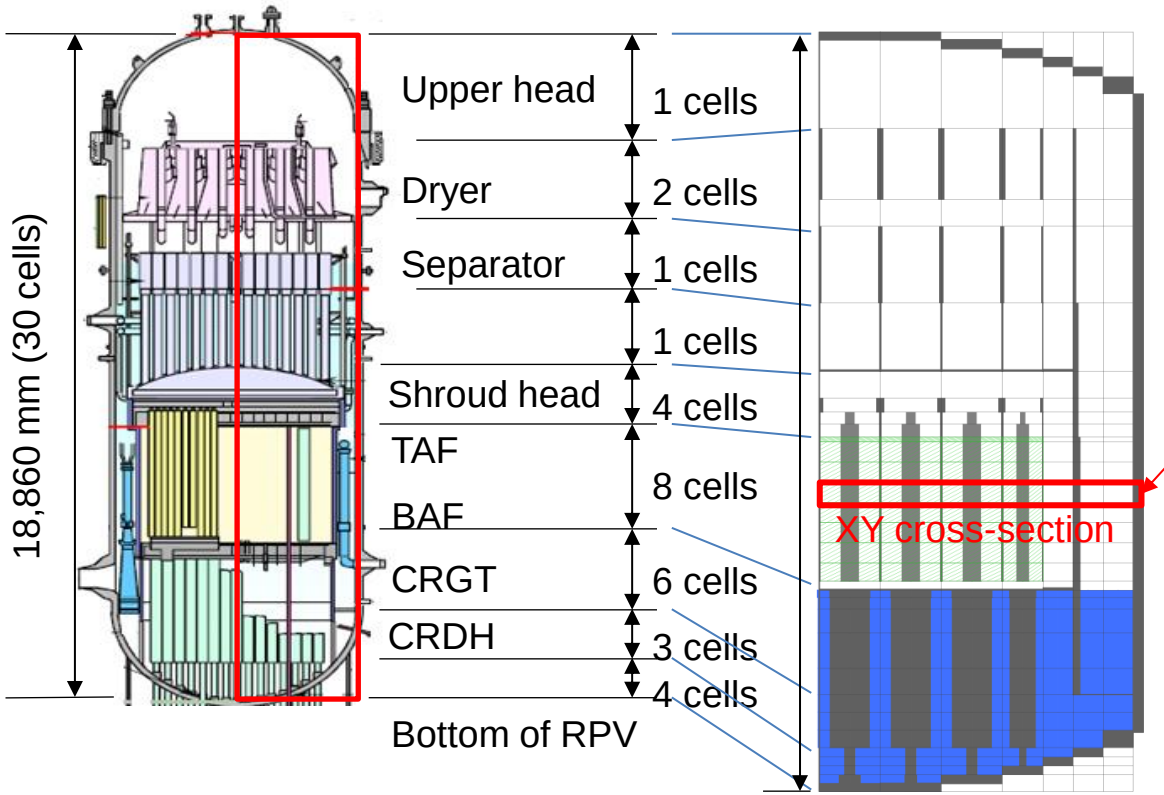
☐ Initial Parameters:

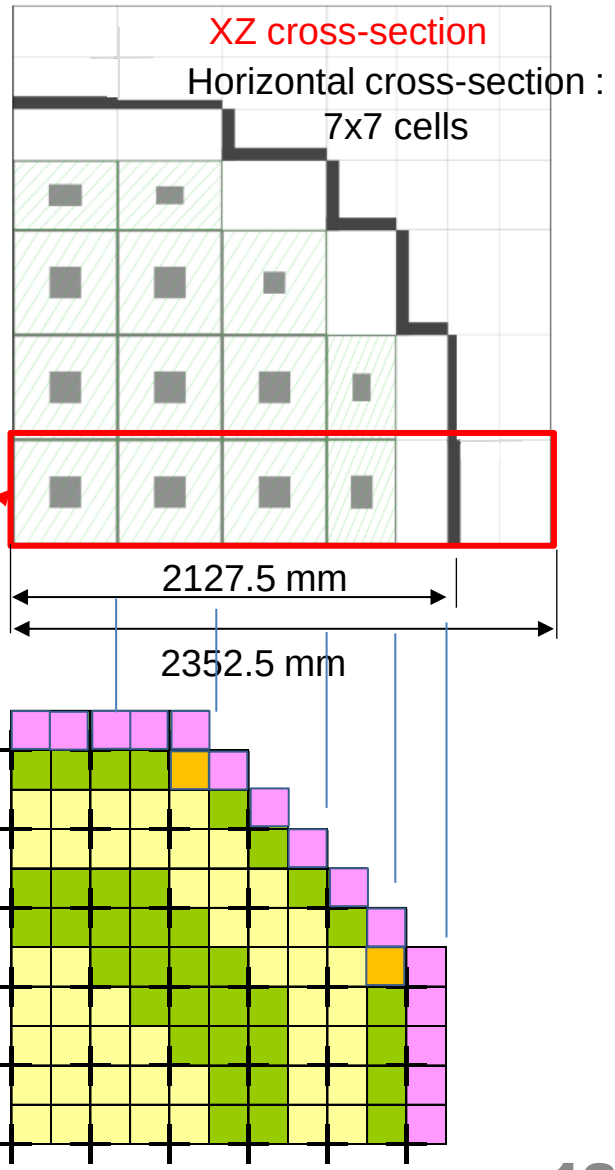
To quantify sensitivities of initial parameters for the BWR core degradation phenomena, the following parameters were selected.

i. Initial water level (BAF*/TAF)
ii. Decay heat (10% of thermal output*/Way-Wigner equation)
iii. Radial power shape (Cosine*/ Flat)
iv. Oxidation of fuel cladding (Depleted*/Fresh fuel)

*Reference case: Very high power

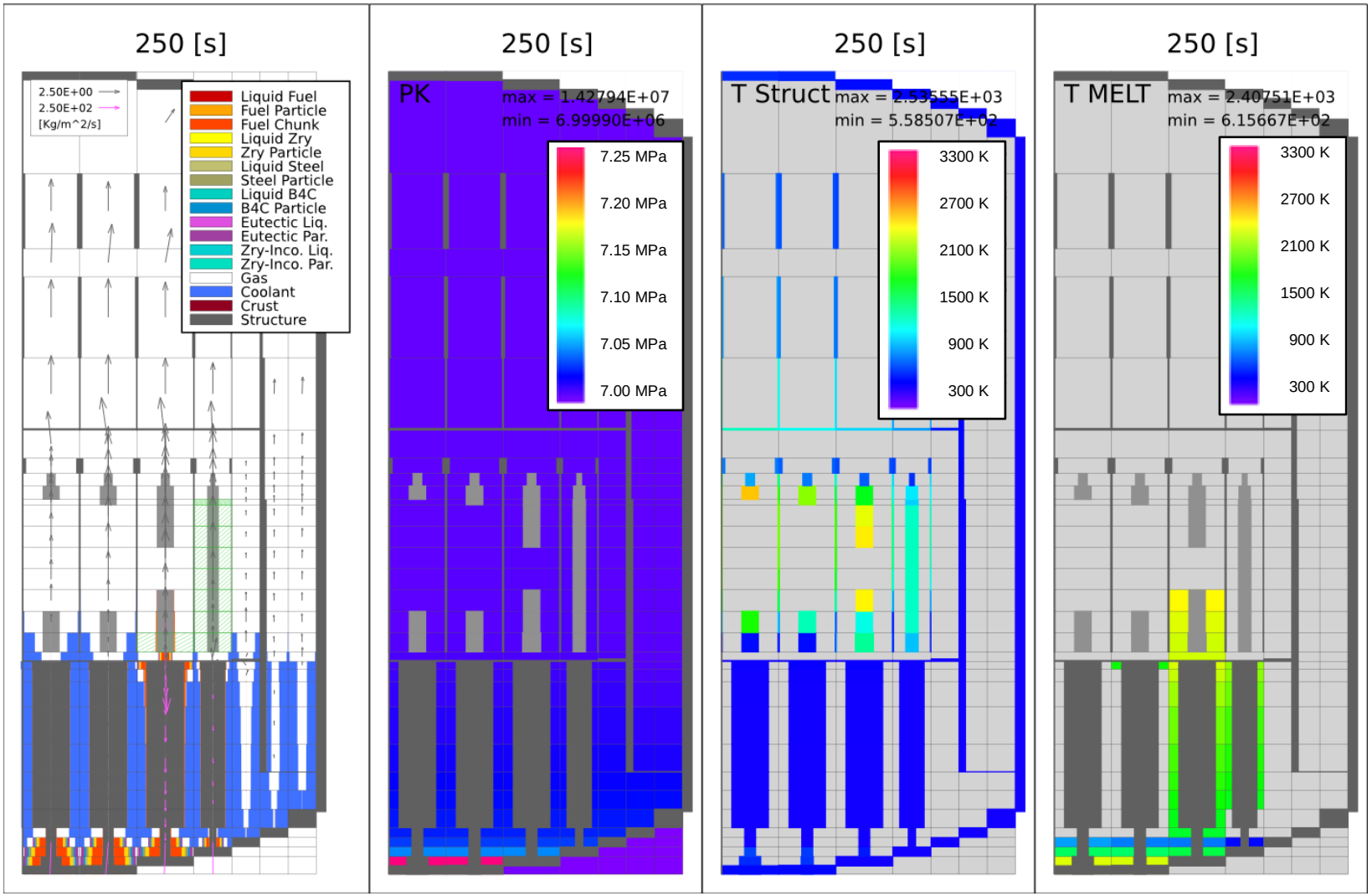
Exploratory Analyses: Simulation Model of ¼ Sector-Core Geometry





i. Water level: BAF
ii. Simulation time: 250s
iii. Decay heat level: 10%
iv. Pressure: 7 MPa

Exploratory Analyses: Results for Middle-Power-Level BWR In-Vessel Core Degradation



(a) Distribution of material volume fraction

(b) Pressure distribution

(c) Structural temperature distribution

(d) Molten corium temperature distribution

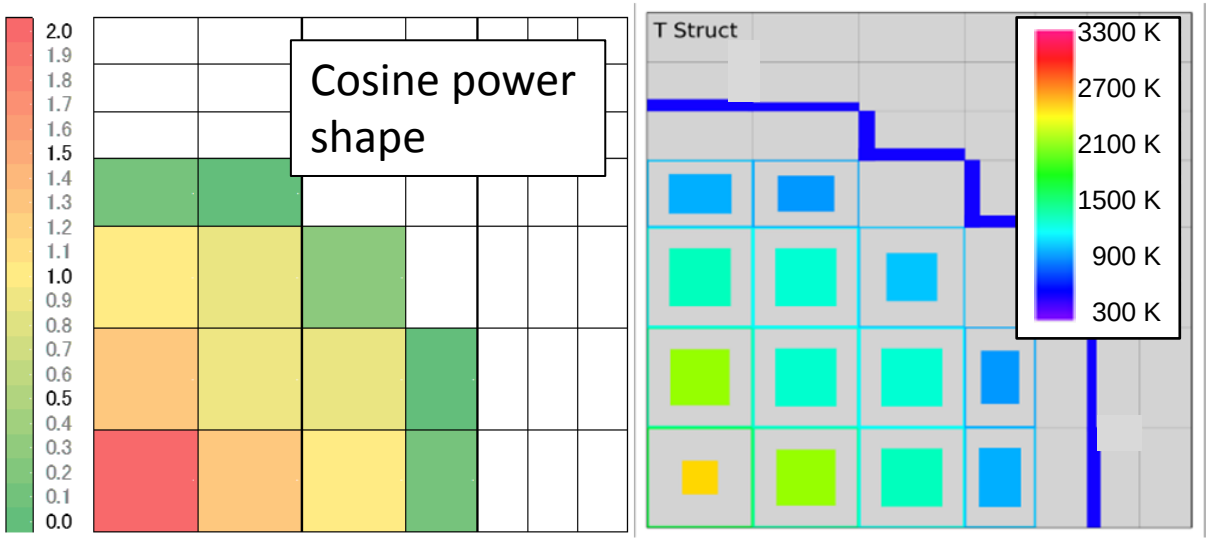
Exploratory Analyses: Calculation Results from Sensitivity Analyses

Variables	Reference case	Initial water level (TAF)	Decay Heat (Way-Wigner equation)	Radial Power shape (Flat)	Oxidation of fuel cladding (Fresh fuel loading)
Timing of fuel failure [s]	131	255*	586*	243	102
Fuel failure mode	Pellet melting	Pellet melting	Pellet melting	Pellet melting	Cladding melting
Hydrogen production [kg]	13.7	2.3	1.6	16.7	12.6
Weight of corium on the bottom of lower head [kg]	7,300	760	0	7,200	13,200

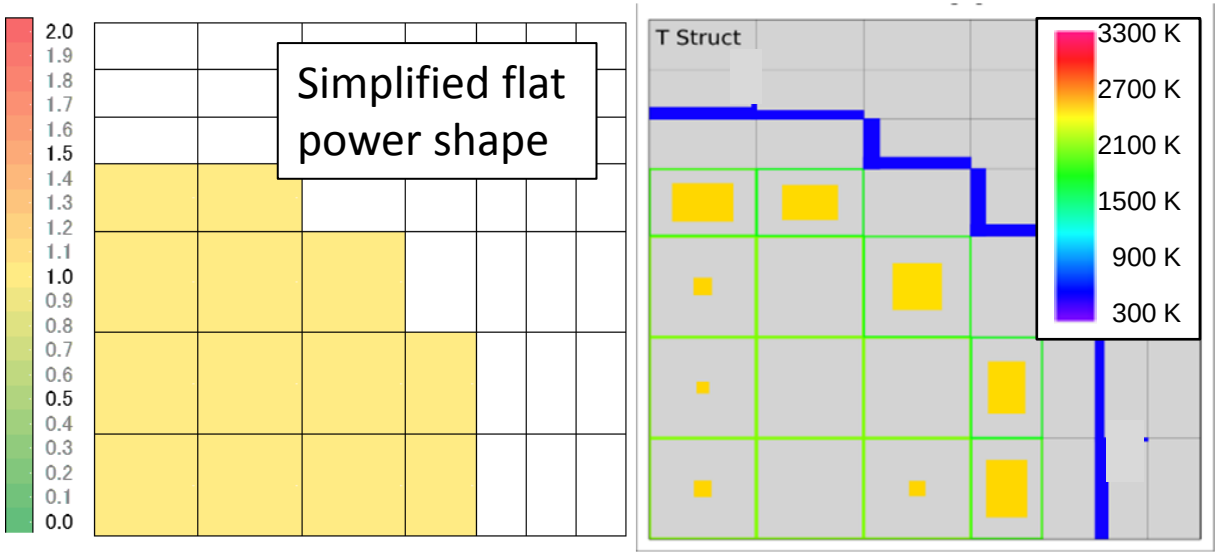
* The calculations were continued up to the fuel failures to evaluate the fuel failure mode.

Sensitivity Analyses of Radial Power Shape: Temperature Distributions & Core Degradation Appearances

- The reference (a) : radially cosine-power-shape
- The other case (b) : simplified flat radial power shape
- The time to fuel failure of the reference (a) was fast due to the higher radial peaking factor



(a) Radial cosine-power shape at 134s



(b) Simplified homogenous power shape at 245s

6. Summary

- Tight coupling calculation of the neutronic, thermal-hydraulic and fuel pin behavior modules was verified in the single fuel assembly geometry.
- Until now, validation has been conducted with major existing experiments: DF-4, QUENCH-06, CORA-16&18, XR-2, KROTOS-37 and FARO L-19.
- These validation results showed the capability of three-dimensional simulation code for calculation of BWR in-vessel core degradation.
- Exploratory analyses regarding initial conditions of BWR in-vessel core degradation were executed. It was found that the initial parameters such as water level, etc. were sensitive for BWR core degradation progression.

Implementation of QUENCH-10 experiment modelling experience for the modelling of severe accidents in spent fuel pools

Tadas Kaliatka, Lithuanian Energy Institute, Breslaujos str. 3, LT-44403 Kaunas, Lithuania

ABSTRACT

QUENCH – 10 experiment performed at the Karlsruhe Research Center consist of three stages: (1) pre-oxidation in steam, (2) period of air ingress, and (3) reflood. Because of the long period of peroxidation in steam and air atmosphere, this experiment support understanding of the consequences of a possible failure of heat removal in a spent fuel pool. Evaporation of the pool water may lead to fuel element degradation and consequently fission release from the damaged fuel elements.

For the analysis of QUENCH-10 experiment the latest version of RELAP/SCDAPSIM mod 3.5 was used. This code is the semi-mechanistic lumped parameter code for transient analysis of a reactor core and coolant system. In the last version of the code several new zirconium oxidation correlations in steam and air ambient were recently implemented by Innovative Systems Software (code maintenance group) and Paul Scherrer Institute. QUENCH-10 experiment modelling results received using new zirconium oxidation models and standard parabolic correlation were compared with experimental data. Based to the comparison of total hydrogen generation rates and formatted oxide thickness it was made conclusion, what the new oxidation correlations more precise reflects the experimental data and sufficiently replicates measured data of QUENCH-10 experiment.

Using gained experience from the modelling of the QUENCH-10 experiment, the model of spent fuel pool (BWR-4 type reactor and Mark-I type containment) was developed. Analysis of loss of coolant accident without water make-up was performed using the new zirconium oxidation models (the same as for QUENCH-10 experiment). The results of calculation shows that the amount of generated hydrogen using new zirconium oxidation models is 30% lower than using old zirconium oxidation model.



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Implementation of QUENCH-10 experiment modelling experience for the modelling of severe accidents in spent fuel pools

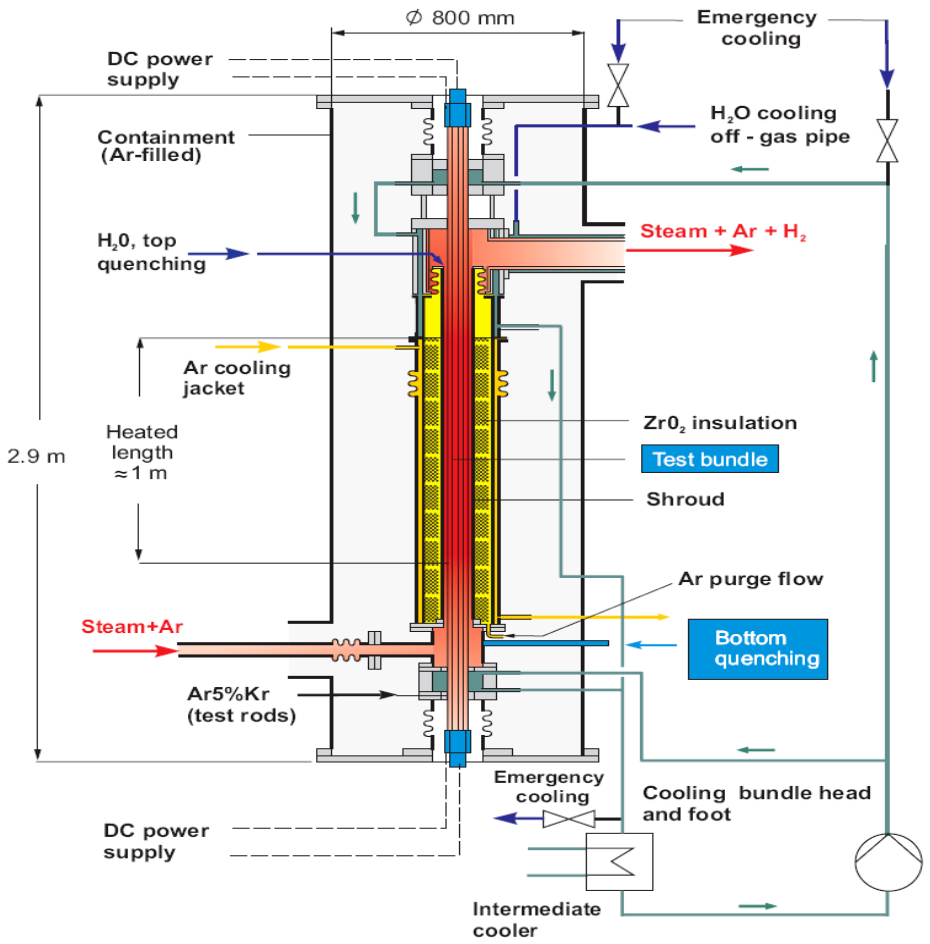
Tadas Kaliatka

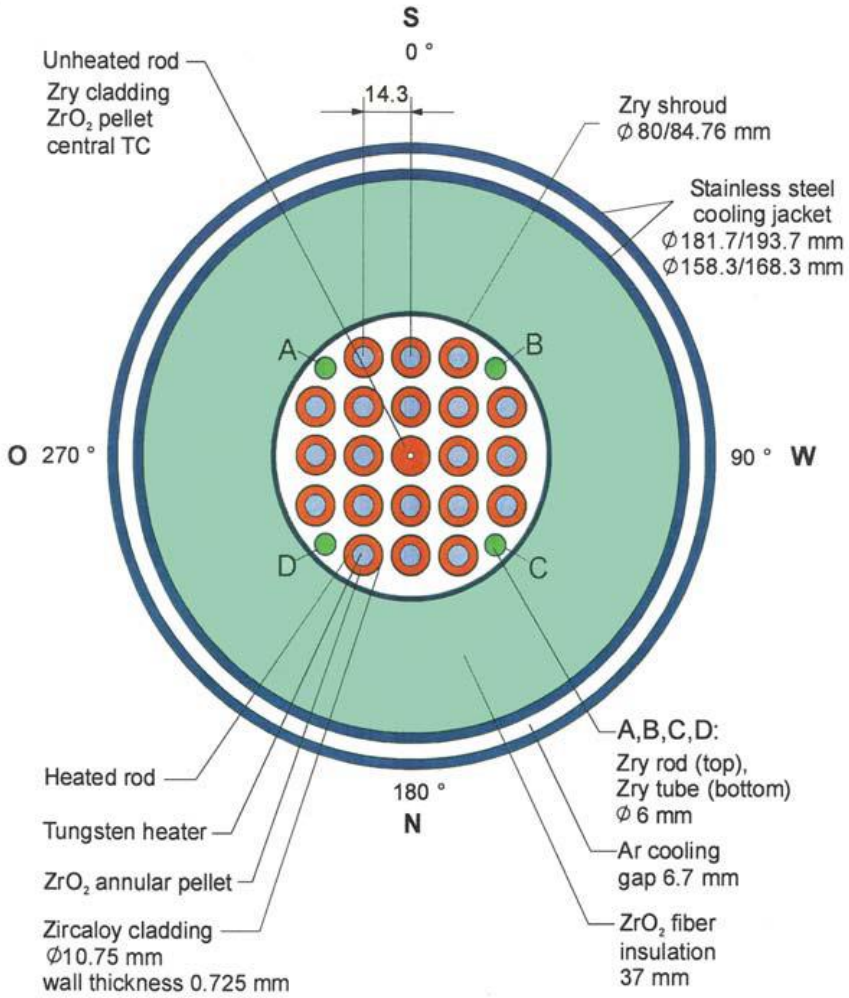
Outline

- Modelling and results of QUENCH-10
- Modelling of LOCA accident in the BWR spent fuel pool
  - RELAP5 and RELAP/SCDAPSIM model of BWR SFP
  - RELAP5 calculation results
  - RELAP/SCDAPSIM calculation results
  - Comparison with the older version of RELAP/SCDAPSIM which is using standard parabolic correlation

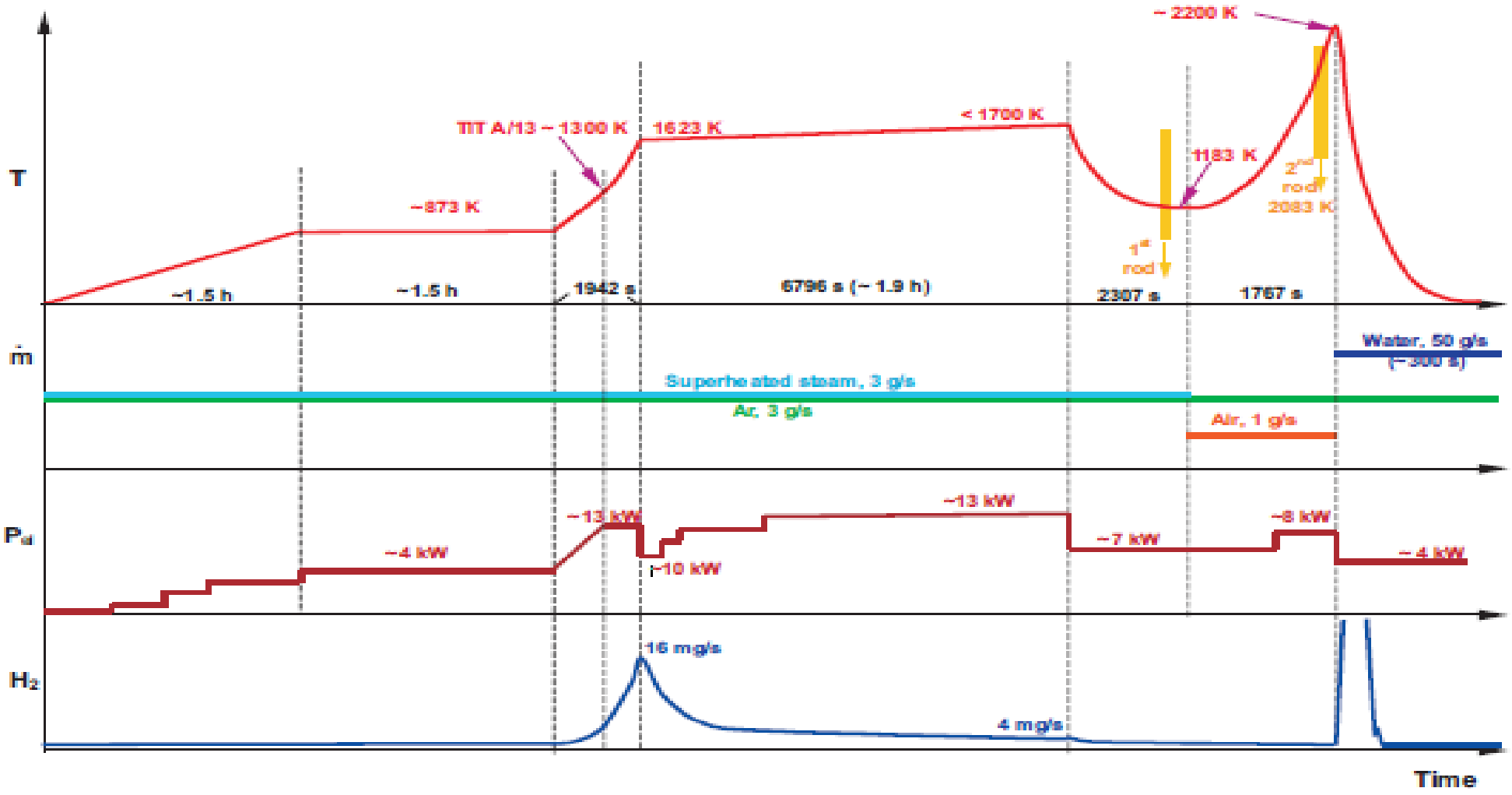
QUENCH test

- The main focus of the QUENCH tests lies on the analyses of the hydrogen generation during the reflood.



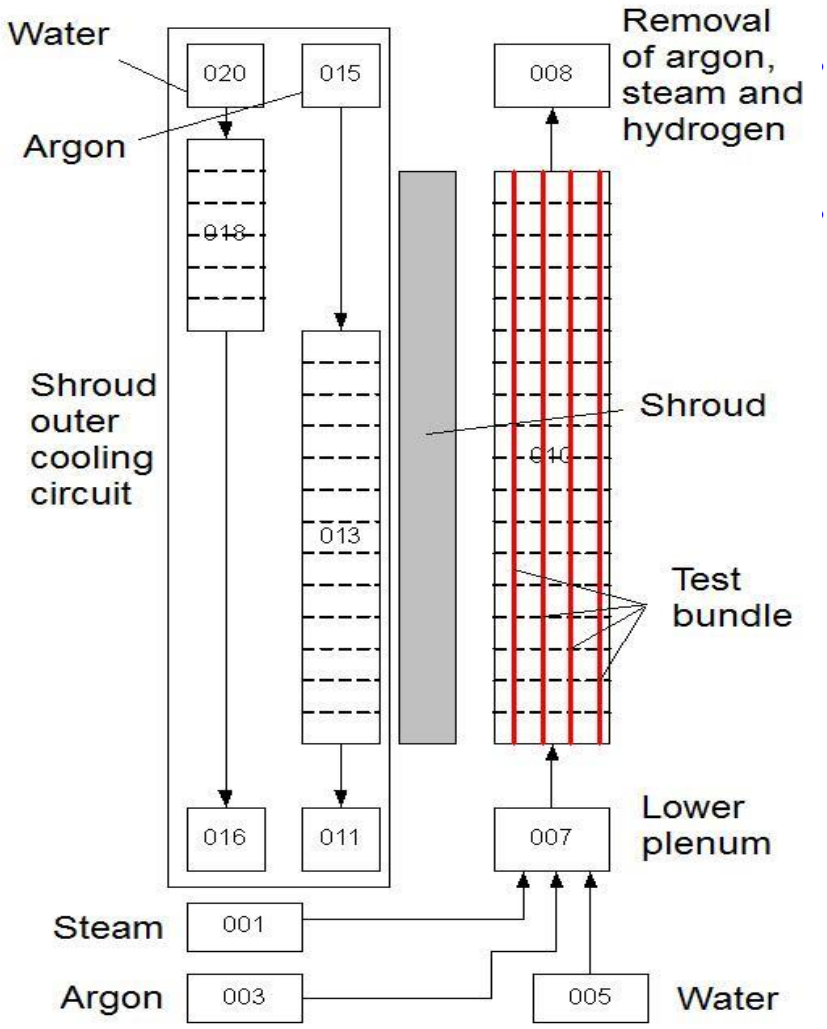


QUENCH-10 tests



RELAP/SCDAPSIM model of the QUENCH test

RELAP/SCDAPSIM is designed to describe the overall reactor coolant system thermal hydraulic response and core behaviour under normal operating conditions or under design basis or severe accident conditions.



RELAP module (thermal hydraulics):

- All Elements

SCDAP module (behaviour of the core and vessel structures):

- component 1 - one central rod (not heated), modelled as a "fuel" element is composed of ZrO_2 pellets in the centre, a gas-filled gap and cladding of Zircaloy;
- component 2 - 8 heated rods (around the central rod) which simulates heated fuel rods, modelled using "Cora" component which is composed of tungsten heating elements in the centre, ZrO_2 pellets, gas-filled gap and cladding of Zircaloy;
- component 3 - 12 heated rods, modelled using "Cora" component;
- component 4 - the four rods in the corners, modelled as "fuel" component;
- component 5 – shroud of the bundle, modelled as "shroud" component, which consists of the inner Zircaloy layer, an insulating layer of ZrO_2 and Inconel layer (using properties of stainless steel).

RELAP/SCDAPSIM models for the steam and air oxidation

RELAP/SCDAPSIM MOD3.5 (dp)

Correlations could be used for oxidation in the presence of **steam**:

- Cathcart-Pawel/Urbanic-Heidrick,
- Leistikow-Schanz/Prater-Courtwright,
- Sokolov,
- **Standard parabolic (used in the previous versions of RELAP/SCDAP),**
- **Shattering (correlation evaluating shattering effect of Zr tubes).**

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Analysis of QUENCH-10 and -16 air ingress experiments with SCDAPSim3.5

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ABSTRACT

A recently developed air oxidation model has been implemented in the code version SCDAPSim3.5 and used to analyse the air ingress experiments QUENCH-10 and QUENCH-16 as part of the model assessment. The experiments comprised pre-oxidation in steam, a period of air ingress, and reflood. The analysis focused on the thermal response and possible breakaway during the oxygen consumption. Of particular interest were the effect of the oxygen concentration on the kinetics and the effect of oxygen starvation on the oxidation excursion during the subsequent reflood. The large excursion observed in QUENCH-16 during reflood was not reproduced by the model; the possible causes were explored and indicated the need of further improvement in the model.

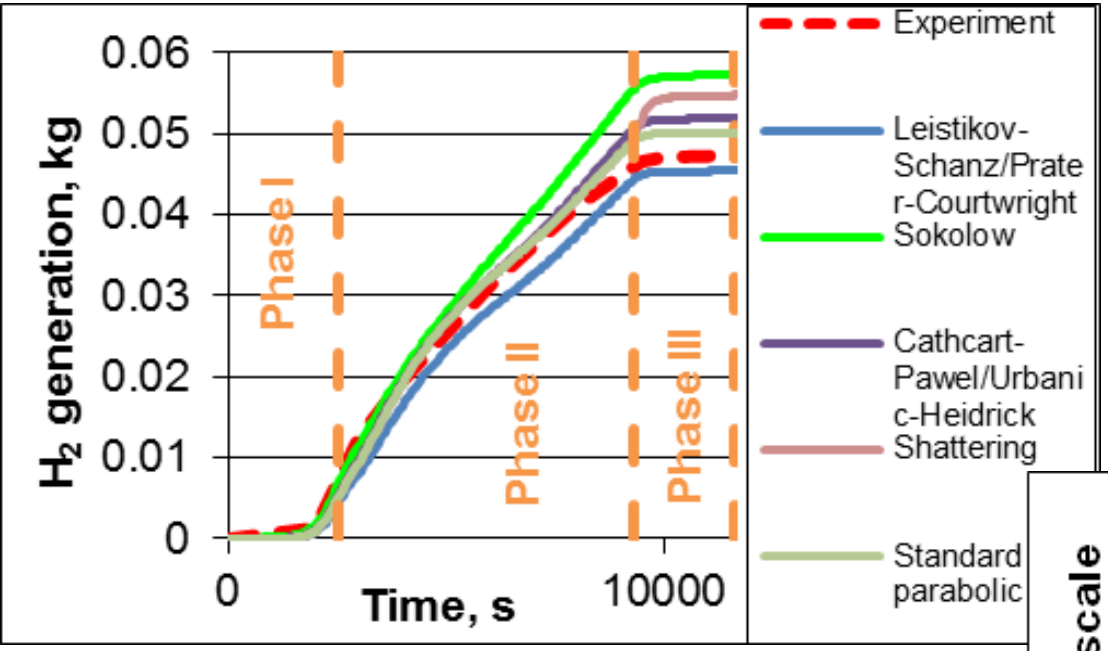
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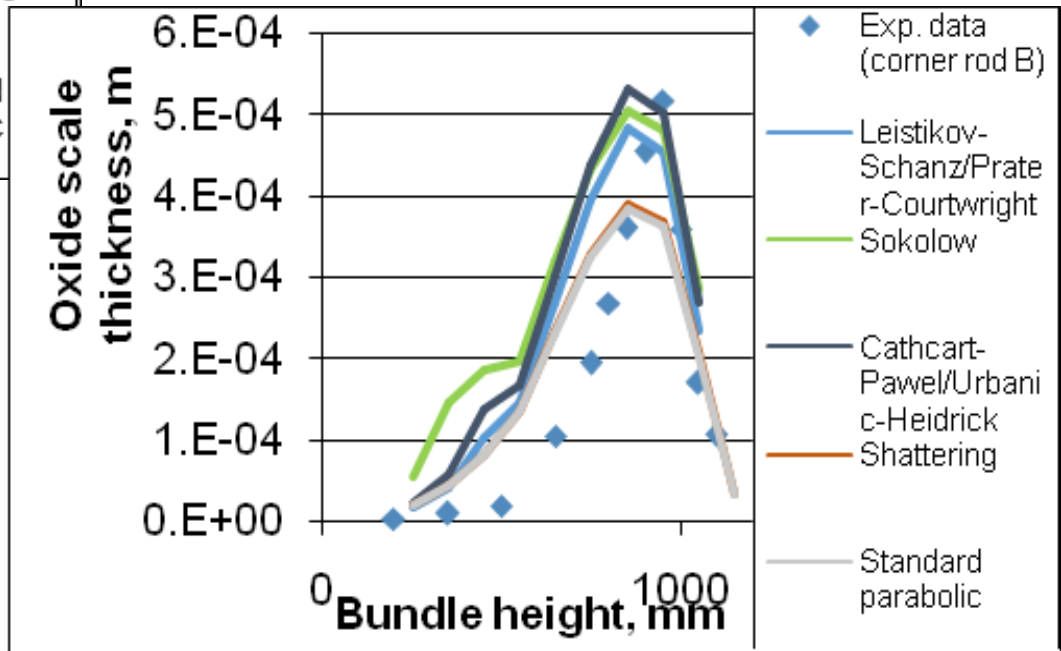
Correlations could be used for oxidation in the presence of **air**:

- Hofmann-Birchley,
- Hayes-Roberson/Leistikov-Berg (NUREG1),
- Powers (NUREG2),
- MELCOR,
- MOZART.

QUENCH-10 test

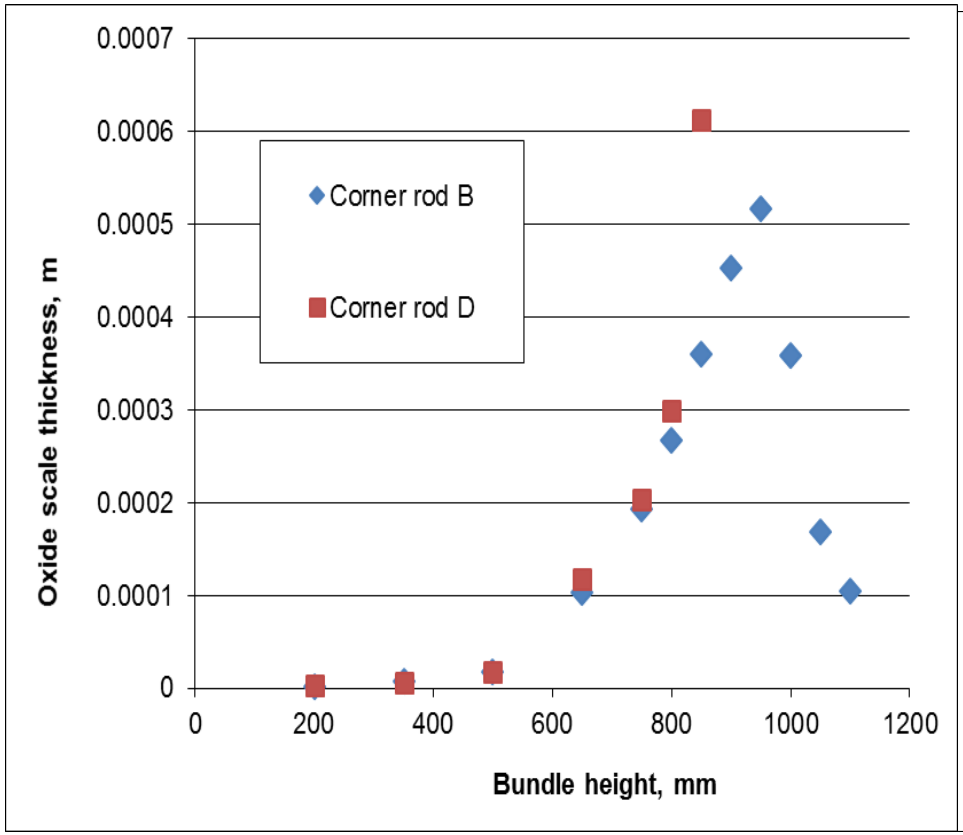
Influence of **steam oxidation** model to calculation results

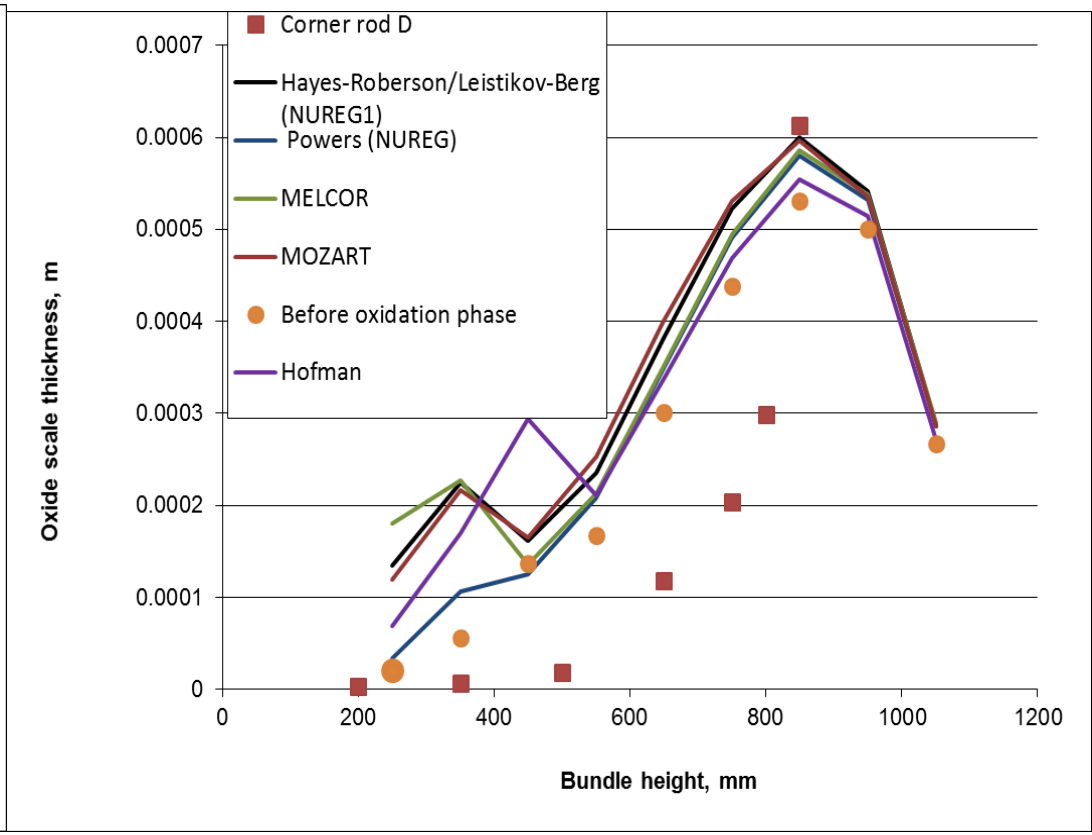




QUENCH-10 test

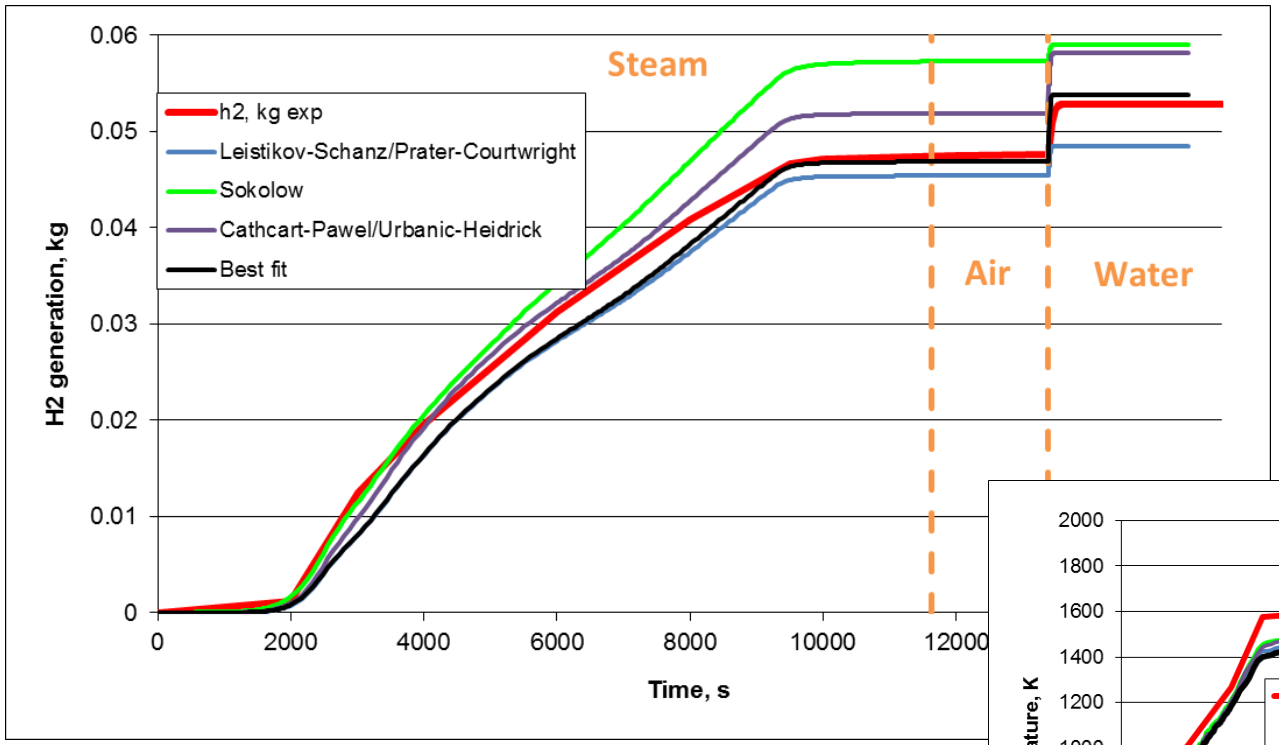
Influence of **air oxidation** model to calculation results



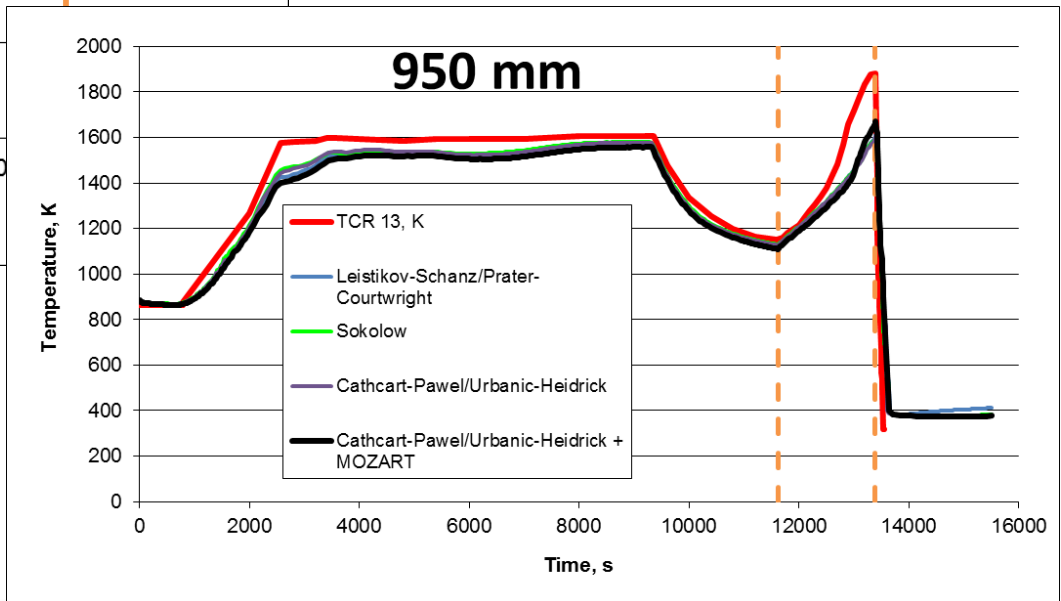


QUENCH-10 test

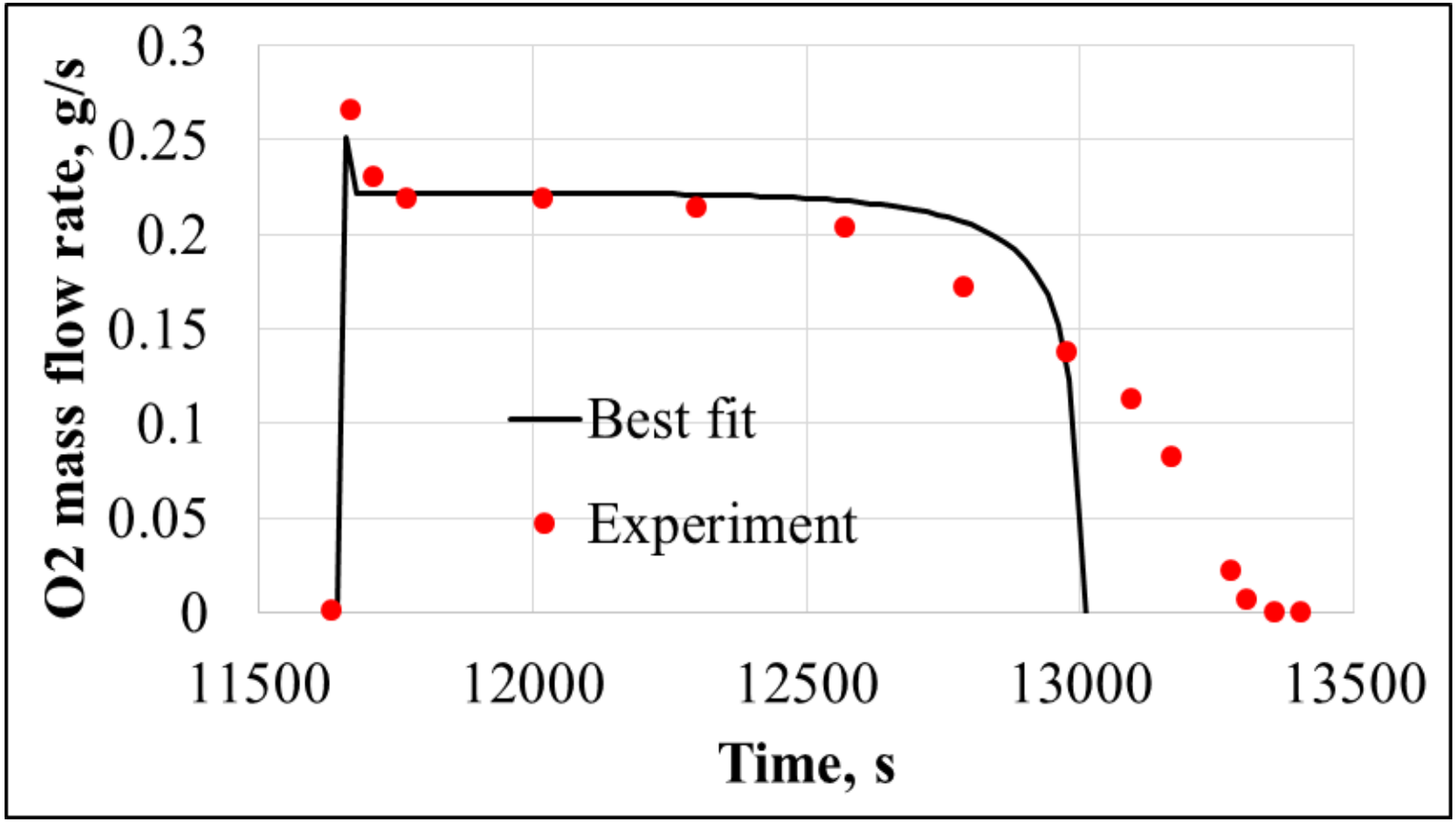
Best fit calculation



Calculation results using Cathcart-Pawel/Urbanic-Heidrick and MOZART oxidation models and **3.7 mOhm** resistance of sliding contacts are presented



QUENCH-10 test



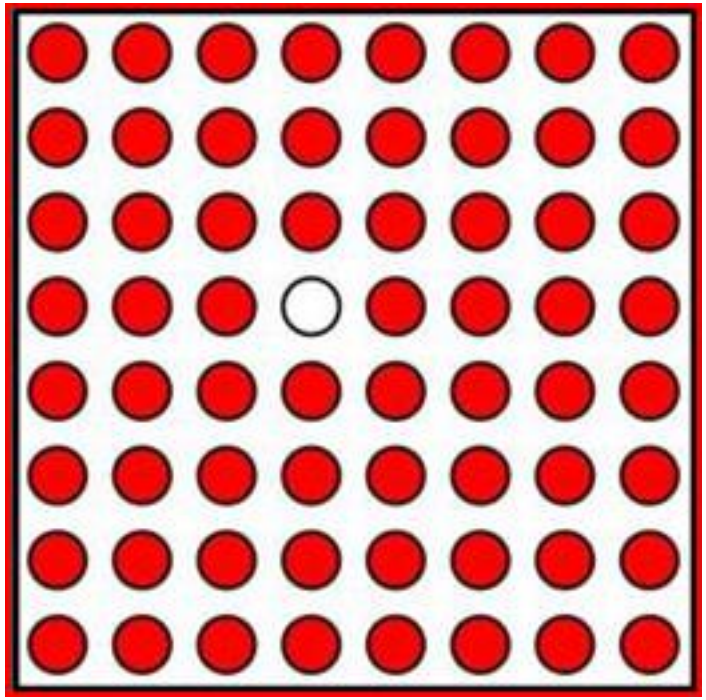
Comparison of the O_2 mass flow rate measured during experiment and calculation results show good agreement in the first part of the air ingress phase. However, calculated O_2 mass flow rate start to decrease latter and decreases sharper than in the experiment (global oxygen starvation in the calculation achieved ~400 s earlier). In general it could be concluded that oxygen starvation process in the QUENCH-10 experiment is predicted reasonably well.

Modelling of LOCA accident in the BWR spent fuel pool

- NPP with the BWR-4 type reactor and Mark-I type containment is capable to store 3055 spent fuel assemblies in the SFP. The air temperature in the pool must be up to 303 K and water temperature up to 313 K.

Parameter	
Pool length, m	12.2
Pool width, m	9.9
Water height, m	11.5
Pool height, m	11.8
Steel liner thickness, m	0.0635
Concrete wall thickness, m	2.0
Pool bottom thickness, m	2.0

Modelling of LOCA accident in the BWR spent fuel pool

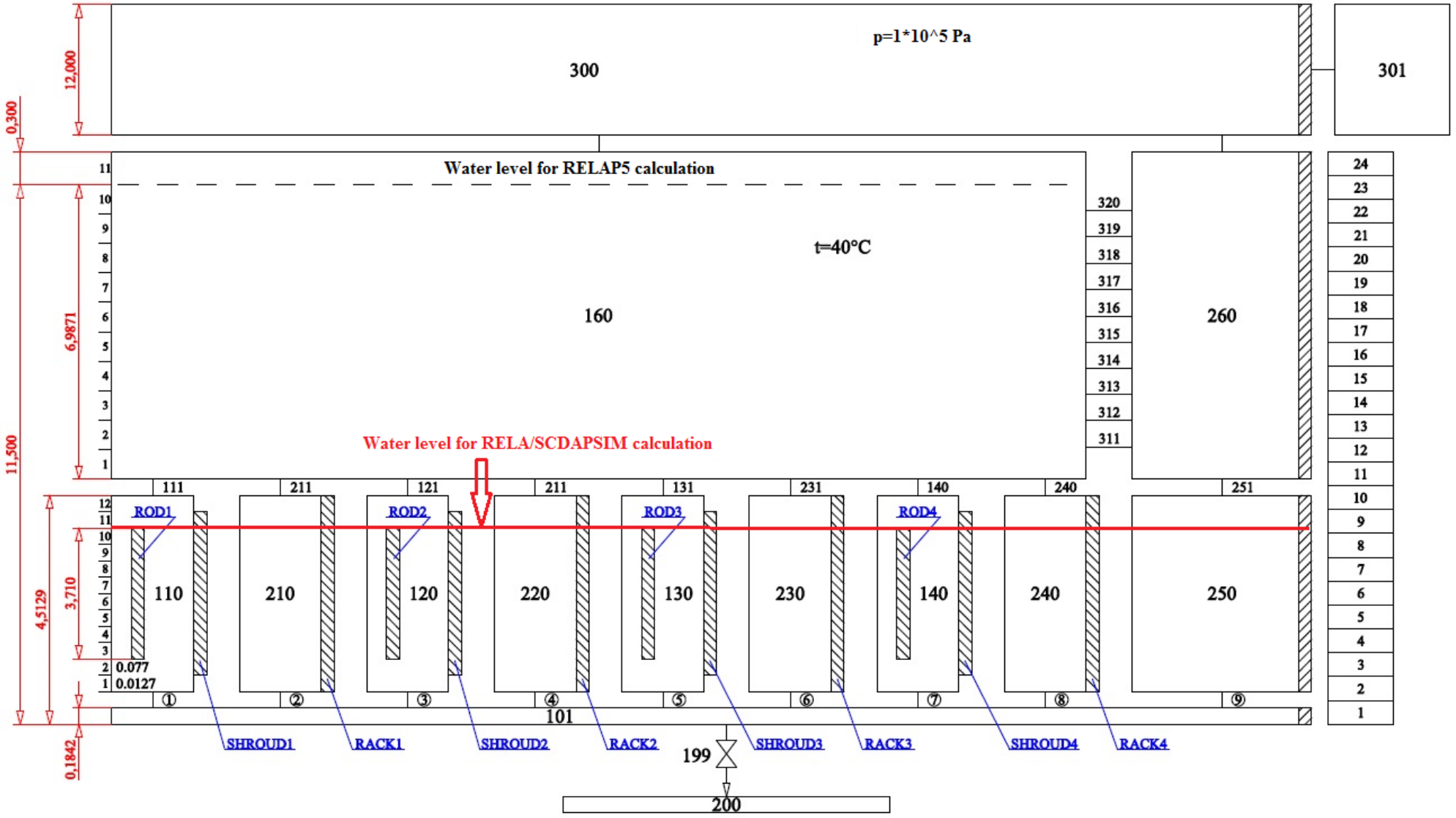


Fuel radius, mm	4.80
Gap, mm	0.1
Cladding radius (inside), mm	4.90
Cladding radius (outside) , mm	5.60
Cladding thickness, mm	0.70
Pitch, mm	14.5
Active fuel height, mm	3710
Fuel assembly height, mm	4276
Number of rods per FA	63
Internal pressure of rods, Pa	$3.0*10^6$
Width of canister (inside), mm	134.0
Width of canister (outside), mm	138.0
Canister thickness, mm	2.0
Grids elevations, m	0.5; 1.11; 1.72; 2.33; 2.94; 3.55
Grid height, mm	50
Grid thickness, mm	1.0

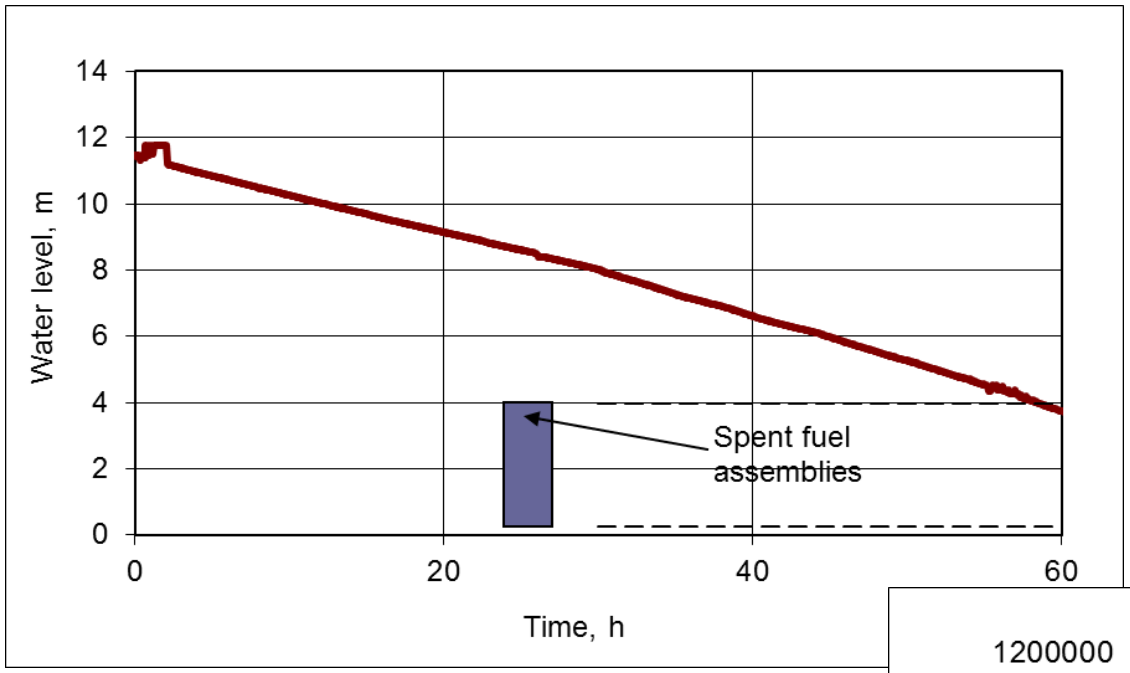
RELAP5 and RELAP/SCDAPSIM model of BWR SFP

Data			RELAP5 part			RELAP/SCDAPSIM		
Group	**Number of FA**	**Power of group, MW**	**Fuel rods**	**Fuel box**	**Racks**	**Fuel rods**	**Fuel box**	**Racks**
1	88	1.144	HS 11100	HS 12111	HS 12151	Fuel1	Shroud1	HS 12151
2	196	1.533	HS 11200	HS 12211	HS 12251	Fuel2	Shroud2	HS 12251
3	315	0.33	HS 11300	HS 12311	HS 12351	Fuel3	Shroud3	HS 12351
4	2456	0.558	HS 11400	HS 12411	HS 12451	Fuel4	Shroud4	HS 12451

RELAP5 and RELAP/SCDAPSIM model of BWR SFP

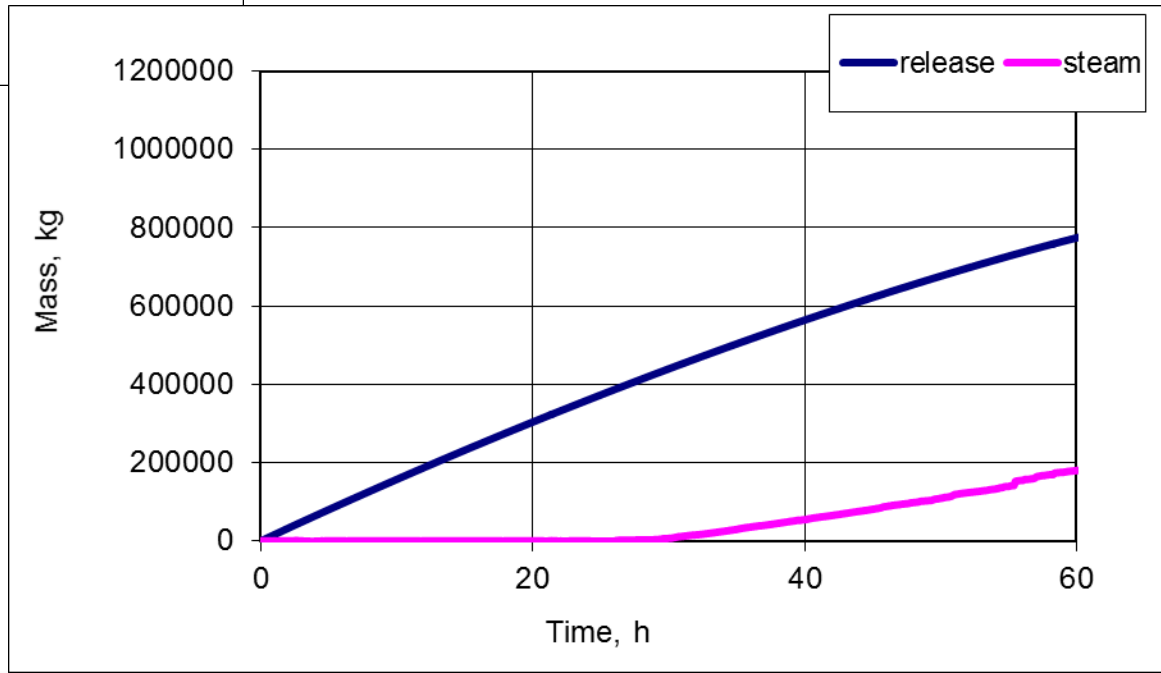


RELAP5 calculation results

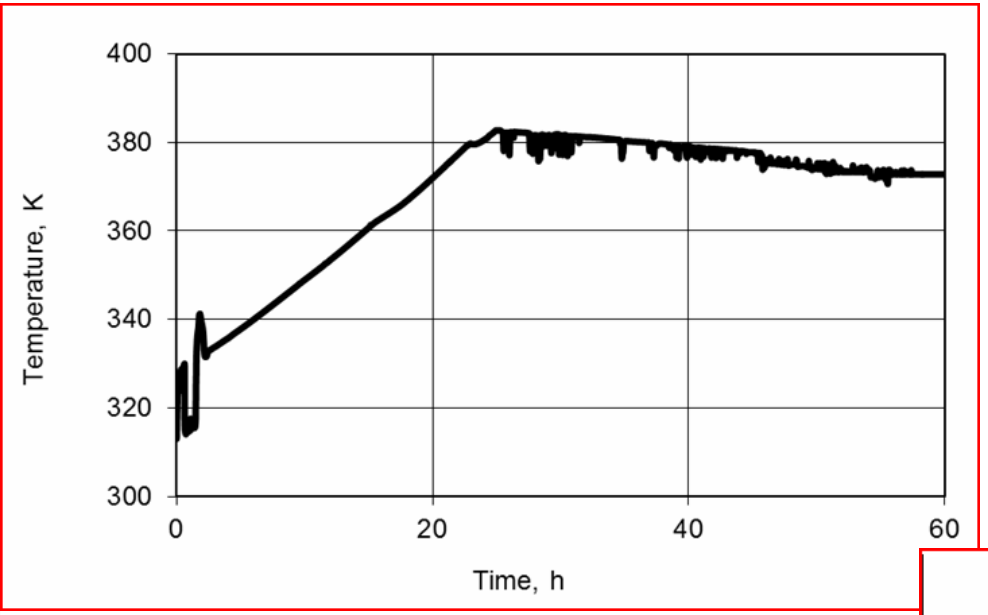


Behavior of pool water collapsed level

Comparison of integral water release and integral steam loss

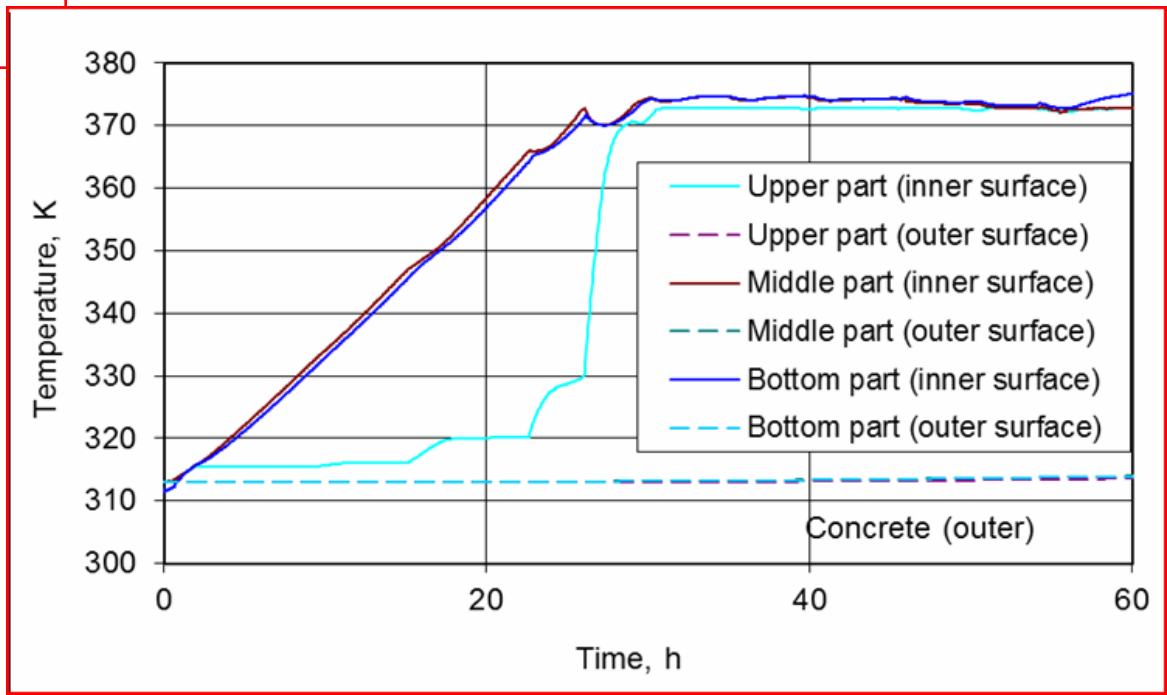


RELAP5 calculation results

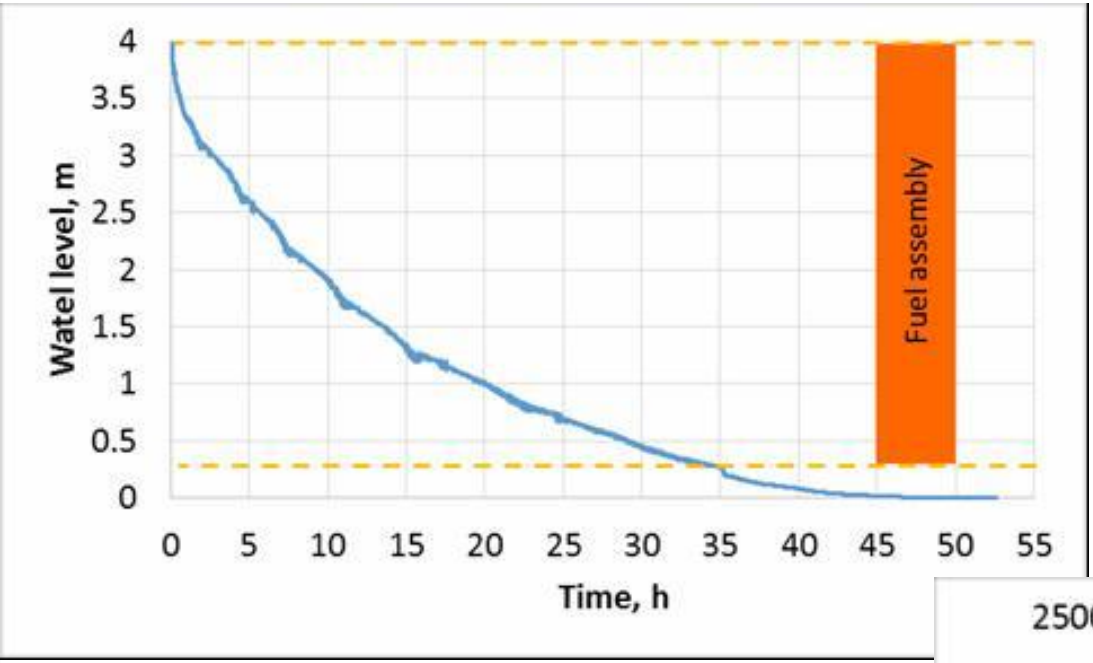


Behavior of pool coolant temperature (top of active fuel)

Temperature of SFP wall at bottom, middle and upper part of the pool

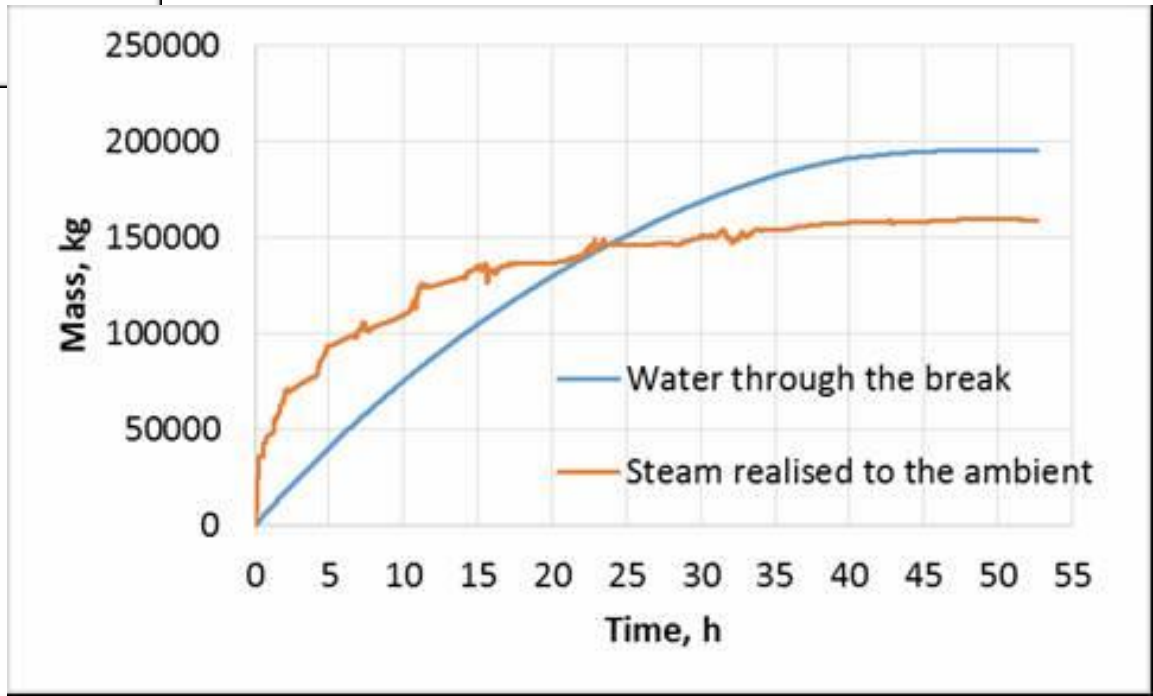


RELAP/SCDAPSIM calculation results

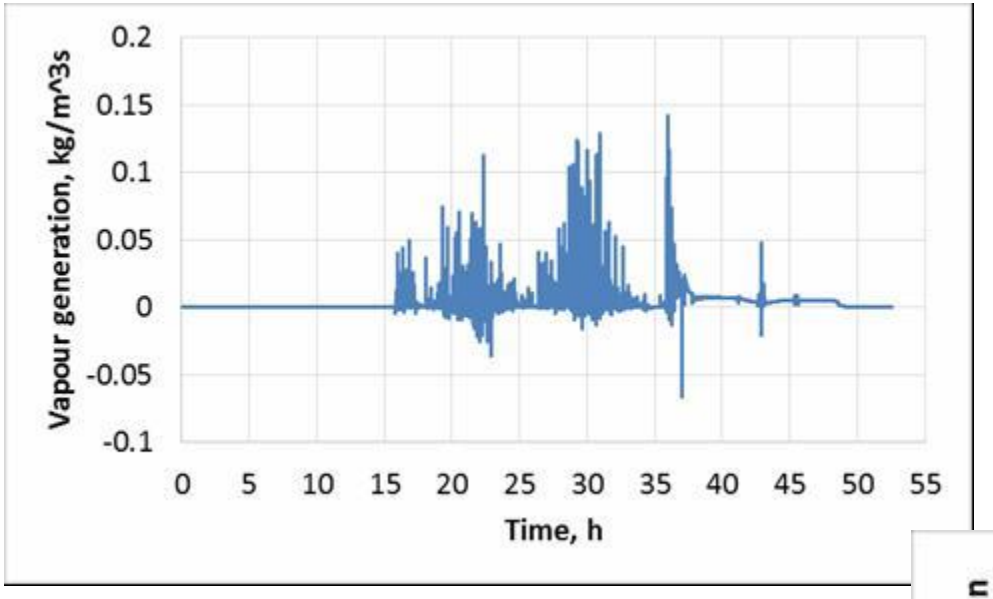


Water level in the pool

Water and steam released from the pool

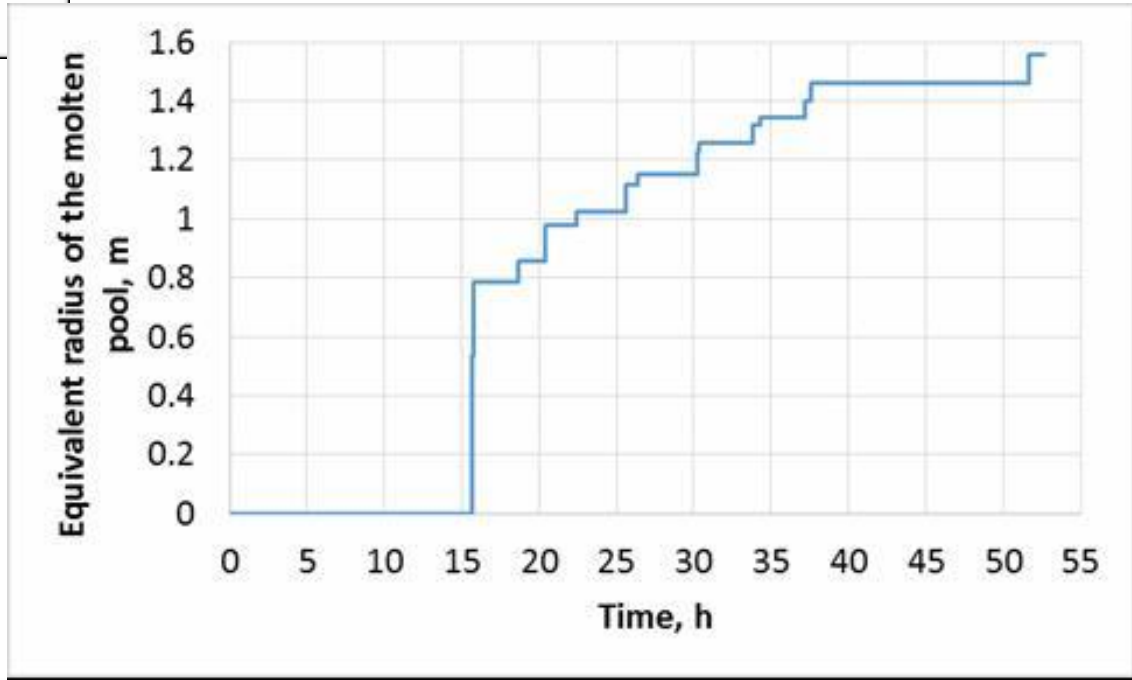


RELAP/SCDAPSIM calculation results



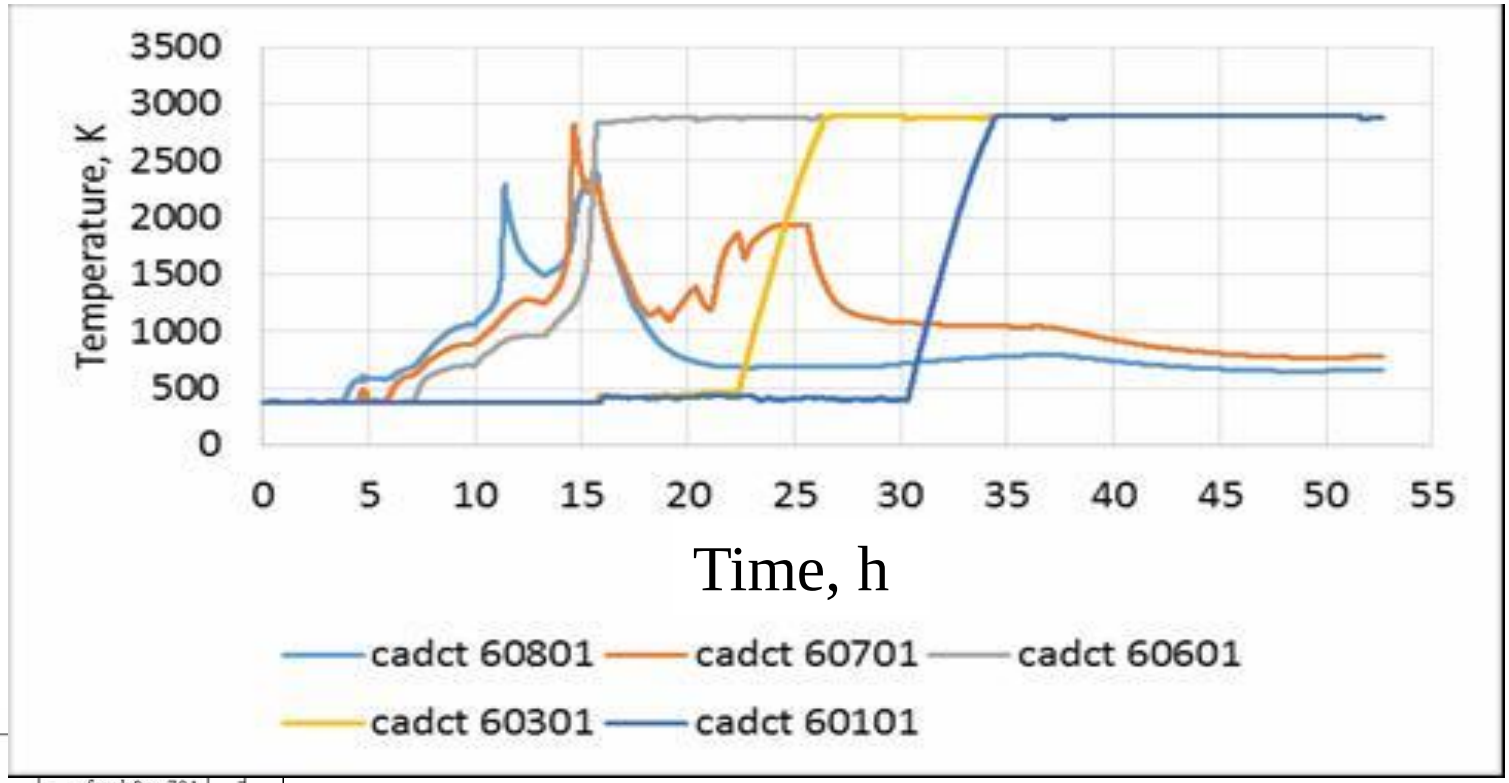
Vapour generation rate at the bottom part of the pool

Equivalent radius of the molten pool

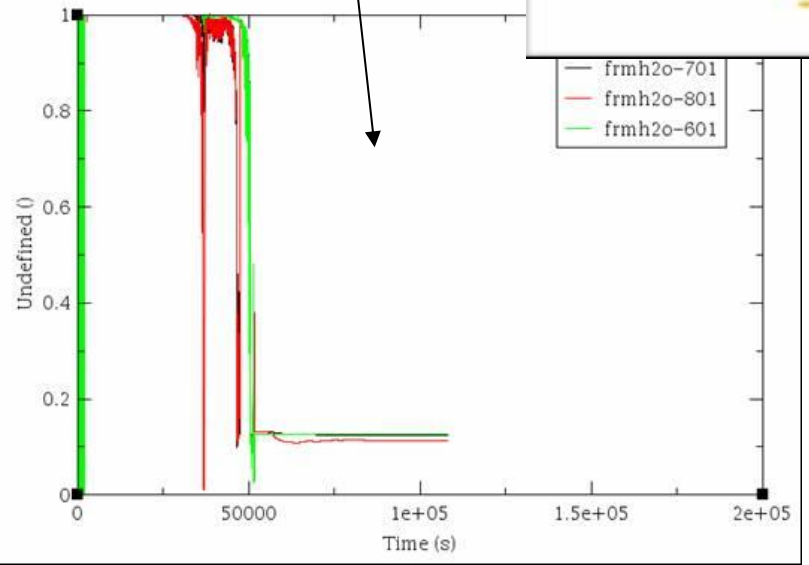


RELAP/SCDAPSIM calculation results

Cladding temperatures in the first group, nodes 8,7,6,3,1

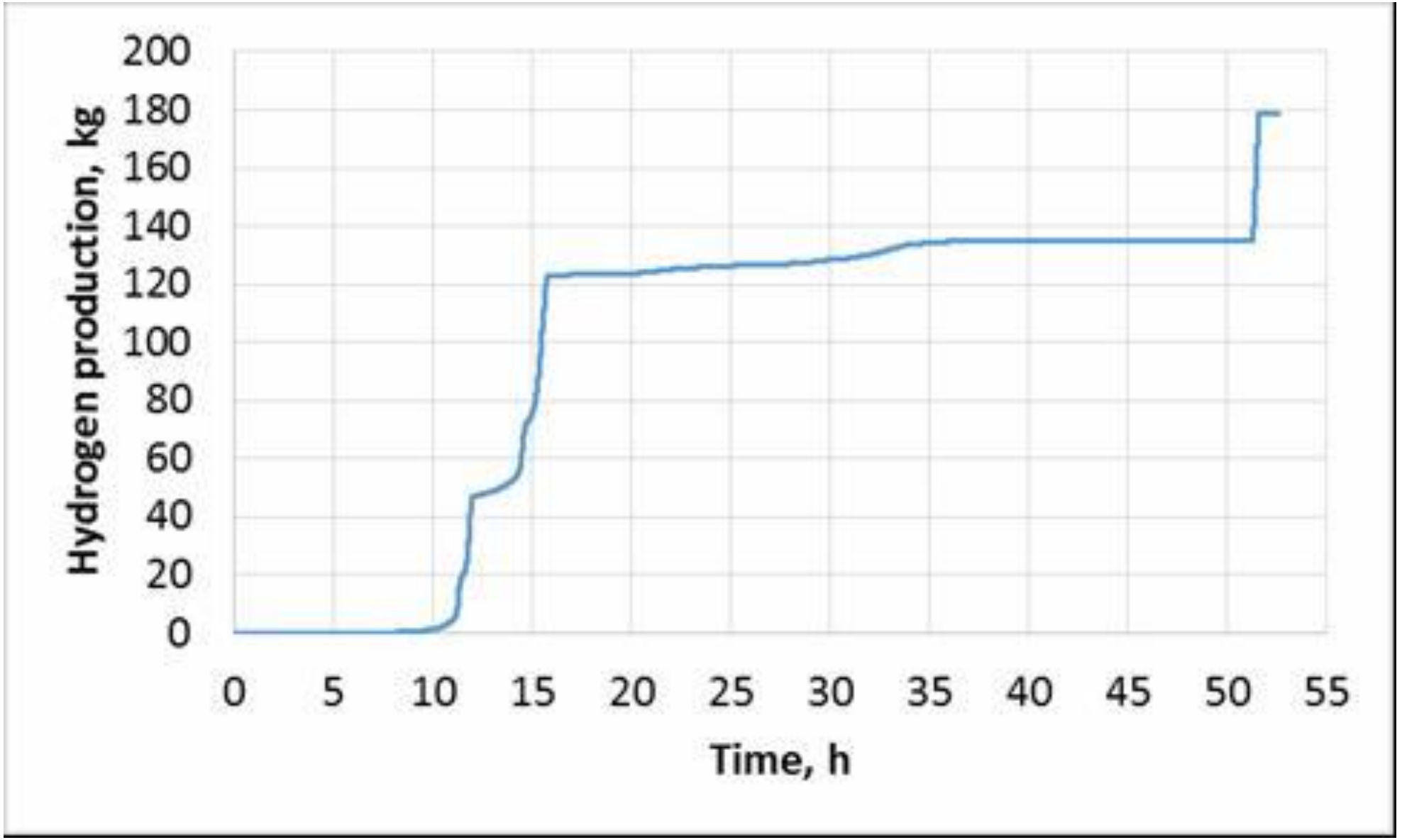


Steam mole fraction of the upper nodes of fuel rods



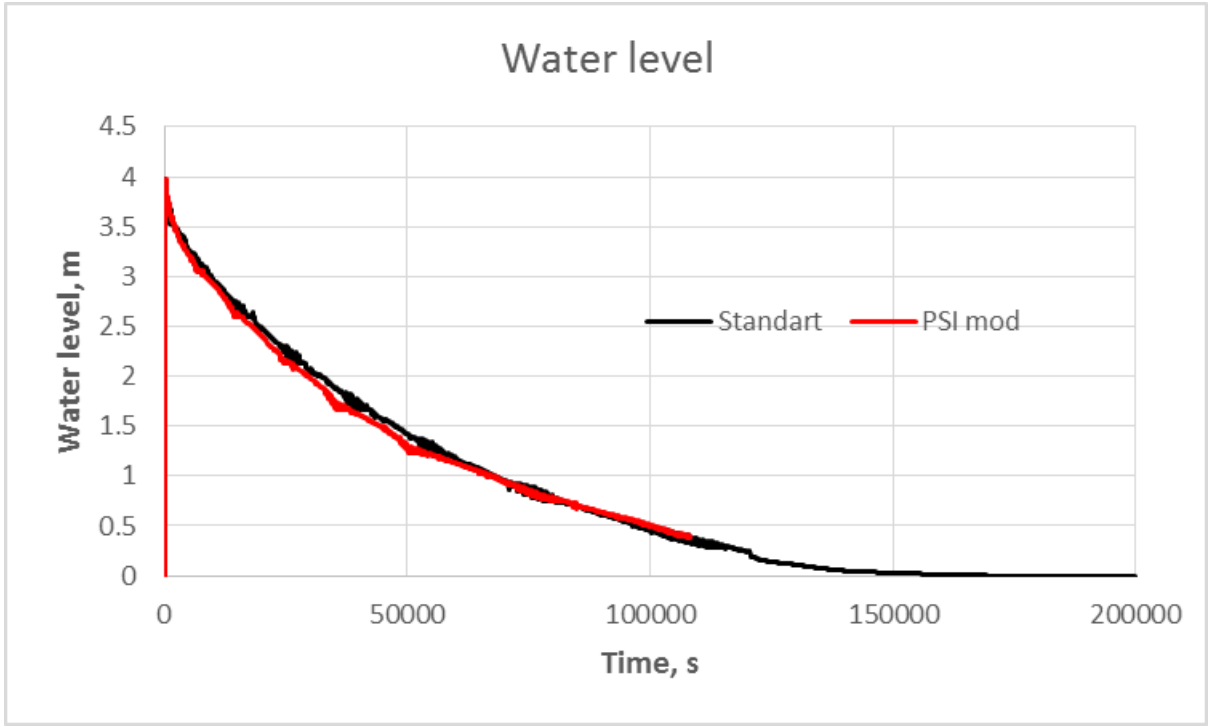
Temperatures at low elevations will start to increase sharply, while temperature decrease at the upper elevations. This is due to the fast consumption of oxygen at the upper elevation which will cause oxygen starvation. Once the upper elevations get oxygen starved, the lower elevations become more prompt to oxidize and they sharply increase their temperature.

RELAP/SCDAPSIM calculation results

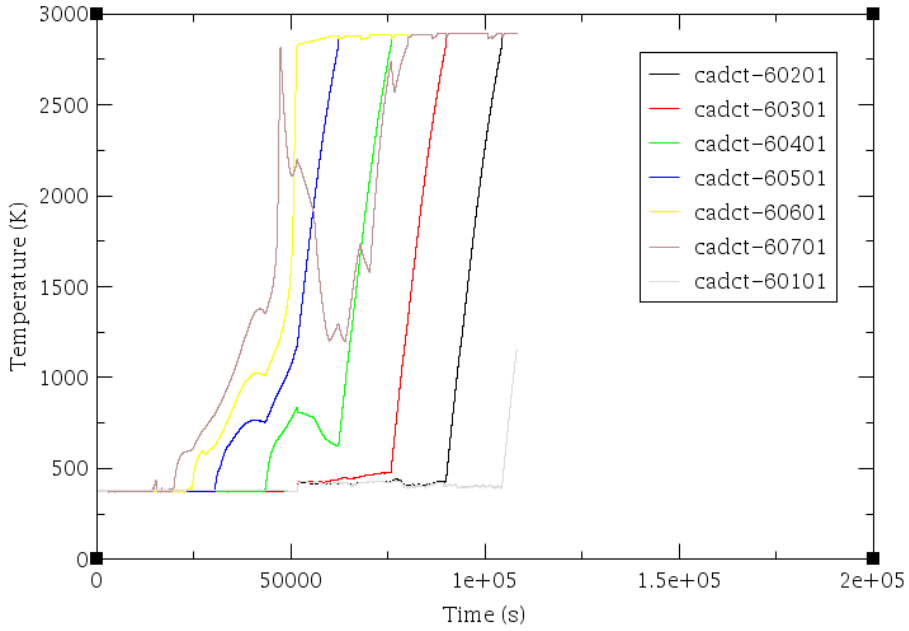


Comparison with the older version of RELAP/SCDAPSIM

- Cathcart-Pawel/Urbanic-Heidrick and MOZART oxidation models comparison with using standard parabolic correlation

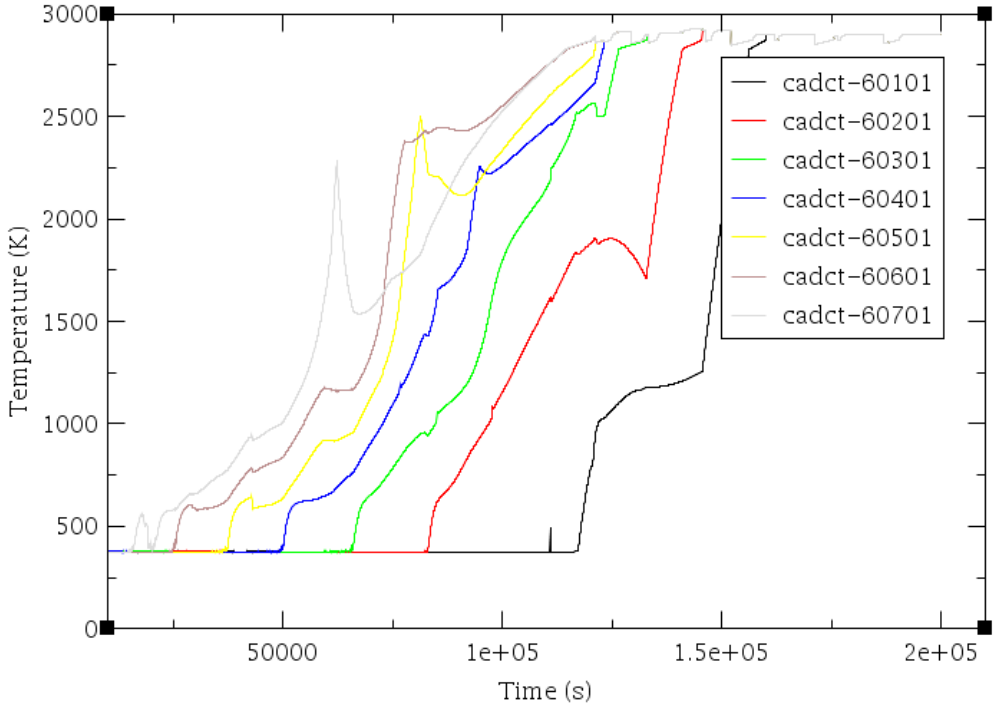


Comparison with the older version of RELAP/SCDAPSIM

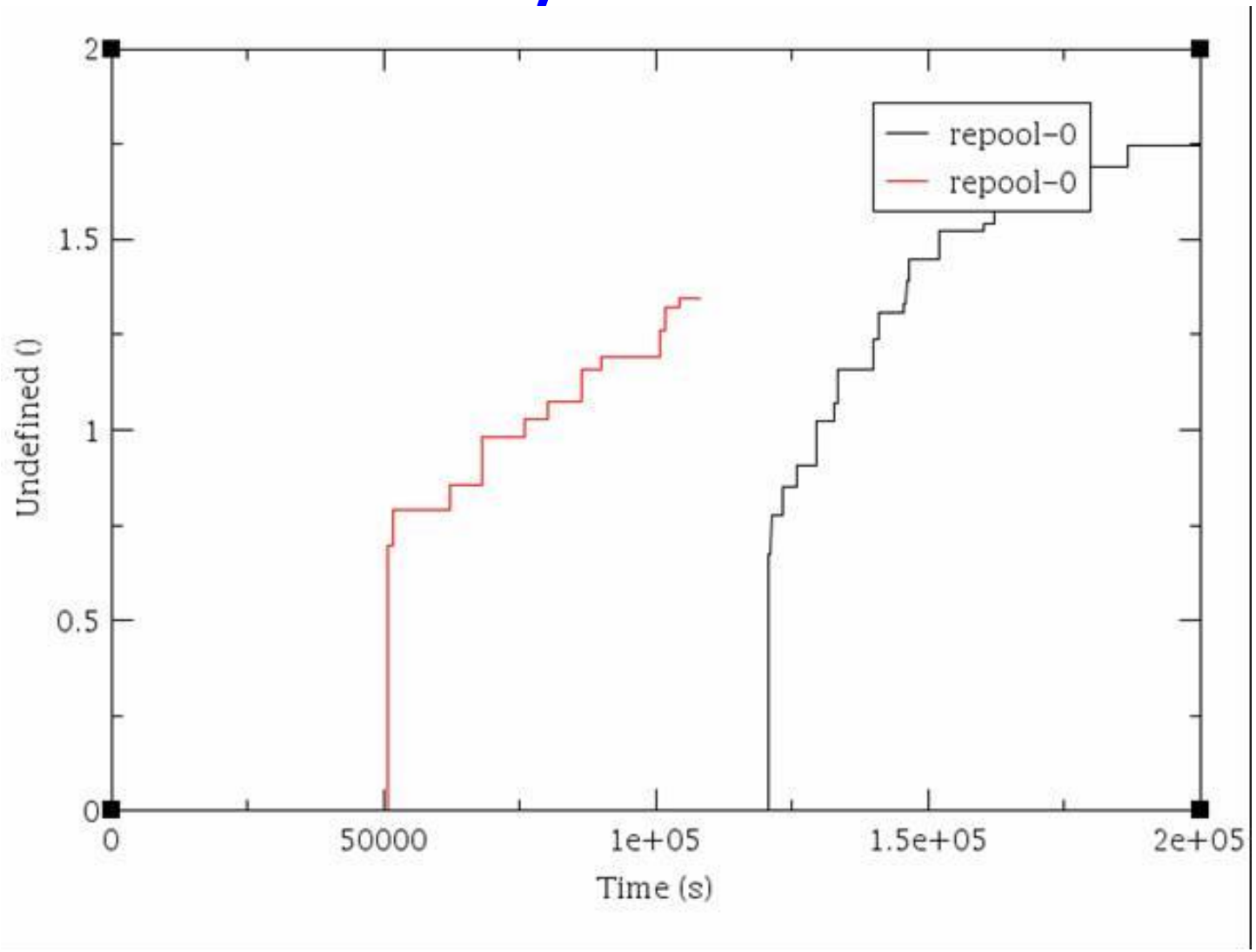


Temperature in the fuel rods. Calculation results using Cathcart-Pawel/Urbanic-Heidrick and MOZART oxidation models

Temperature in the fuel rods. Calculation results using standard model

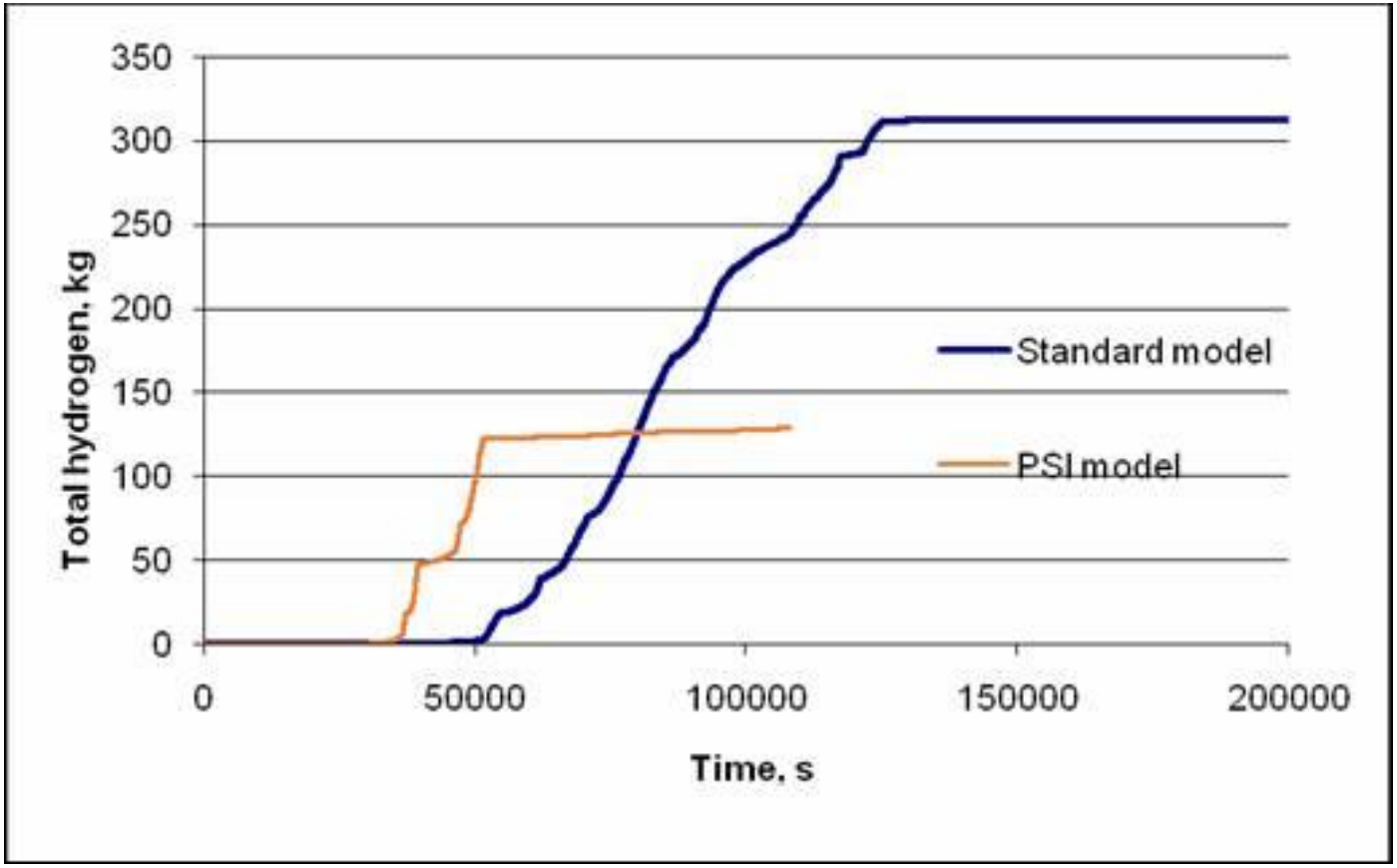


Comparison with the older version of RELAP/SCDAPSIM



Equivalent radius of the molten pool

Comparison with the older version of RELAP/SCDAPSIM



From the comparison it is clear what using Cathcart-Pawel/Urbanic-Heidrick and MOZART oxidation models the hydrogen generation start earlier, but after latter it stabilizes and increasing just a little bit. It is was not expected, because temperature still increasing very sharply, fuel rods starts to melt and the molten material is slumping to the bottom of the pool. So it's generates the steam and h_2 must be produced.

Conclusions

- Modelling of QUENCH-10 it was preparation for the modelling of BWR spent fuel pool. Modelling was done in order to understand the main phenomena which accruing during the experiment and to select the oxidation models.
- Modelling of LOCA accident in the BWR spent fuel pool have been provided using RELAP5 and RELAP/SCDAPSIM program codes.

Conclusions

- RELAP/SCDAPSIM calculations showed what up to 25 h after the calculation, water level in the pool decreases mainly due to the contact with fuel assemblies which have residual heat (mainly in the group 1 and 2). When the water level decreases to the bottom of the fuel assemblies steam generation decreases and the dominant water release from the pool is due to water discharge through the break. However, steam is also producing even then there is no water contact with the fuel assembly. This is due to molten fuel and metal slumping to the lower part of the SFP pool and contact with the hot surfaces of the walls.

Conclusions

- Calculation results using Cathcart-Pawel/Urbanic-Heidrick and MOZART oxidation models were compared with calculation results using standard parabolic correlation. Using Cathcart-Pawel/Urbanic-Heidrick and MOZART oxidation models the hydrogen generation start earlier, but after latter it stabilizes and observed only slow hydrogen increase. It was not expected because temperature still increasing, fuel rods melts and slumping to the bottom of the pool and steam is generated in the bottom part of the pool.

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Thank you for the attention

Tadas Kaliatka

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Novel percolation model in post-transition growth kinetics of heterogeneous zirconium oxide

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Material Science and Engineering, University of California, Los Angeles, CA 90095, USA

Abstract

We develop novel percolation framework for attaining the effective diffusivity within the given porous medium. Our model captures the shortest pathways as well as the effective transport volume (ETM). (Figure 1)We utilized this methodology in particular to predict the high-temperature corrosion kinetics of cracked zirconium oxide. Consequently, we generate oxide islands and their cracked boundaries stochastically. Using cellular automata, and starting from Dijkstr'as shortest path algorithm, we extract the effective tortuous percolation *rivers* and compute the homogenized diffusivity based on their geometry by means of site and bond percolation.[1, 2, 3, 4] The state-dependent model reproduces the jump in corrosion rate effectively, consistent with experimental observations.[5, 6]

Keywords

Percolation, corrosion cracking, zirconium oxide

References

[1] Jing-Chao Chen. Dijkstras shortest path algorithm. *Journal of Formalized Mathematics*, 15:144–157, 2003.

[2] Geoffrey Grimmett. What is percolation? In *Percolation*, pages 1–31. Springer, 1999.

[3] Dietrich Stauffer and Ammon Aharony. *Introduction to percolation theory*. CRC press, 1994.

[4] Ying Chen and Christopher A Schuh. Diffusion on grain boundary networks: Percolation theory and effective medium approximations. *Acta materialia*, 54(18):4709–4720, 2006.

[5] B Cox. The oxidation and corrosion of zirconium and its alloys v. mechanism of oxide film growth and breakdown on zirconium and zircaloy-2. *Journal of The Electrochemical Society*, 108(1):24–30, 1961.

*Corresponding author: aryanfar@caltech.edu

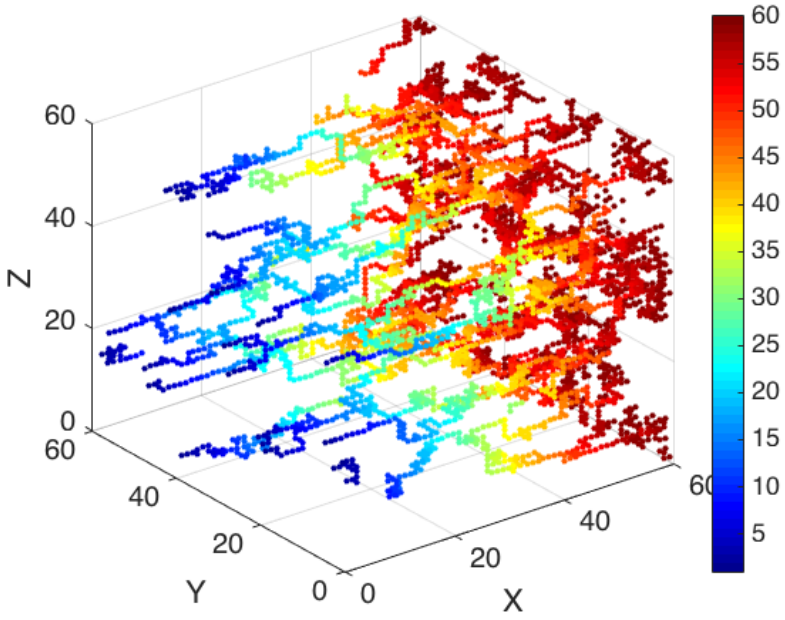


Figure 1: A sample of computed 3D effective transpot volume for a givem heterogenous medium.

[6] Bastien Chopard, Michael Droz, and Max Kolb. Cellular automata approach to non-equilibrium diffusion and gradient percolation. *Journal of Physics A: Mathematical and General*, 22(10):1609, 1989.



Post-transition corrosion kinetics of zirconium oxide

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University of California
Los Angeles, CA



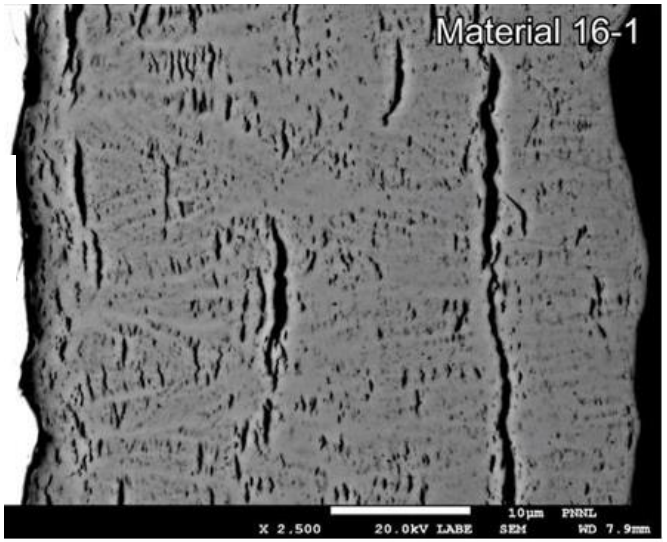
Quench Workshop
KIT , Karlsruhe, Germany
Oct 18, 2017

INTRODUCTION

Oxidation: PRE / POST transition

$$Zr(s) + xH_2O(g) \rightarrow ZrO_x(s) + xH_2(g)$$

Heating/Cooling ~ 600 K: CRACKS!

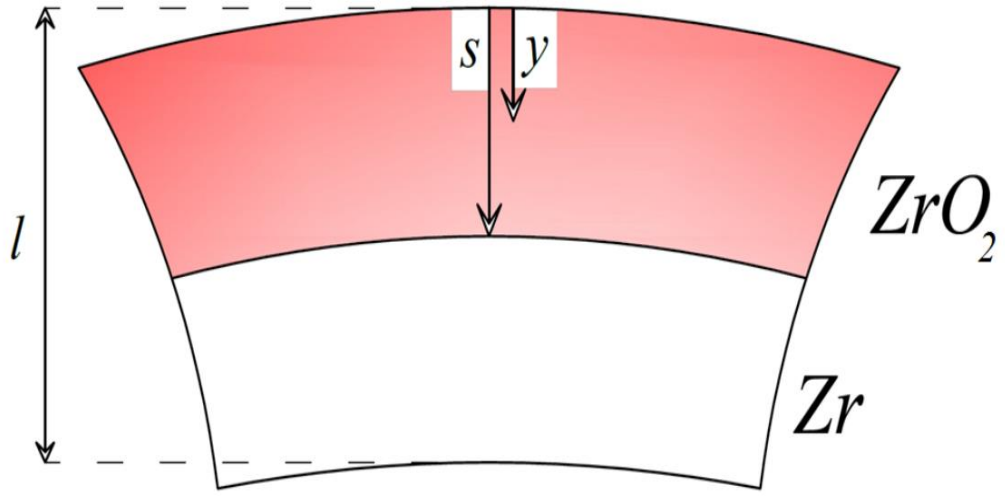


Joe Rachid_PNNL

Other factors:

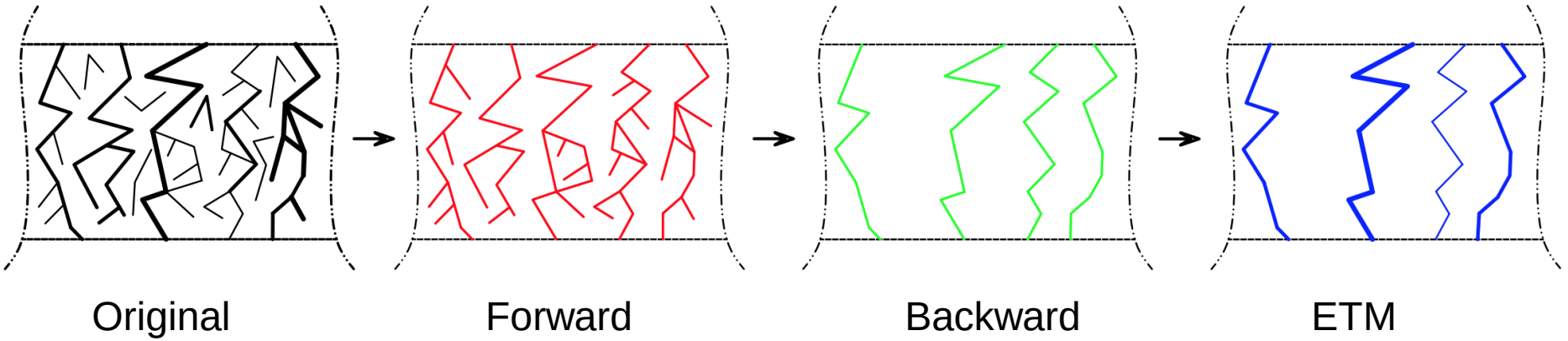
ii. Temperature gradients

iii. (irradiation)



Swelling

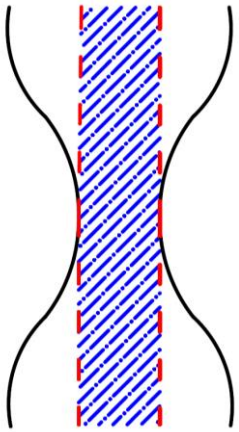
EFFECTIVE TRANSPORT MEDIUM (ETM)



1. Forward percolation

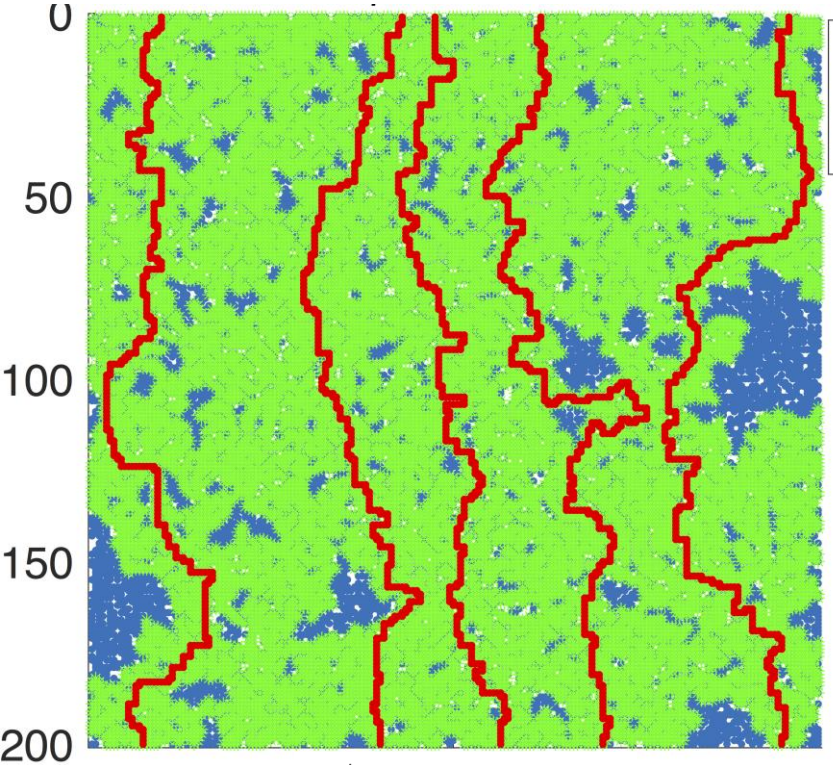
2. Backward percolation

3. Constrictive medium

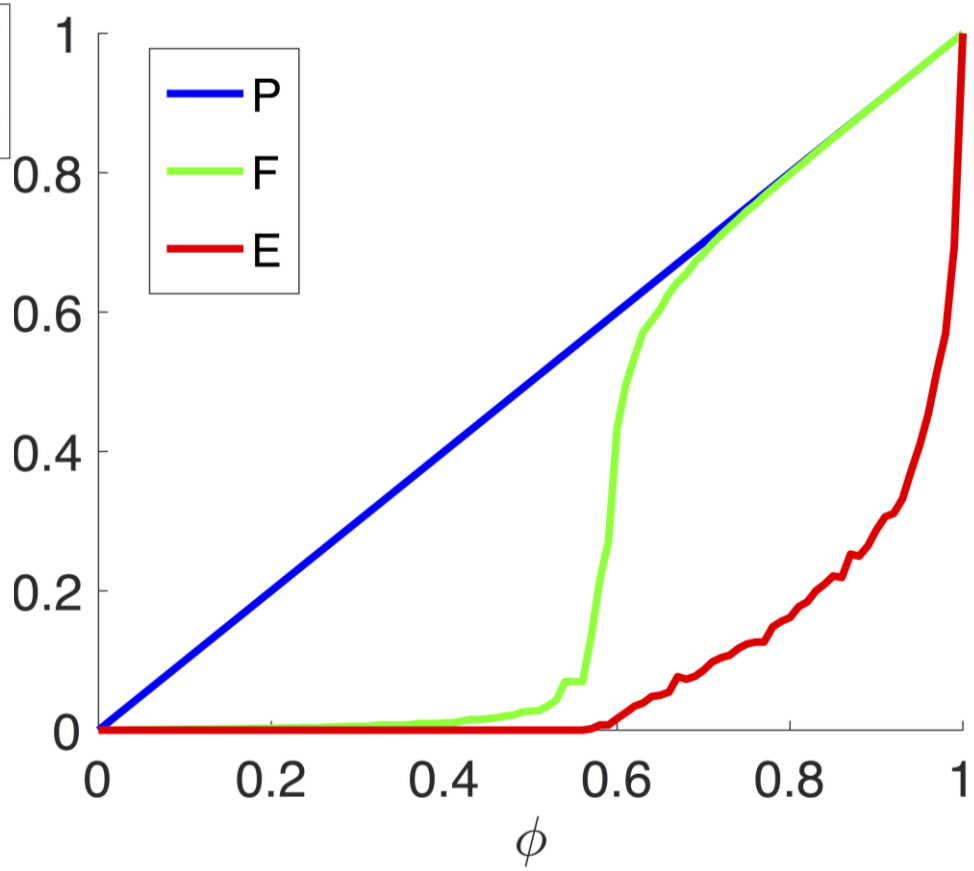


Extended Dijkstra's shortest path algorithm!

POROSITY ROLE



$\phi = 0.62$

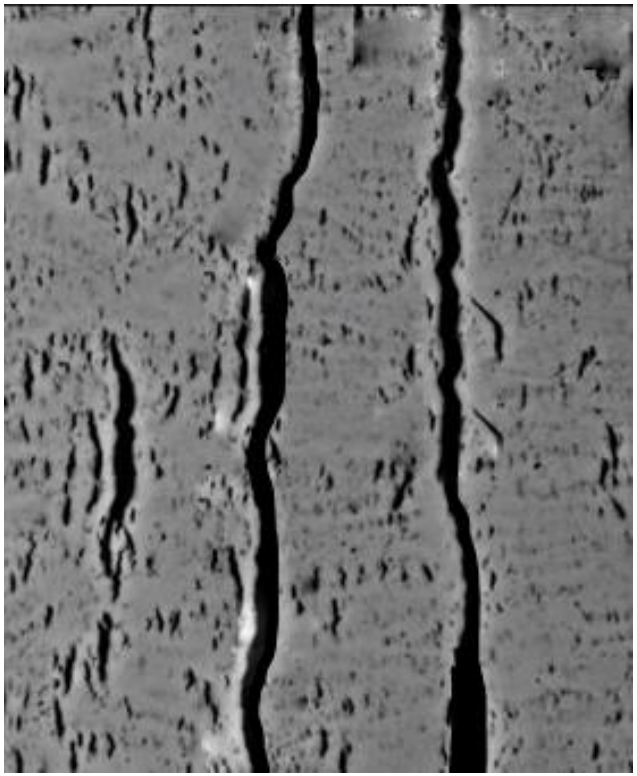


$$E \ll F < P$$

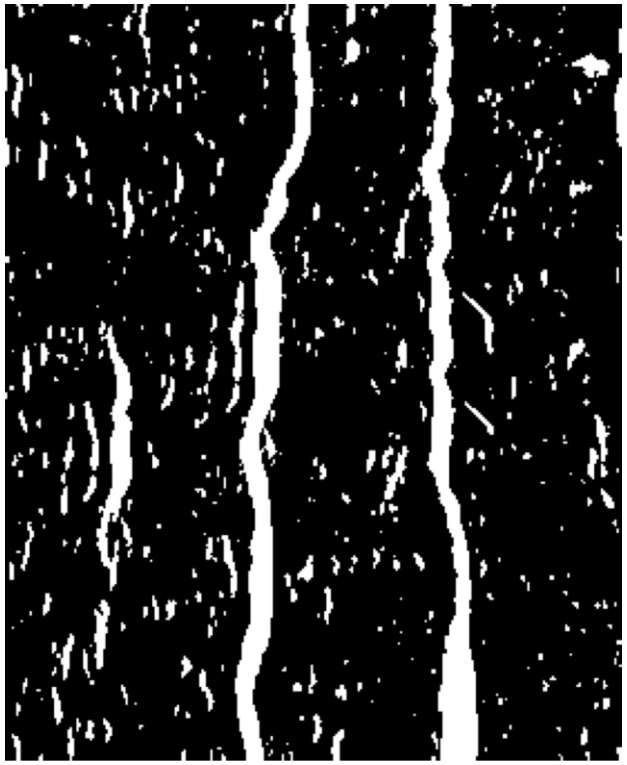
P: States
F: Forward
B: Backward + Constrictivity

$$D_{EFF} = \frac{D_w}{\sum_{i=1}^{n} m_i l_i} \sum_{k=1}^{n} \frac{m_k l_k}{\tau_k}$$

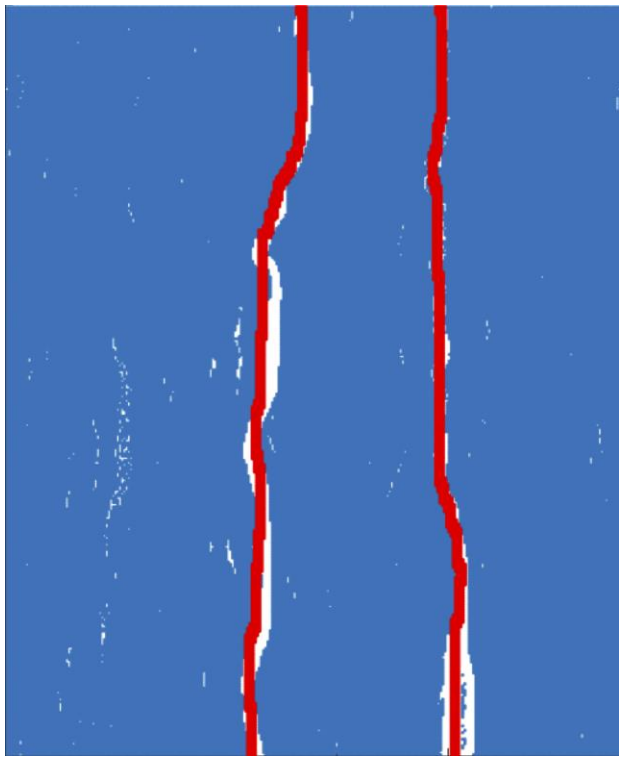
EXPERIMENTAL IMAGES



Original



Binarized



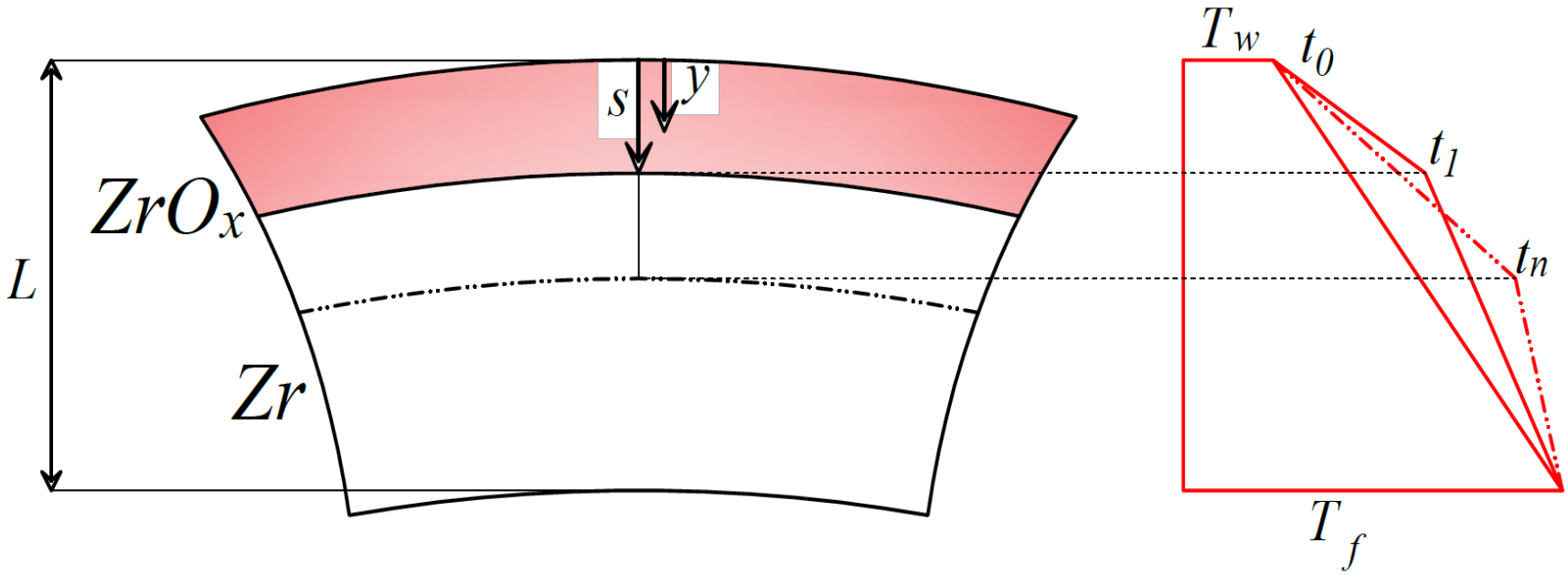
ETM

Otsu's method:

0 : black
1 : white

$$\begin{cases} \sigma^2(I) = w_0(I)\sigma_0^2(I) + w_1(I)\sigma_1^2(I) \\ w_0(I) + w_1(I) = 1 \end{cases}$$

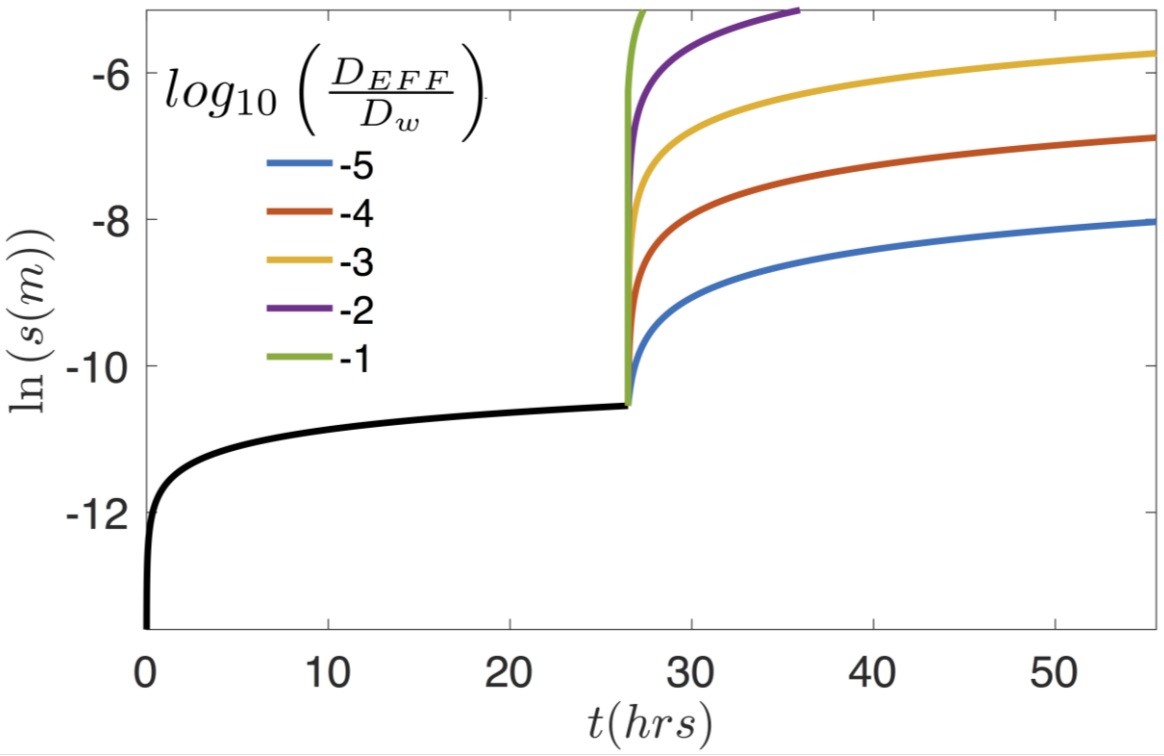
TEMPERATURE PROFILE

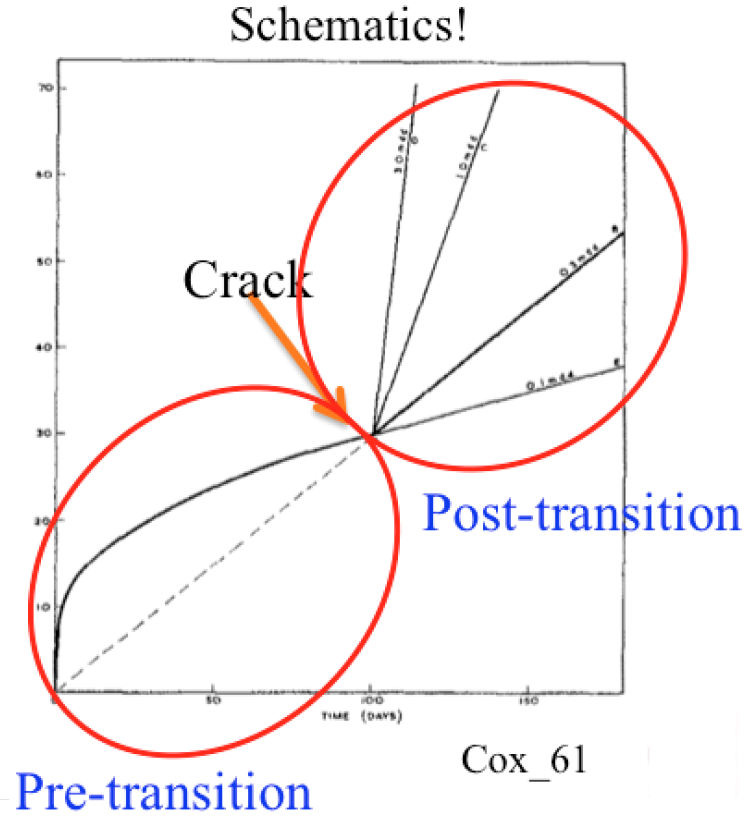


$$\begin{cases} \alpha = \dfrac{\kappa}{\rho c_p} \approx 10^{-5}\,\dfrac{m^2}{s} \\ D \approx 10^{-14}\,\dfrac{m^2}{s} \end{cases} \longrightarrow \frac{\partial T}{\partial t} \gg \frac{\partial [O]}{\partial t}$$

$$D^{ox} = D_0^{ox} \exp\left(\frac{-Q^{ox}}{k_B T}\right)$$

GROWTH KINETICS





Pre:

$$\frac{ds}{dt} = \frac{lD^{ox}}{s^2}$$

Post:

$$\frac{ds}{dt} = \frac{D^{EFF}}{s}$$

SUMMARY

1. A formulation for the effective transport medium based on Dijkstra's shortest path algorithm.

1. The post-transition growth kinetics of zirconium oxide is quantified.

2. The ETM is highly sensitive to original density of states at values close to unity.

THANK YOU!

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Advanced Analytical Model of Zr-Based Claddings Breakaway Oxidation in Steam-Oxygen-Nitrogen Mixtures

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Email: vasil@ibrae.ac.ru

The oxidation of zirconium-based claddings for nuclear reactor in steam-oxygen-nitrogen gas mixtures in temperature range 600-1760°C behaves in much more agressive way compared to oxidation in pure steam. It was clearly demonstrated by recent separate-effects tests as well as by integral experiments. Very complicated oxidation behaviour was observed in breakaway oxidation regime (about 600-1050°C) for which the oxide layer destruction and transition to linear kinetics are characteristic. In particular, those respectively low temperature conditions may be realized in the course of postulated spent fuel pool loss of coolant accident (SFP LOCA) at nuclear power plant (NPP). A deep understanding of such a complicated phenomenon is very important for adequate prediction of SFP LOCA progression.

The special advanced model of Zr-based alloys oxidation in this regime is developed. The model is based on coupled solution of oxygen diffusion and thermal mechanics problems in a cladding. It is supposed that the breakaway takes place when the hoop stress at external interface becomes larger the critical value. Three-layer or two-layer system consisting of porous ZrO_2+ZrN layer, α-Zr(O) and β-Zr layers is considered. Finally, the dependence of cladding oxidation rate on temperature and gas mixture composition is derived.

The model predictions are compared with the results of separate effects tests on oxidation in steam-oxygen-nitrogen gas mixtures. The calculated results are in a reasonable agreement with experimental data which justifies the adequacy of modelling capabilities of advanced analytical model for application to zirconium-based claddings oxidation in breakaway regime.


t für Technologie

Advanced Analytical Model of Zr-Based Claddings Breakaway Oxidation in Steam-Oxygen-Nitrogen Mixtures

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Nuclear Safety Institute of Russian Academy of Sciences (IBRAE),
B.Tulskaya 52, 115191 Moscow, Russia

Content of Presentation

1. Brief Description of Breakaway Cladding Oxidation

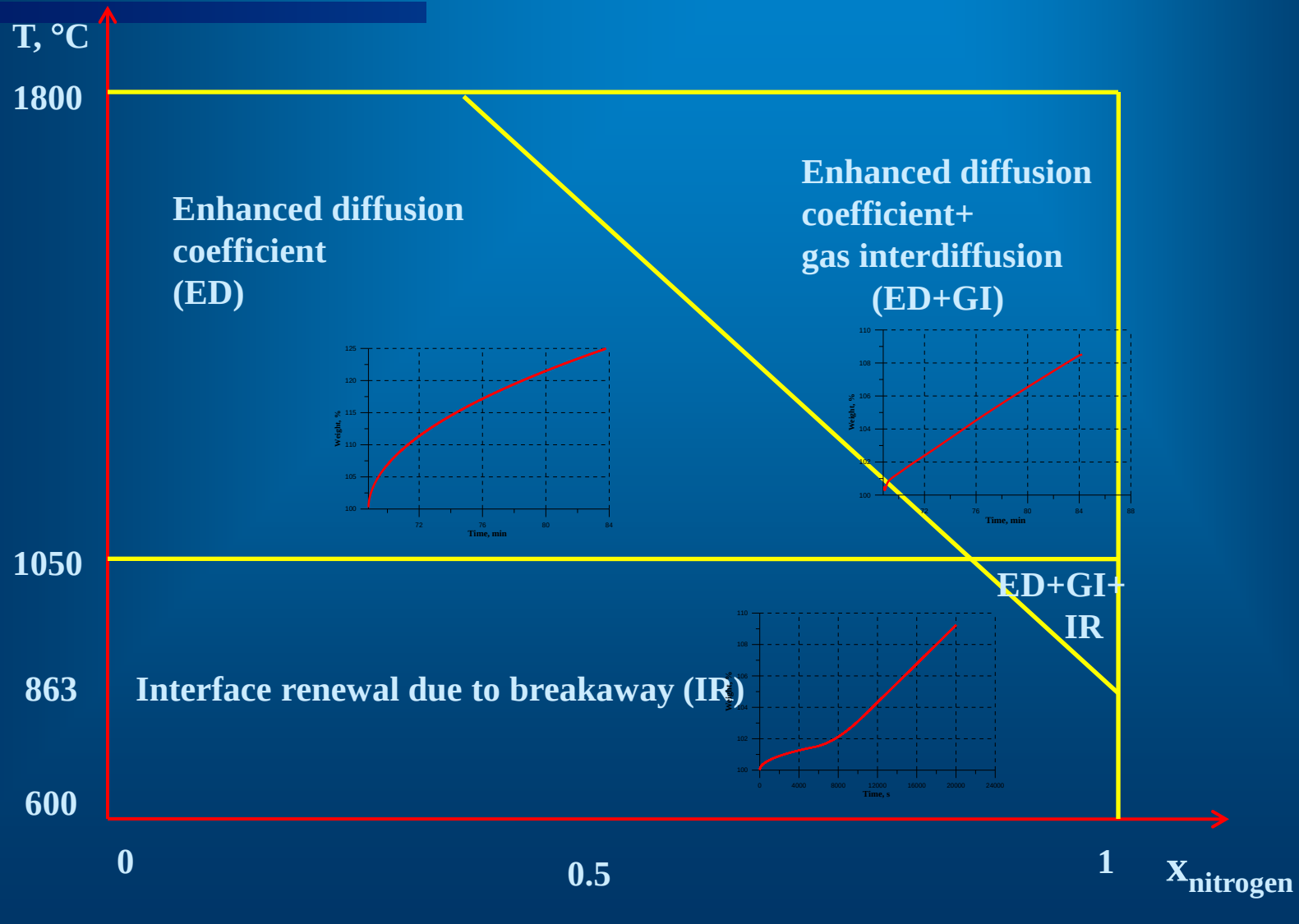
2. Thermo-Mechanical Model of Breakaway

3. Coupled Model to Calculate Breakaway Oxidation in O_2-N_2 and H_2O-N_2 Gas Mixtures

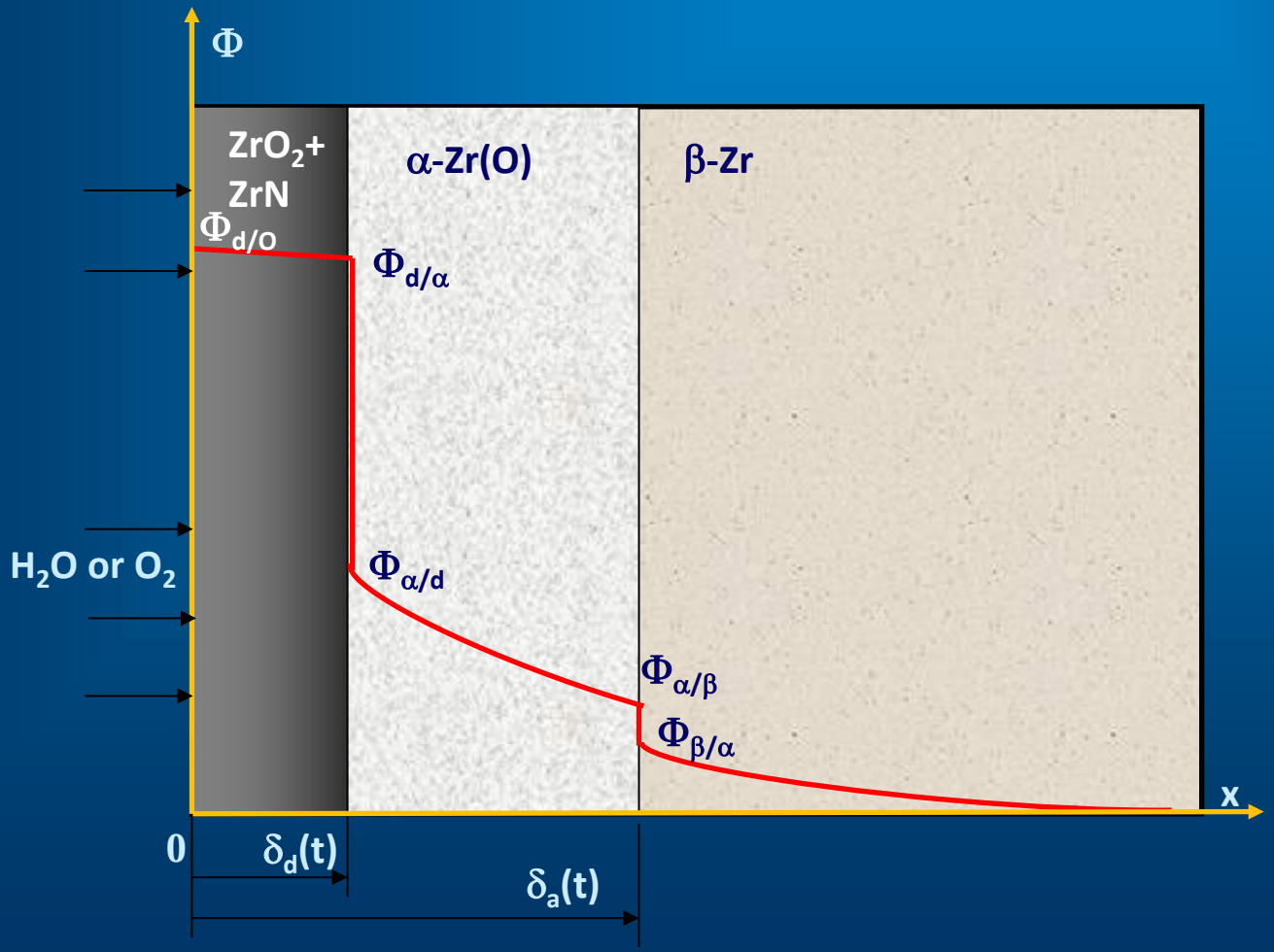
4. Model Application to Separate Effects Tests

5. Conclusions

Claddings Oxidation Model Type Map in Steam-Oxygen-Nitrogen Mixtures



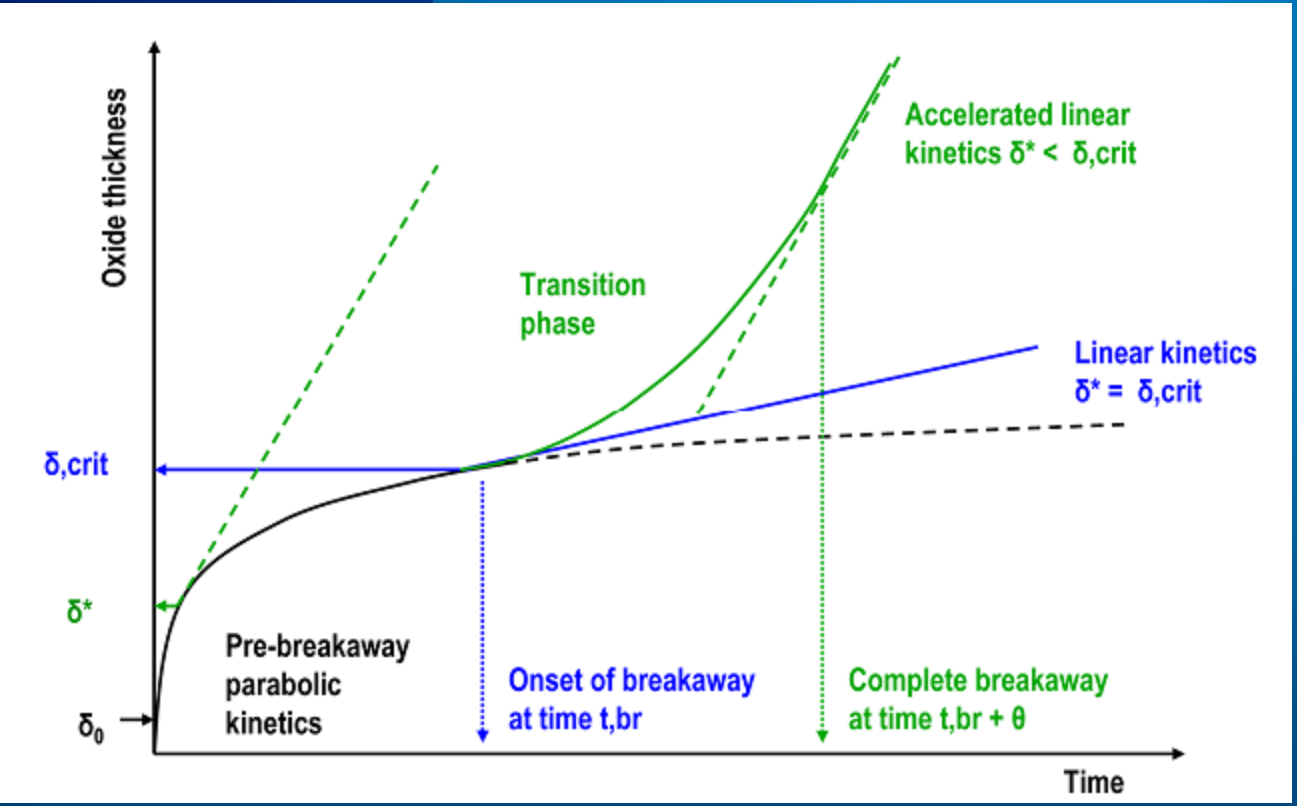
Three-Layers System
T>863°C



Two-Layers System
T<863°C



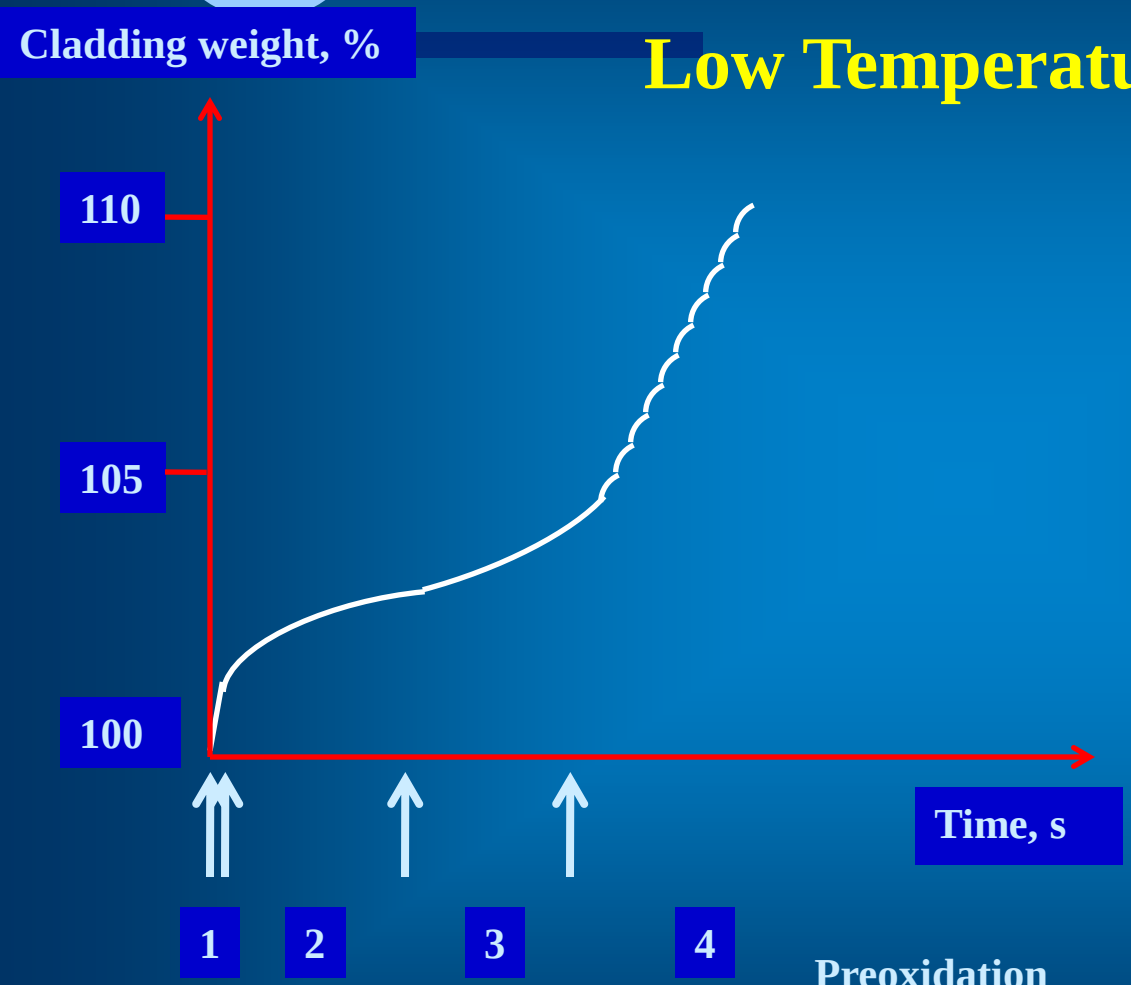
PSI Model of Zircaloy-4 Oxidation in Air



1. B. Jaeckel, J. Birchley, L. Fernandez-Moguel. Spent Fuel Pool under Severe Accident Conditions. Proc. 22nd Int. Conf. on Nuclear Engineering (ICONE22), July 7-11, 2014, Prague, Czech Republic. ICONE22-30729.

2. J. Birchley, L. Fernandez-Moguel, B. Jaeckel. Extended options to PSI oxidation model for Zircaloy-4 (2013-2014): input specification and assessment summary. Report TM-42-14-09, June 2014, PSI, Switzerland.

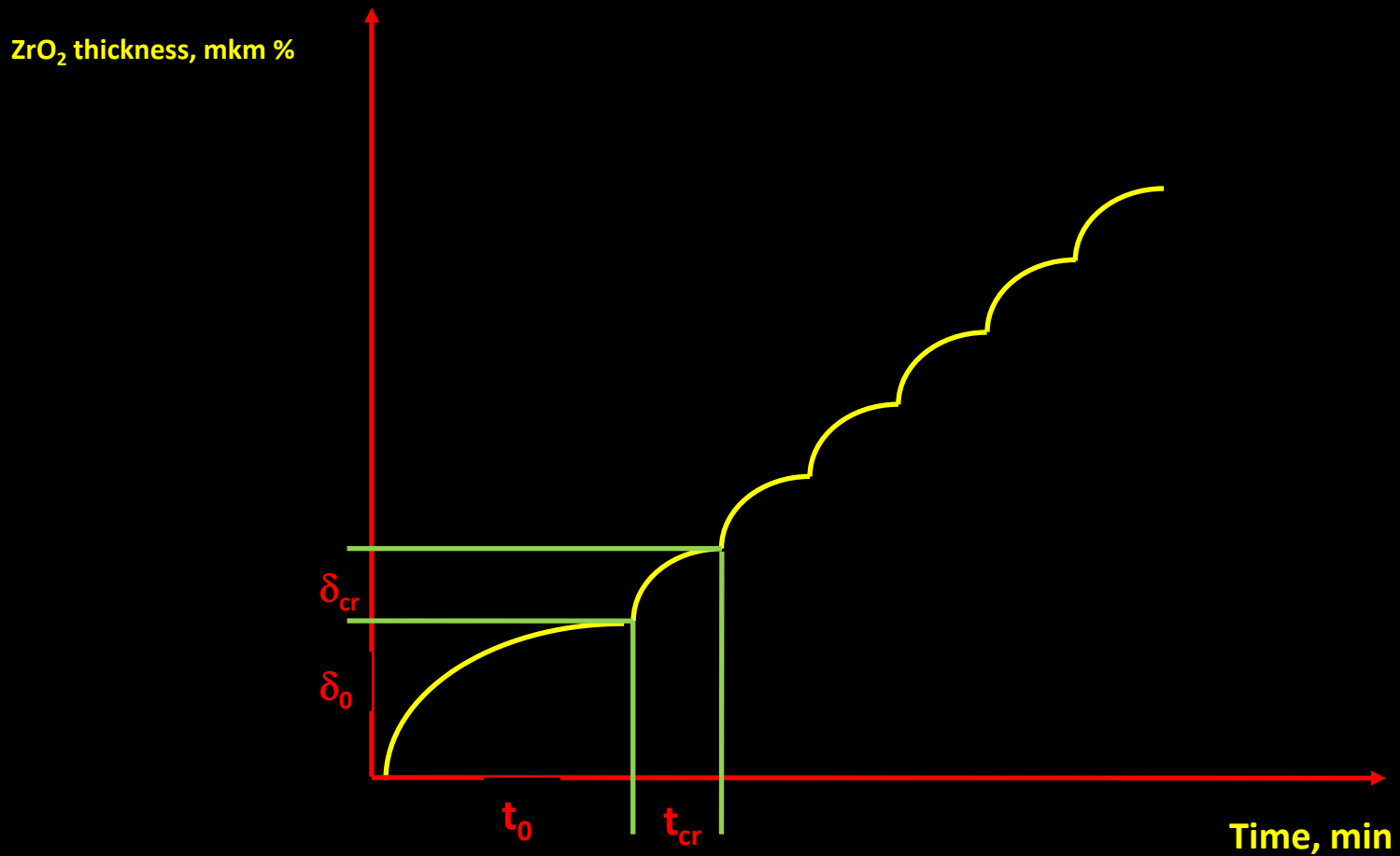
Small Consecutive Parabolas Correspond to Linear Dynamics! Low Temperatures 600÷1050°C



1 – adsorption
2 – first parabola
3 – transition
4 – small consecutive parabolas

Surface Renewal Model

$$\delta^* = K_d \sqrt{t^*}$$



Non-Dimensionless Variables

$$\delta_d = K_d \sqrt{t}$$

$$\tau = \frac{t}{t^*}$$

Non-dimensionless time

$$\delta^* = K_d \sqrt{t^*}$$

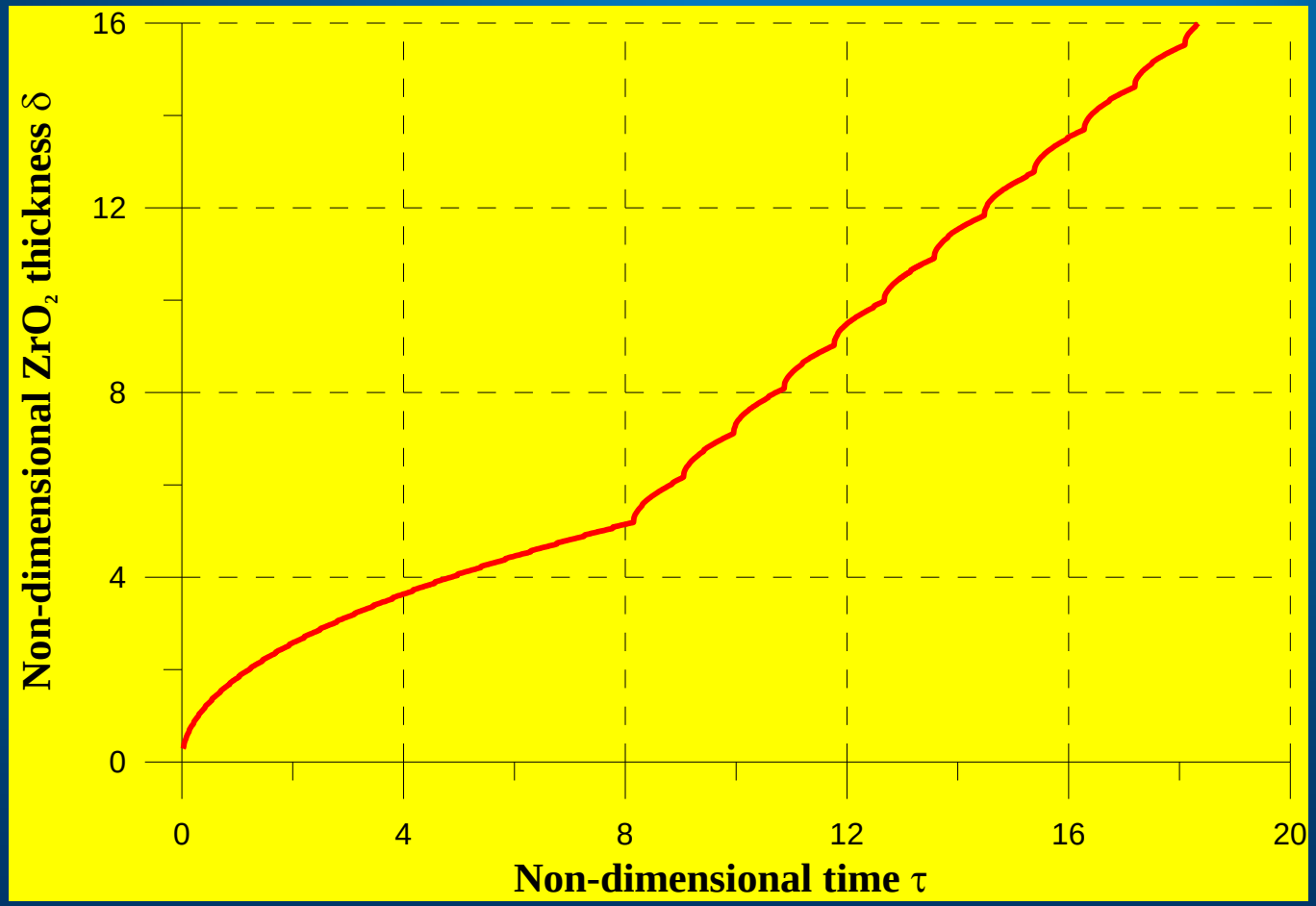
$$\delta = \frac{\delta_d}{\delta^*}$$

Non-dimensionless ZrO_2 layer thickness

$$V = \frac{\delta^*}{t^*} = \frac{K_d^2}{\delta^*}$$

ZrO_2 layer growth velocity

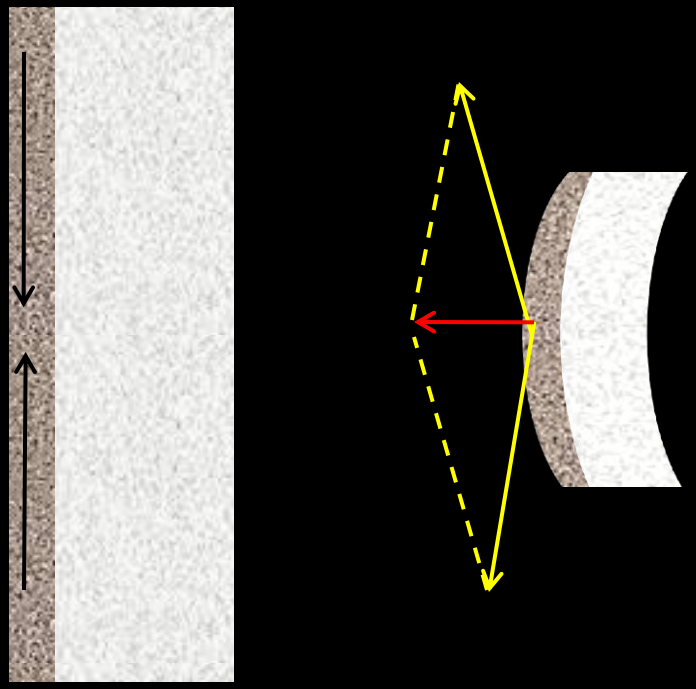
ZrO_2+ZrN Layer Thickness in Non-Dimensionless Variables τ-δ



Why Degradation Takes Place? Comressive Force Acts to Zirconium Dioxide Layer

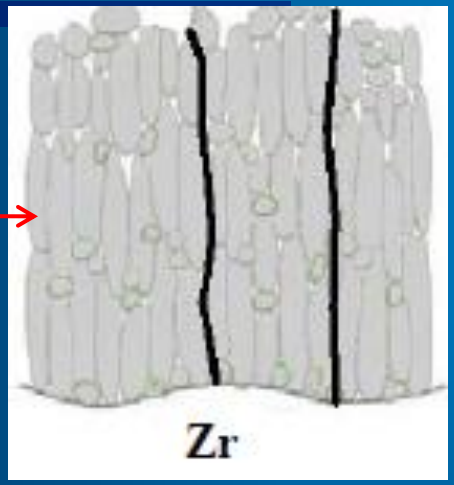
ZrO_2 + ZrN

α-Zr(O)



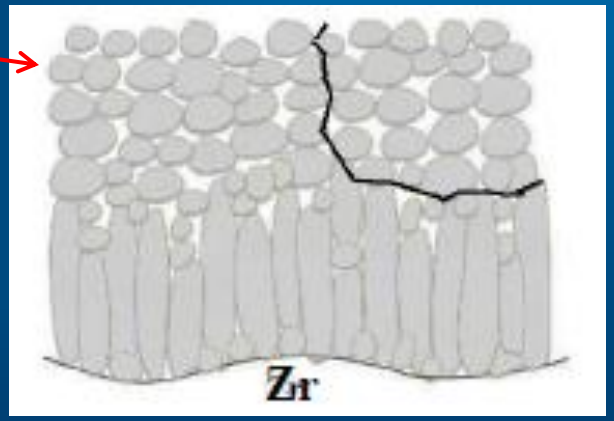
The ratio of the molar volumes of ZrO_2 to Zr (Pilling-Bedworth number) is *1.56* so that the growing oxide film is considered to be under compression.

Degradation Mechanisms of Zr-Based Claddings (E-110)



Break-up





Breakaway Spallation



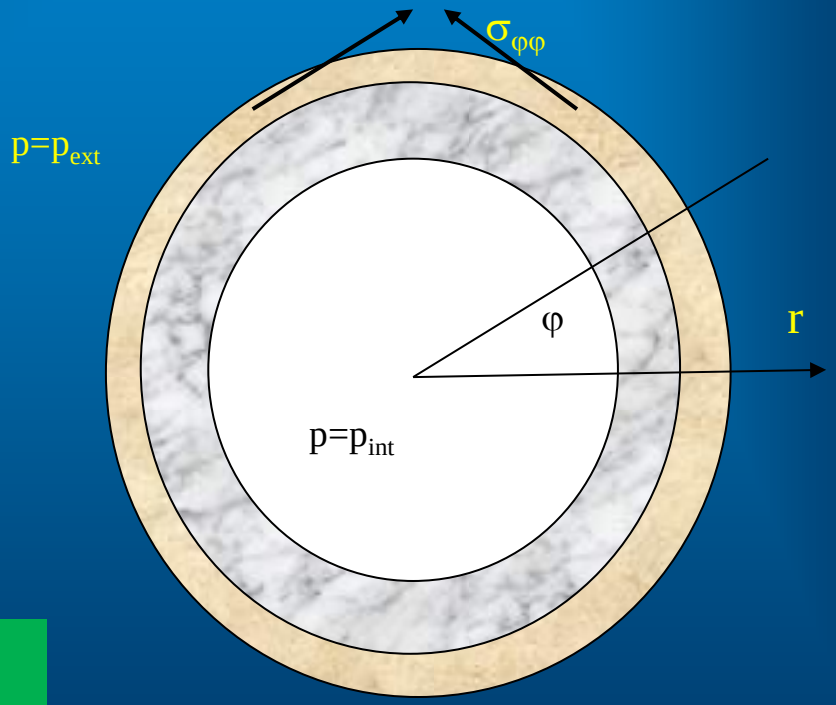
Koteneva M.V. Structure and Degradation of Zr alloys oxide films. PhD Thesis, MISIS, Moscow, 2014.

Diagonal Components of Deformation Tensor ε_{ik} in a Cylindrical Geometry

$$\varepsilon_{rr} = \frac{1}{E}\left(\sigma_{rr} - \nu\left(\sigma_{\phi\phi} + \sigma_{zz}\right)\right)$$

$$\varepsilon_{\phi\phi} = \frac{1}{E}\left(\sigma_{\phi\phi} - \nu\left(\sigma_{rr} + \sigma_{zz}\right)\right)$$

$$\varepsilon_{zz} = \frac{1}{E}\left(\sigma_{zz} - \nu\left(\sigma_{rr} + \sigma_{\phi\phi}\right)\right)$$



E Young modulus

ν Poisson coefficient

Equilibrium Conditions

$$\frac{\partial \sigma_{rr}}{\partial r} + \frac{\sigma_{rr} - \sigma_{\phi\phi}}{r} = 0$$

$$\sigma_{\phi\phi} = \sigma_{rr} + r\frac{\partial \sigma_{rr}}{\partial r}$$

$$\frac{\partial \sigma_{\phi\phi}}{\partial \phi} = 0$$

$$\sigma_{zz} = \nu\left(\sigma_{rr} + \sigma_{\phi\phi}\right)$$

$$\frac{\partial \sigma_{zz}}{\partial z} = 0$$

Basic Equation for Determination of σ_{rr}

$$\varepsilon_{rr} = \frac{\partial u_r}{\partial r}$$

$$\varepsilon_{\phi\phi} = \frac{u_r}{r}$$

$$\varepsilon_{zz} = \frac{\partial u_z}{\partial z} = 0$$

$$\varepsilon_{rr} = \frac{\partial u_r}{\partial r} = \frac{\partial}{\partial r}\left(r\varepsilon_{\phi}\right) = \varepsilon_{\phi} + r\frac{\partial \varepsilon_{\phi}}{\partial r}$$

$$\frac{\left(1-\nu^2\right)}{E} r^2 \frac{d^2\sigma_{rr}}{dr^2} + \frac{3\left(1-\nu^2\right)}{E} r \frac{d\sigma_{rr}}{dr} = 0$$

$$\sigma_{rr} = -p_{int} \qquad r = r_{int}$$

$$\sigma_{rr} = -p_{ext} \qquad r = r_{ext}$$

Clarke's Model for Hoop Deformation

$$\dot{\varepsilon}_{\varphi\varphi} = \dot{\varepsilon}_{\varphi\varphi,elastic} + D_{ox}\dot{\delta}$$



due to Pilling-Bedworth number

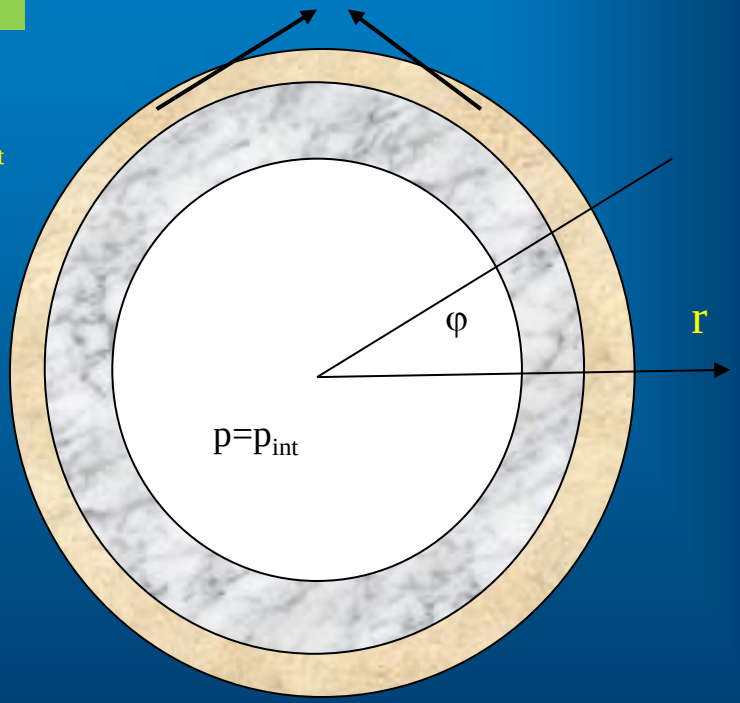
Solution of Equation

$$\sigma_{rr} = A_1 + \frac{B_1}{r^2} + C \ln \frac{r}{r_{ext}}$$

$r_{ext} \geq r \geq r_{ext} - \delta_d$

$$\sigma_{rr} = A_2 + \frac{B_2}{r^2}$$

$r_{ext} - \delta_d \geq r \geq r_{int}$.



Approximate Estimation of Critical Zro2+ZrN Layer Compression at Breakaway

$$\sigma_{\varphi\varphi} \approx D_{ox} \frac{E_d}{1-\nu^2} \delta_d$$

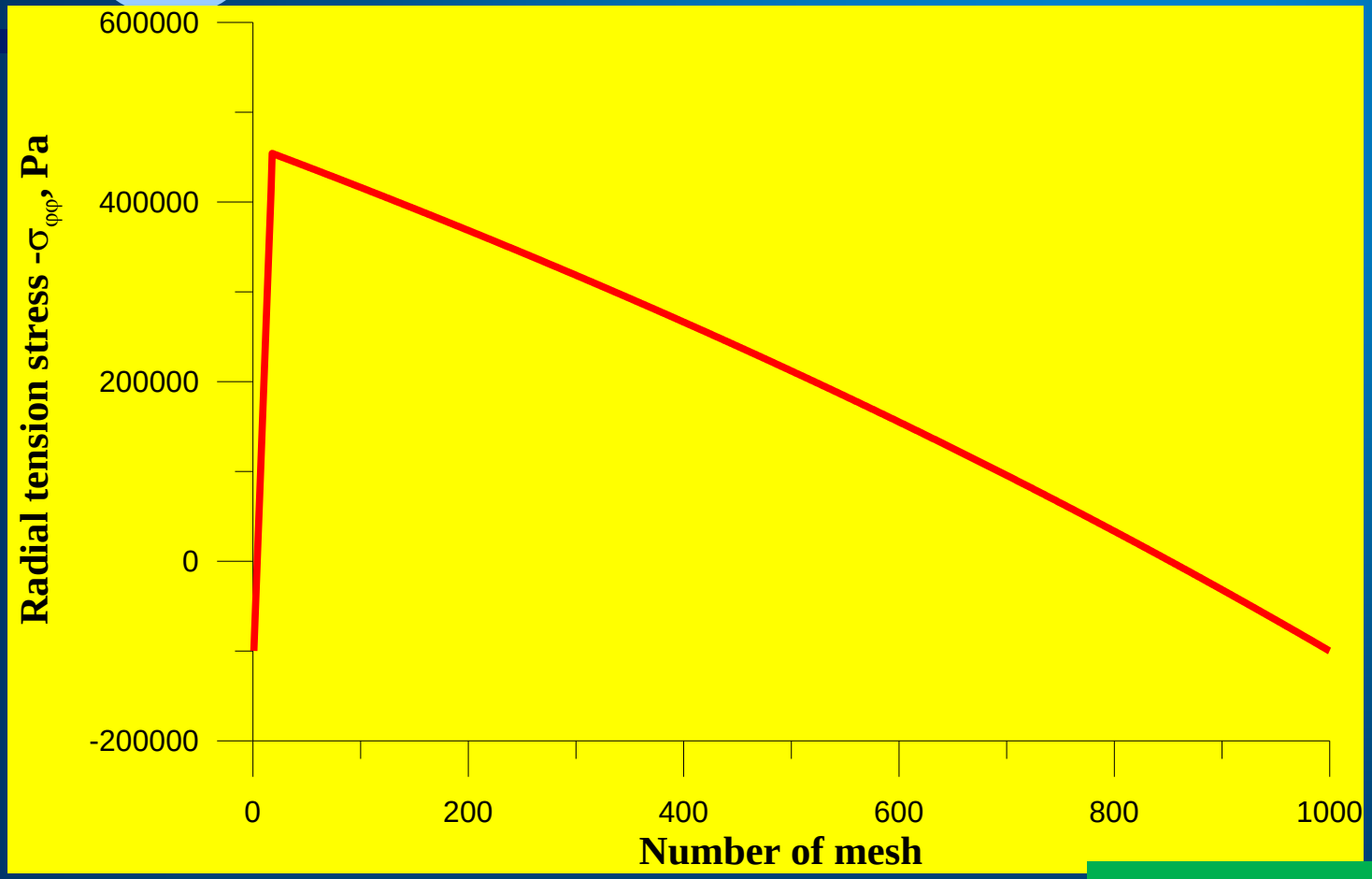
$\sigma_{\varphi\varphi} \cong \sigma_{break}$

Young modulus	$E_d \cong 172$ GPa
at room T	$E_{met} \cong 98$ GPa

$\sigma_{break} \approx 96.28 \cdot 10^6$ Pa

SCDAP/RELAP5/MOD2 Code Manual, Volume 4: MATPRO-A Library of Materials Properties for Light-Water-Reactor Accident Analysis. NUREG/CR-5273 EGG-2555. Vol.4., 1990

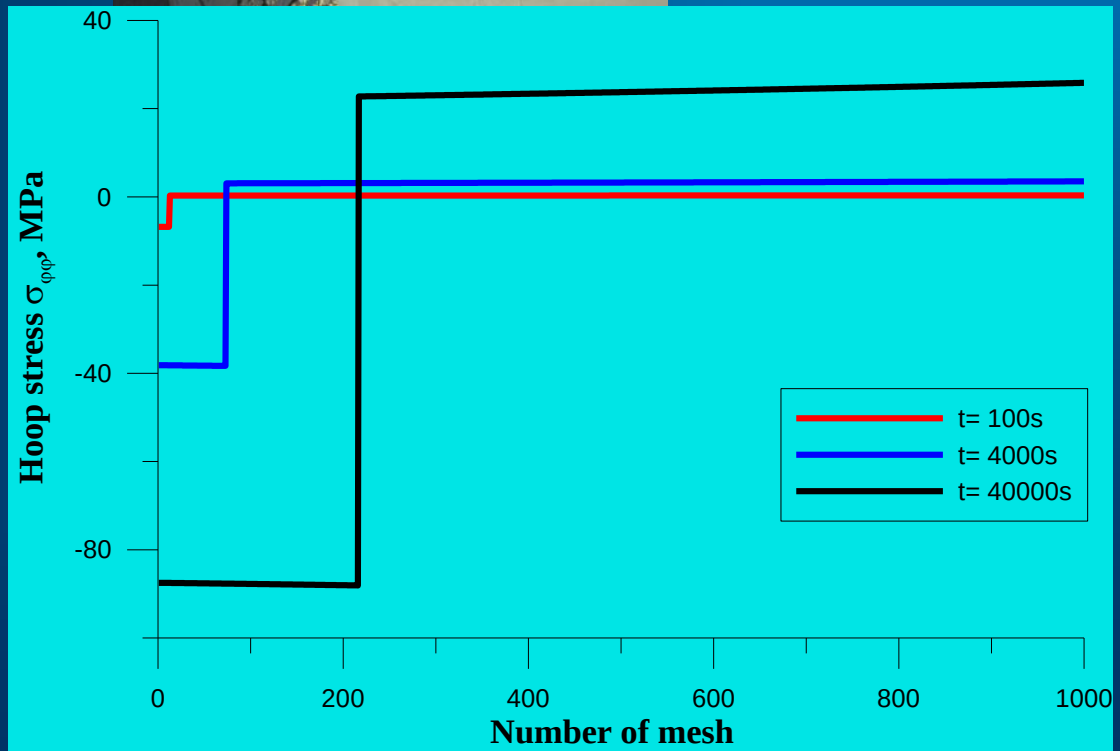
Radial Stress across Cladding



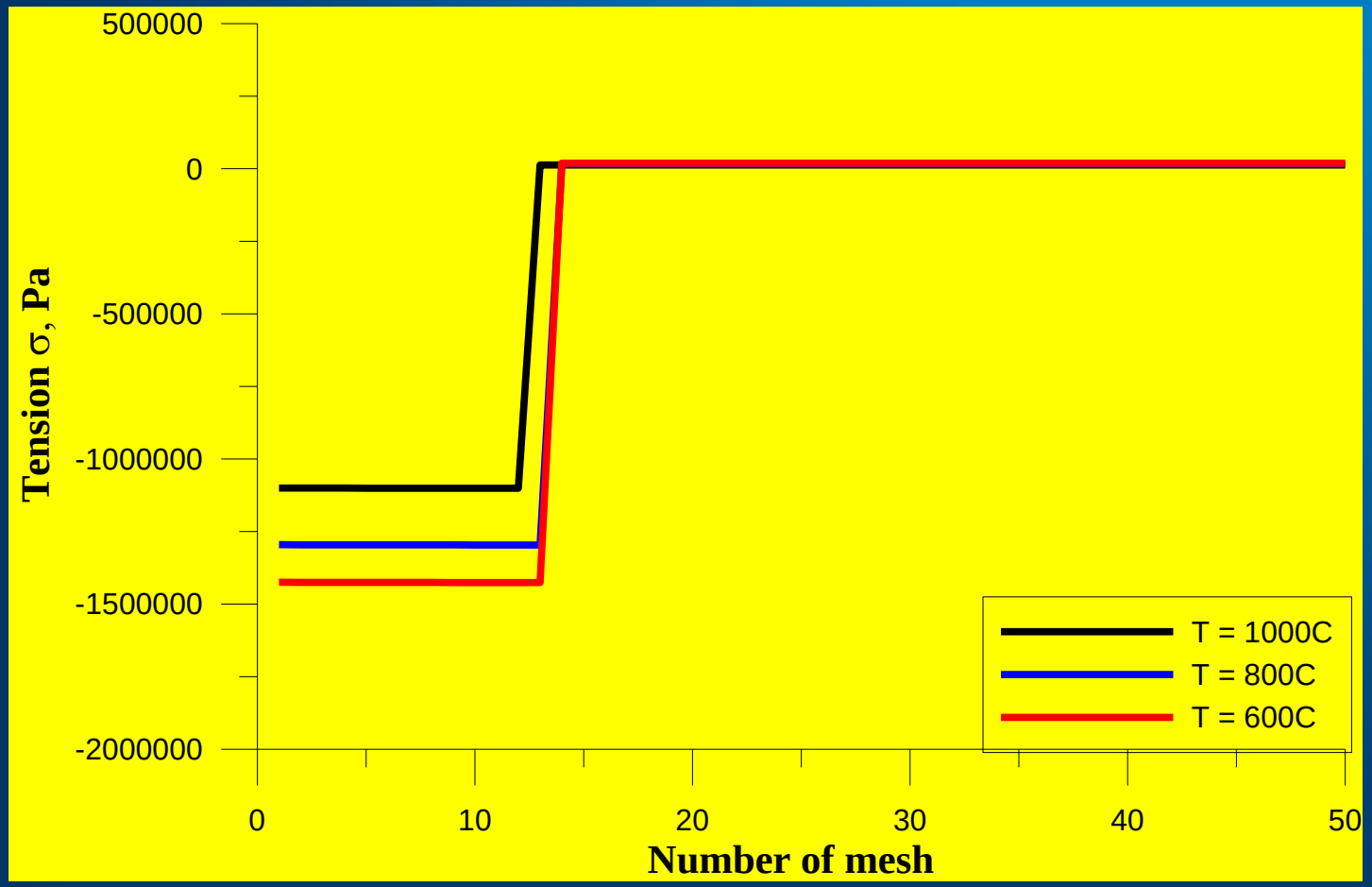
$$\sigma_{\phi\phi} = \sigma_{rr} + r\frac{\partial \sigma_{rr}}{\partial r}$$

Compression and Tension Dynamics. T=1000°C





Hoop Tension: Comparison between Different Temperatures

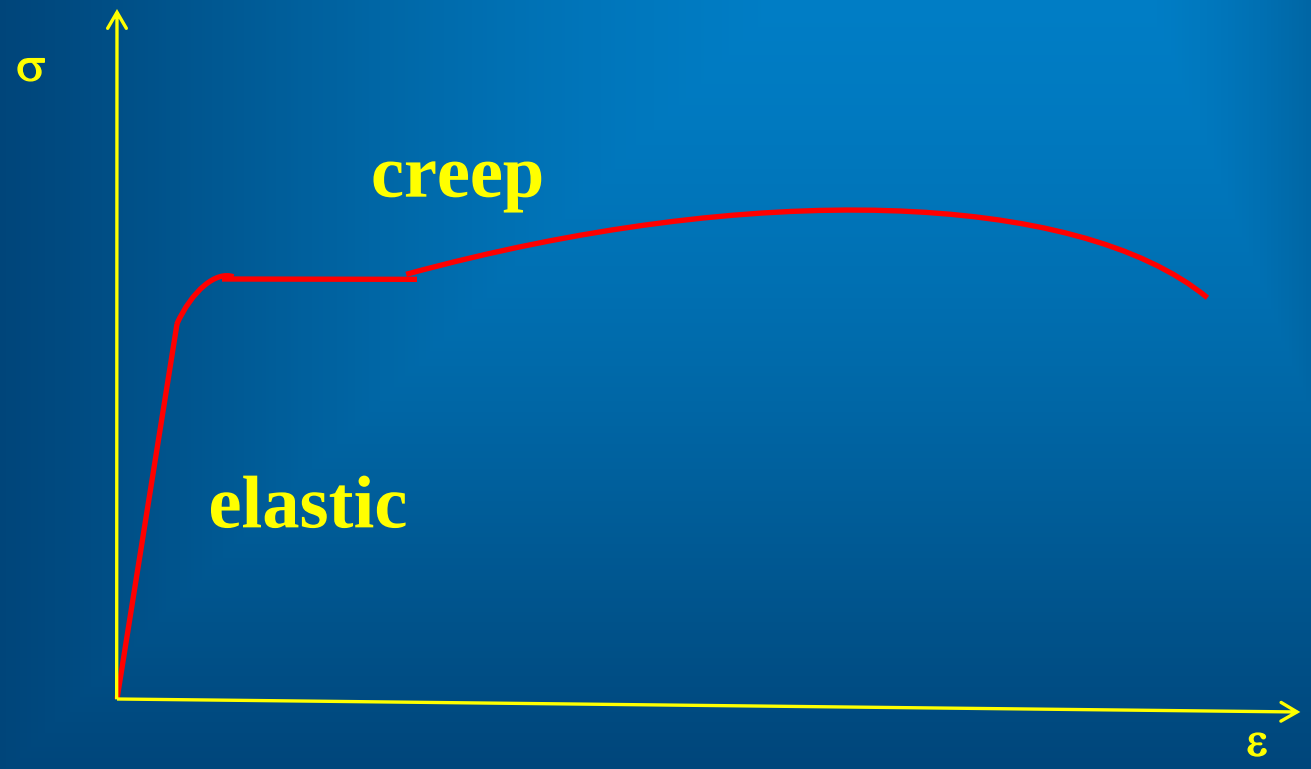


t=100 s T=1000°C

t=1900 s T=800°C

t=140000 s T=600°C

Diagram ε-σ



Coble's Model Creep Strain Rate

$$\dot{\varepsilon} = \frac{\alpha_{CO}\delta D_{gb}}{kT} \cdot \frac{\Omega\pi}{g^3} \cdot \sigma$$

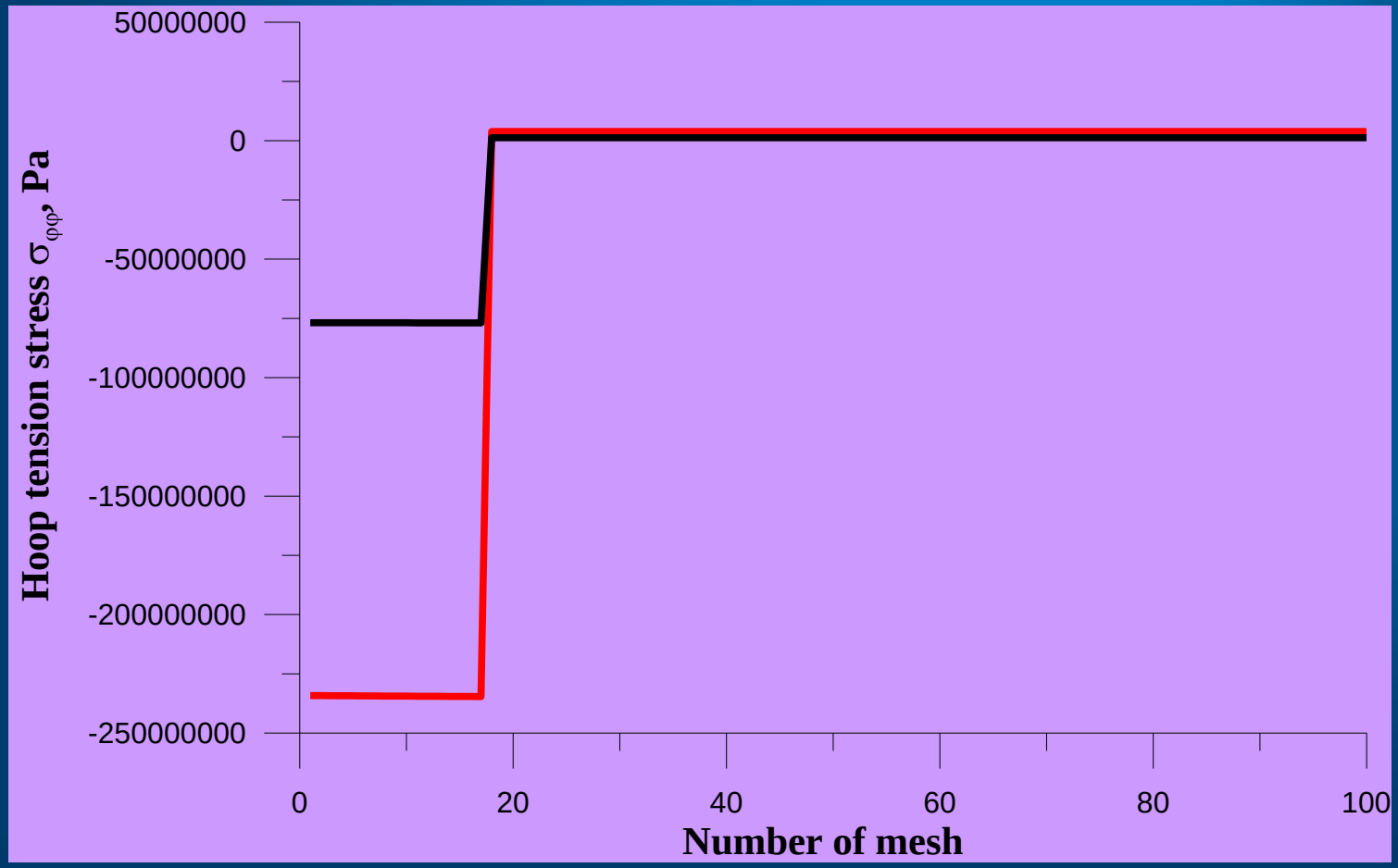
$$\delta D_{gb} = D_o \exp\left[-\frac{Q_{gb}}{RT}\right]$$

α_{CO}	const
δ	grain boundary width
D_{gb}	grain boundary diffusion coefficient
k	Boltzmann const
T	temperature
Ω	atomic volume of Zr
g	average grain size

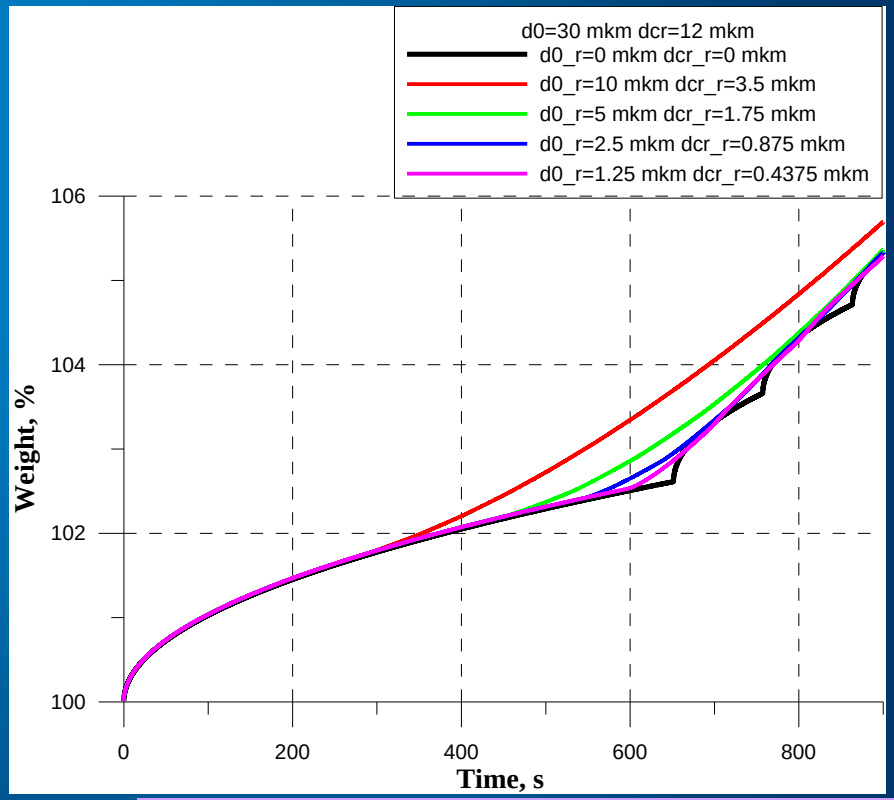
$D_0 = 1.0 \cdot 10^{-4}$ mm^3/s

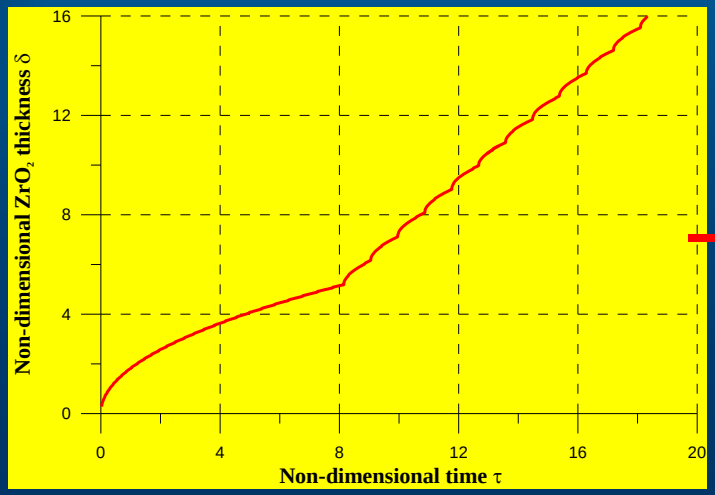
Qgb = 144 kJ/mole

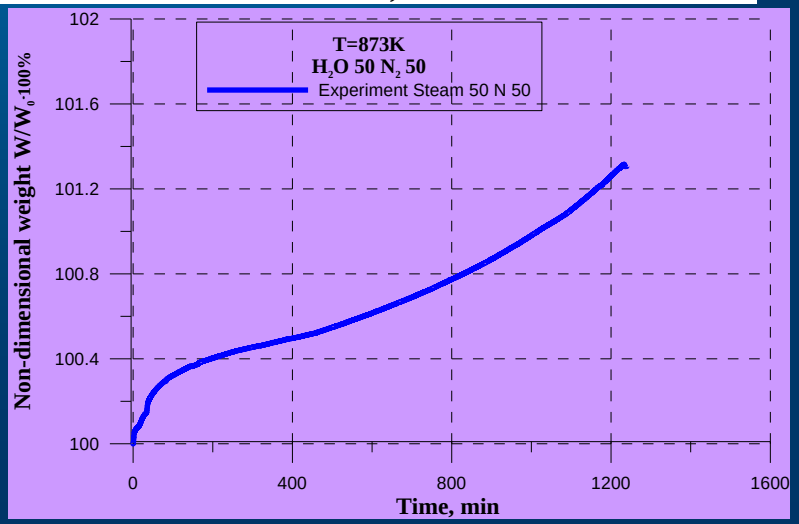
Addition of Creep Strain will Result in Lowering Compression Stress



Averaging on Ensemble







Comparison with separate effects tests

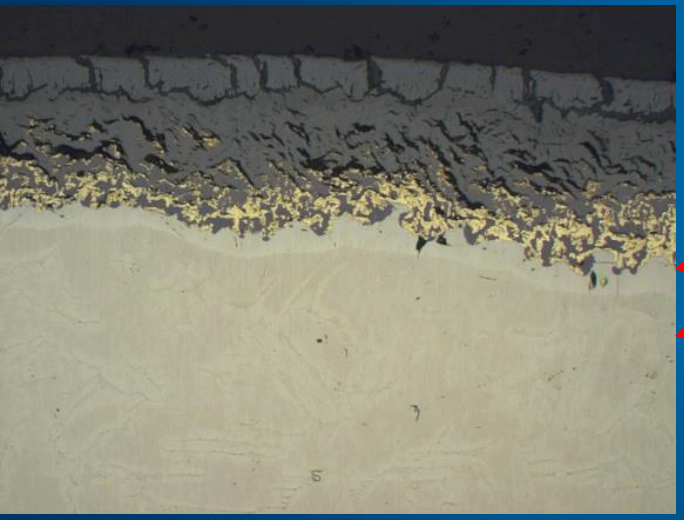
High-temperature oxidation of Zircaloy-4 in steam-nitrogen mixtures.

M. Steinbrück*, F. Oliveira da Silva, H.J. Seifert

NuMat2014: The Nuclear Materials Conference, 27-30 October 2014, Clearwater Beach, Florida, USA.

1000°C and 800°C

1000°C, 5%O_2, 95%N_2



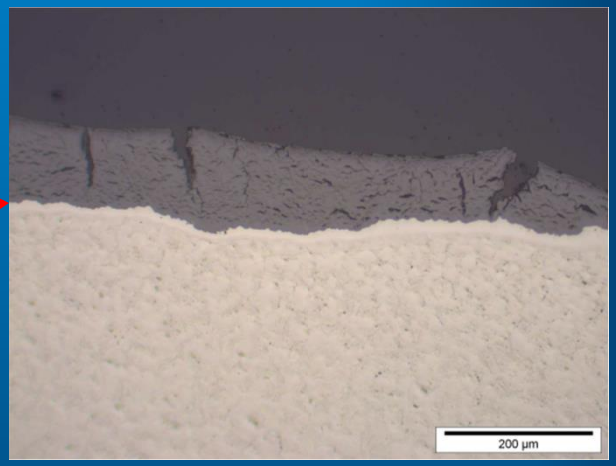
ZrO_2+ZrN

α-Zr(O)

β-Zr

α-Zr

800°C, 99%H_2O, 1%N_2

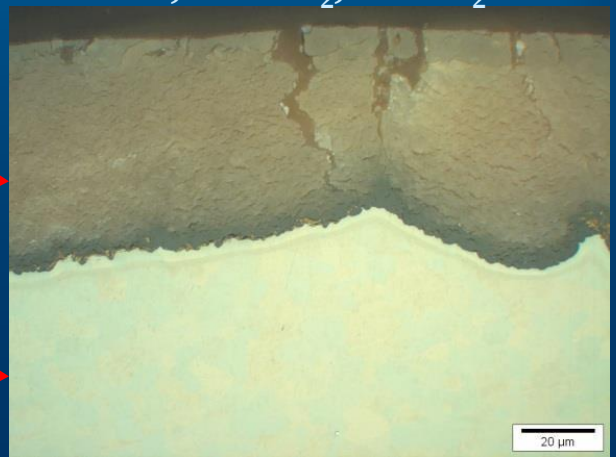


800°C, 60%O_2, 40%N_2

ZrO_2+ZrN

α-Zr(O)

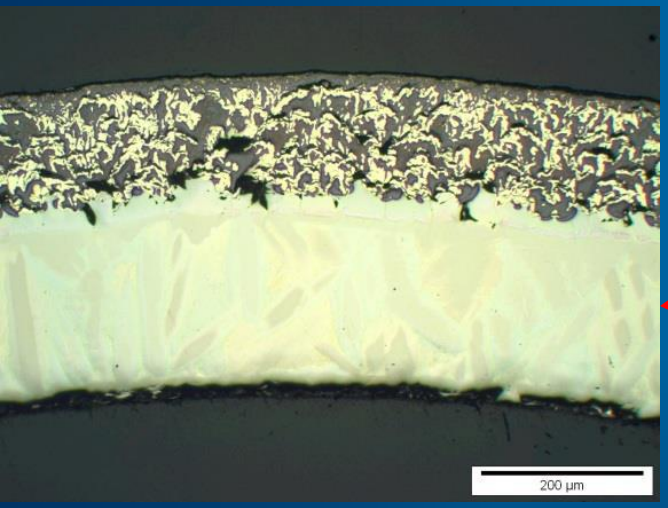
α-Zr



1200°C and 600°C

1200°C, 5%O_2, 95%N_2

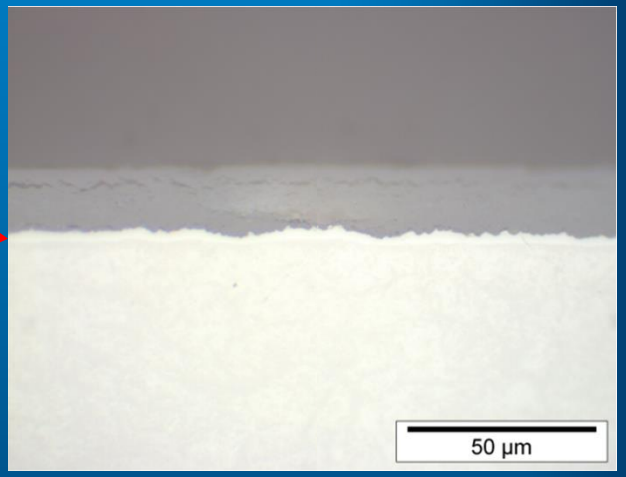
600°C, 99%H_2O, 1%N_2



ZrO_2+ZrN

α-Zr(O)

β-Zr α-Zr

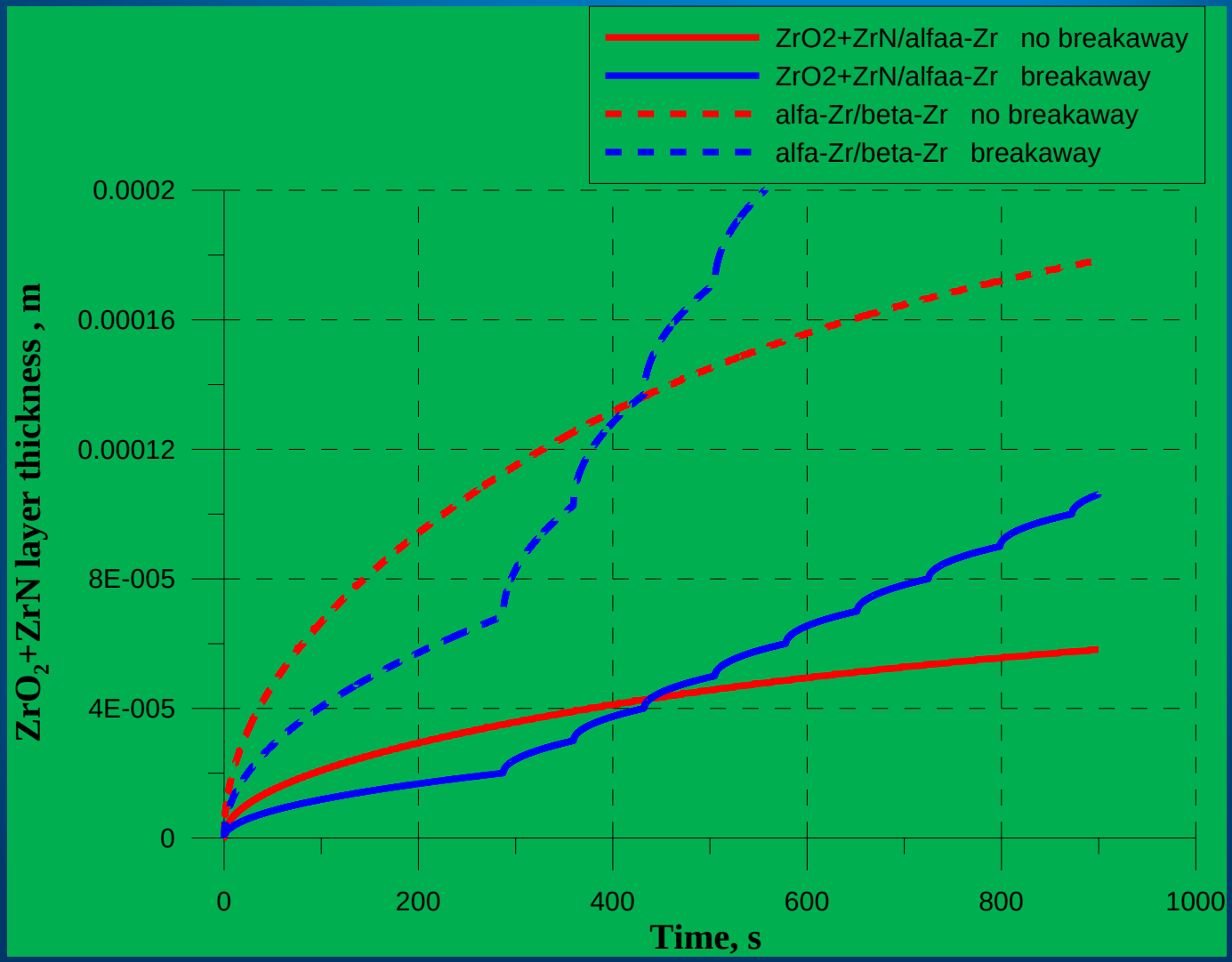


In reality, ZrN at 600°C is not descernible!

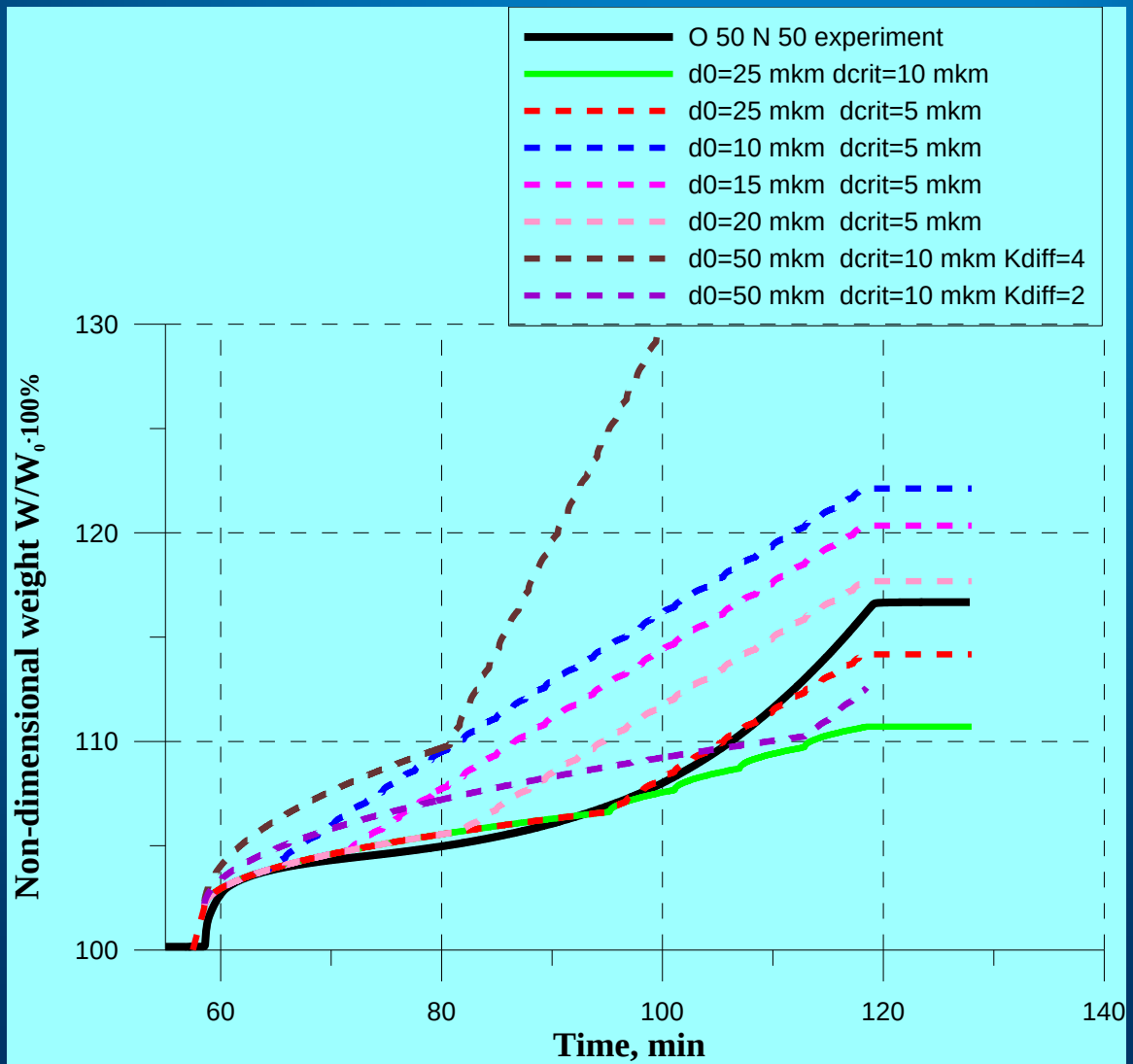
In all cases oxygen diffusion coefficient through ZrO_2+ZrN layer is enhanced!

$$D_{d,eff} = K_{diff} D_d$$

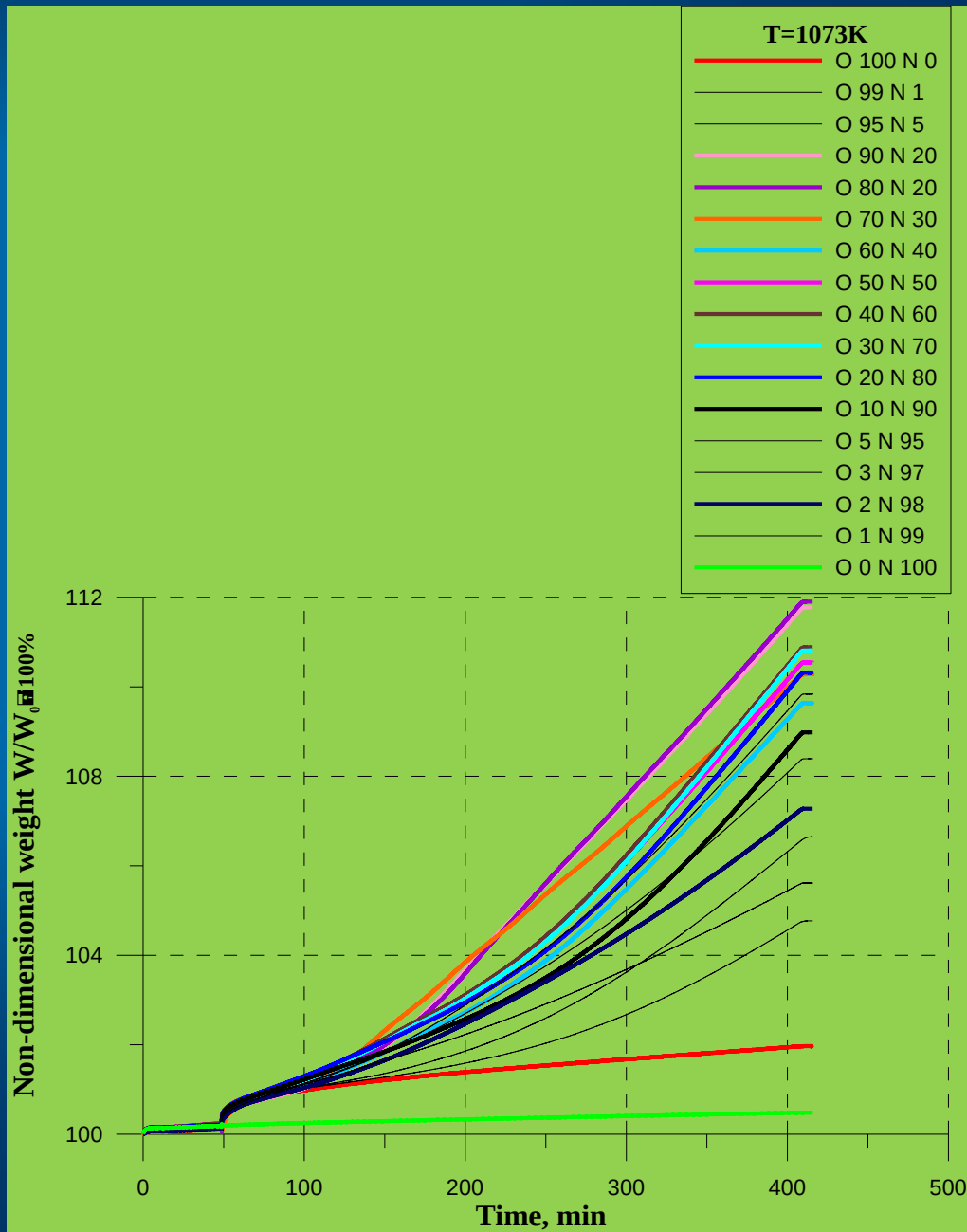
No Breakaway and Breakaway Dynamics



Change of Model Parameters Will Change a Shape of Oxidation Kinetics

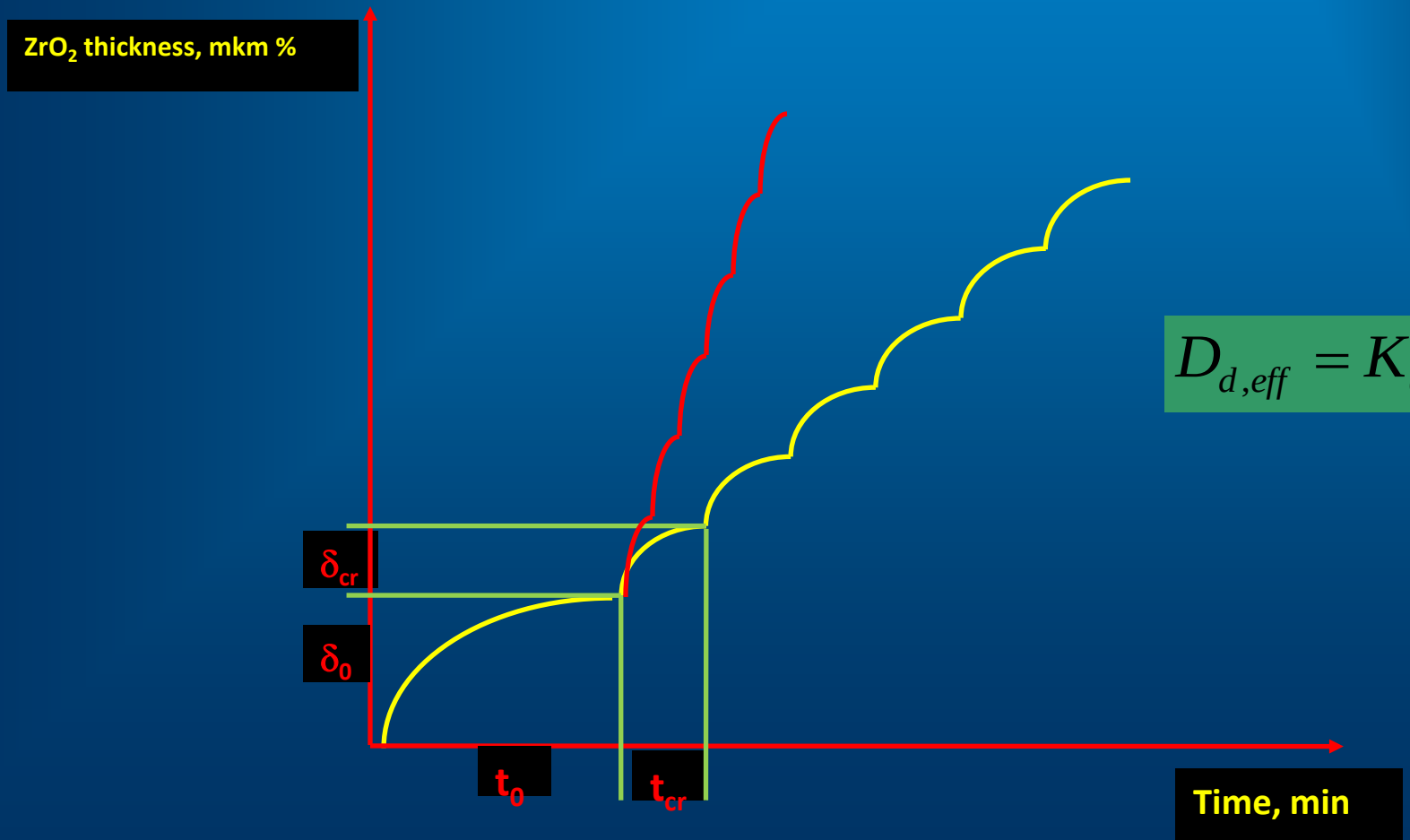


Experimental Weight Gain T=800°C



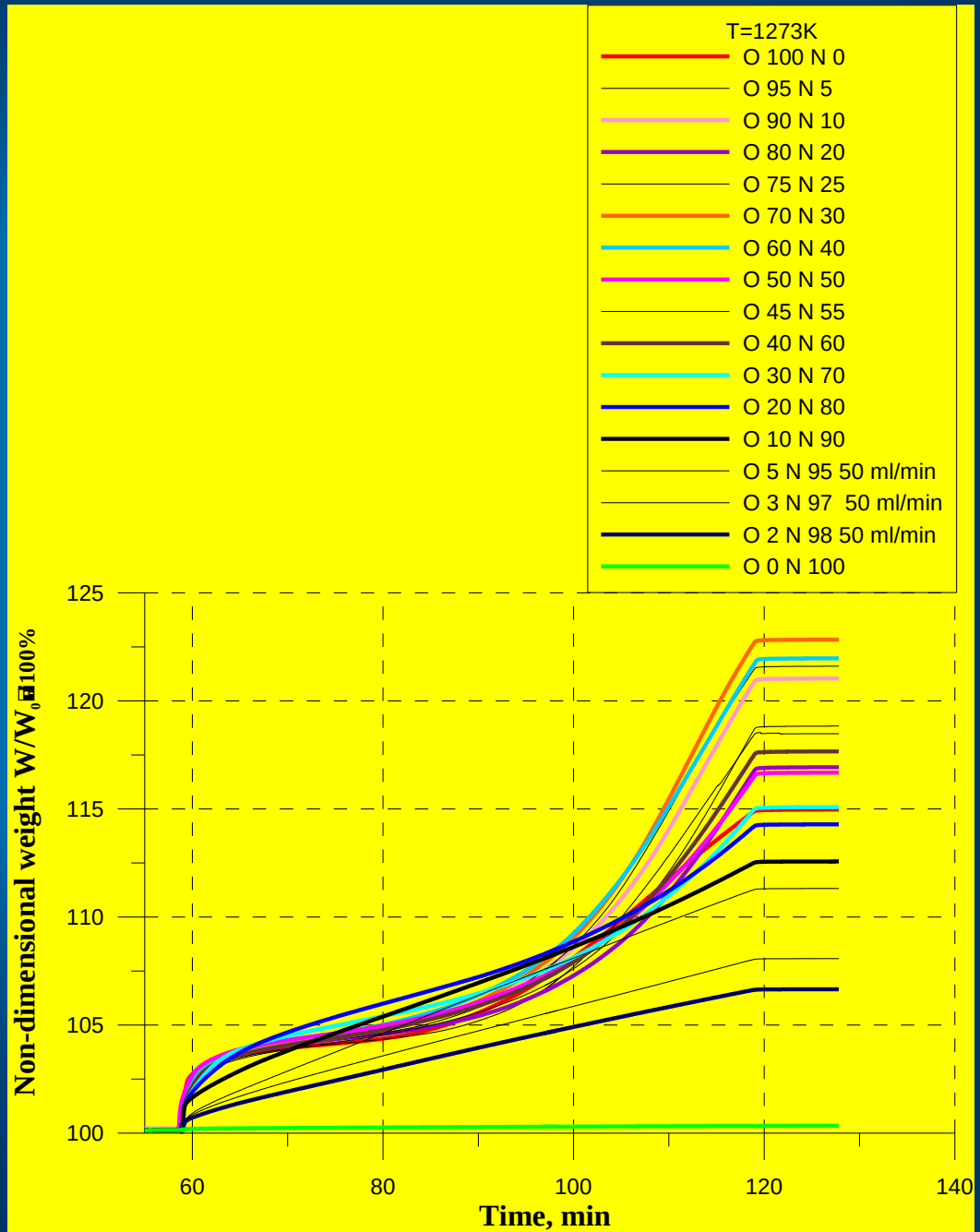
Surface Renewal Model

$$\delta^* = K_d \sqrt{t^*}$$



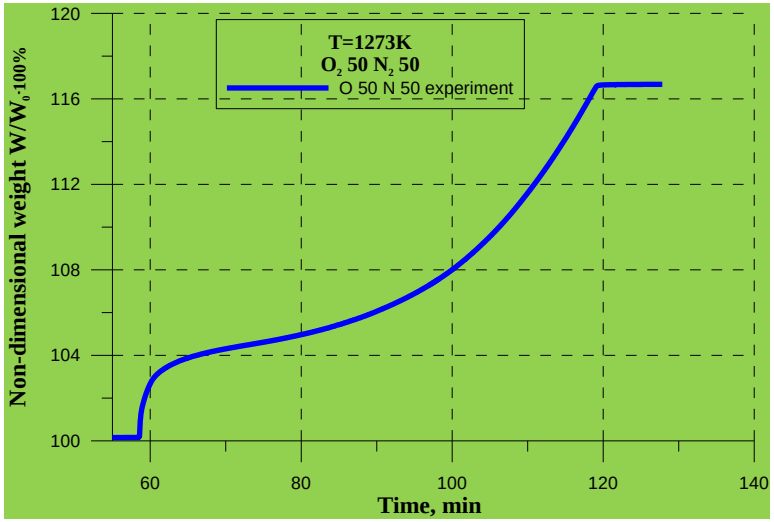
$$D_{d,eff} = K_{diff} D_d$$

Experimental Weight Gain T=600°C



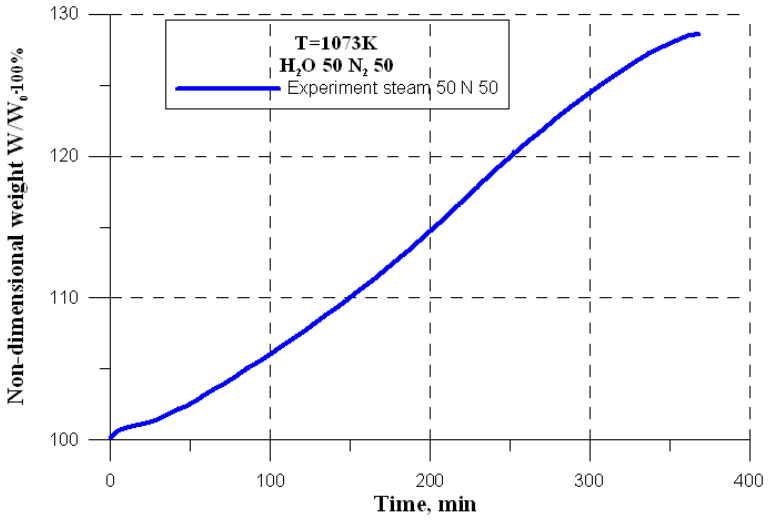
T=1000°C

	δ0	DELCR	Kdiff	Jump (%)
N0	5.30E-05	1.90E-05	1	1.5
N5	5.60E-05	1.20E-05	1	1.2
N10	5.66E-05	9.80E-06	1	1.7
N20	5.85E-05	1.20E-05	1	1.5
N25	5.67E-05	9.00E-06	1	1.4
N30	5.60E-05	9.00E-06	1	1.5
N40	5.60E-05	9.20E-06	1	1.6
N50	5.77E-05	1.30E-05	1	1.7
N55	5.85E-05	1.02E-05	1	1.5
N60	5.80E-05	1.13E-05	1	1.7
N70	5.80E-05	1.40E-05	1	1.7
N80	8.10E-05	3.87E-05	1.7	1.1
N90	4.50E-05	3.20E-05	1	1
N95	3.10E-05	3.70E-05	1	

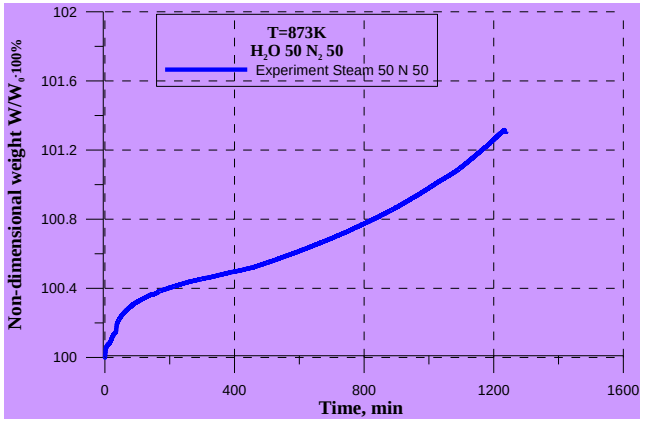


T=800°C

	δ0	δCR	Kdiff	Jump (%)
N5	2.37E-05	9.60E-06	1	0
N10	1.62E-05	6.80E-06	1	0
N20	1.70E-05	6.50E-06	1	0
N30	1.44E-05	8.30E-06	1	0
N40	2.13E-05	7.00E-06	1	0.2
N50	2.08E-05	6.50E-06	1	0.2
N60	2.05E-05	6.30E-06	1	0.2
N70	2.15E-05	6.00E-06	1	0.2
N80	2.17E-05	6.30E-06	1	0.2
N90	2.42E-05	6.10E-06	1	0.2
N95	2.05E-05	7.10E-06	1	0.1
N97	2.07E-05	8.50E-06	1	0.1
N98	2.05E-05	1.02E-05	1	0
N99	2.10E-05	1.45E-05	1	0.1



T=600°C



	δ_0	δcr	Kdiff	Jump, %
N0.5	8.00E-06	4.80E-06	2.5	0.15
N0.7	6.00E-06	3.70E-06	1.5	0.15
N1	5.50E-06	2.70E-06	1.5	0.15
N2	8.00E-06	5.00E-06	2.5	0.15
N5	8.00E-06	5.80E-06	2.5	0.15

Summary of Low Temperature Oxidation. Influence of Temperature on δ_0 and δ_{cr} – Key Parameters of Model O 50% N 50%

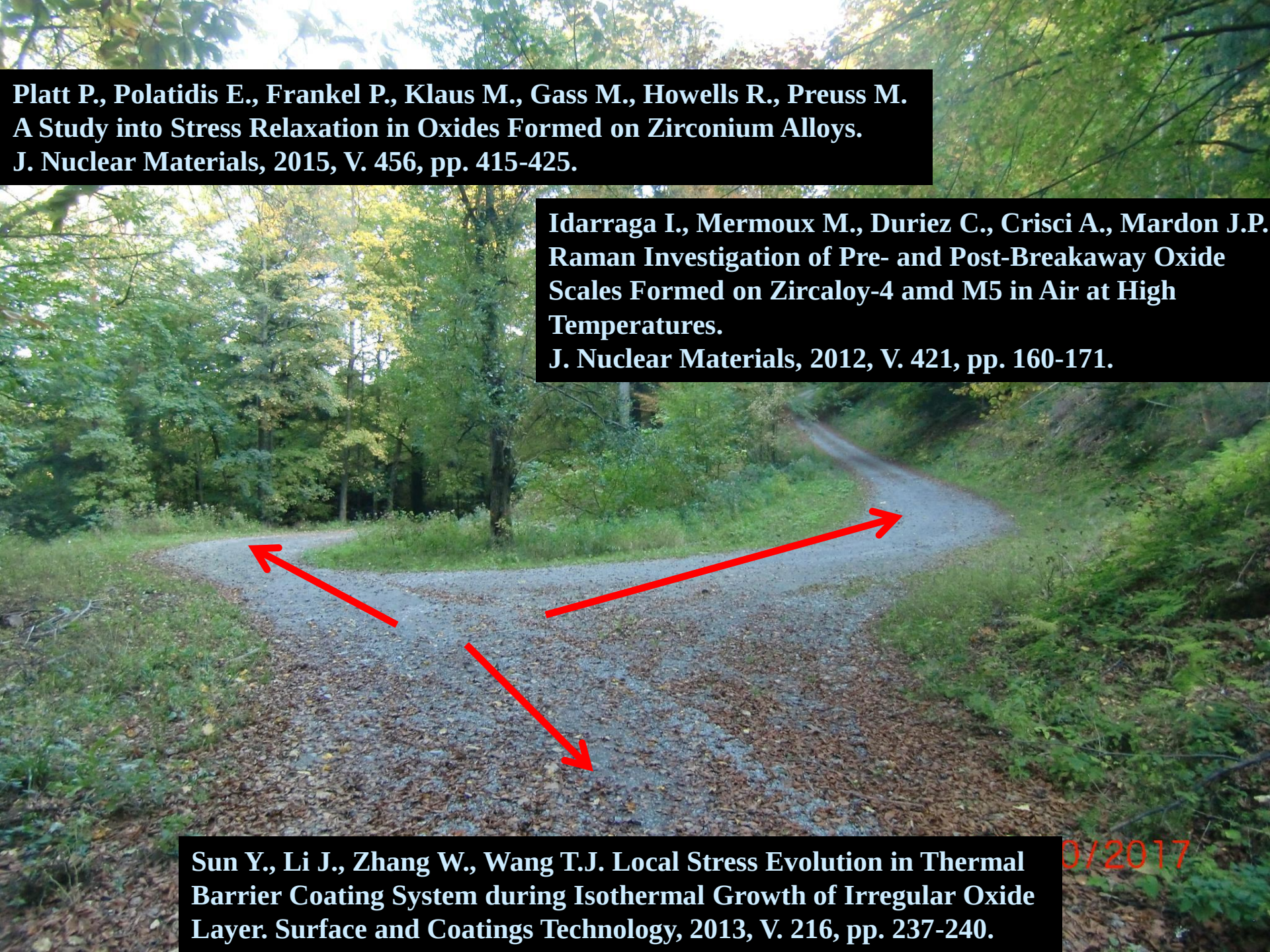
T, °C	δ_0 [mkm]	δ_{cr} [mkm]
600	7÷9	3÷4
800	15÷20	6÷8
1000	30÷50	11÷20

Sanggil Park. Overview of the Air Oxidation Kinetic Modeling. QUENCH-20, November 11-13, 2014, Karlsruhe, Germany.

$-8498.81+5.79\cdot T-4.59\cdot 10^{-21}\cdot T^{7}+A/\delta_{cr}{}^{B}=0$ - criterium of ZrO_2 film degradation

$A \cong 46.5$ from experimental data

$B=1/3$



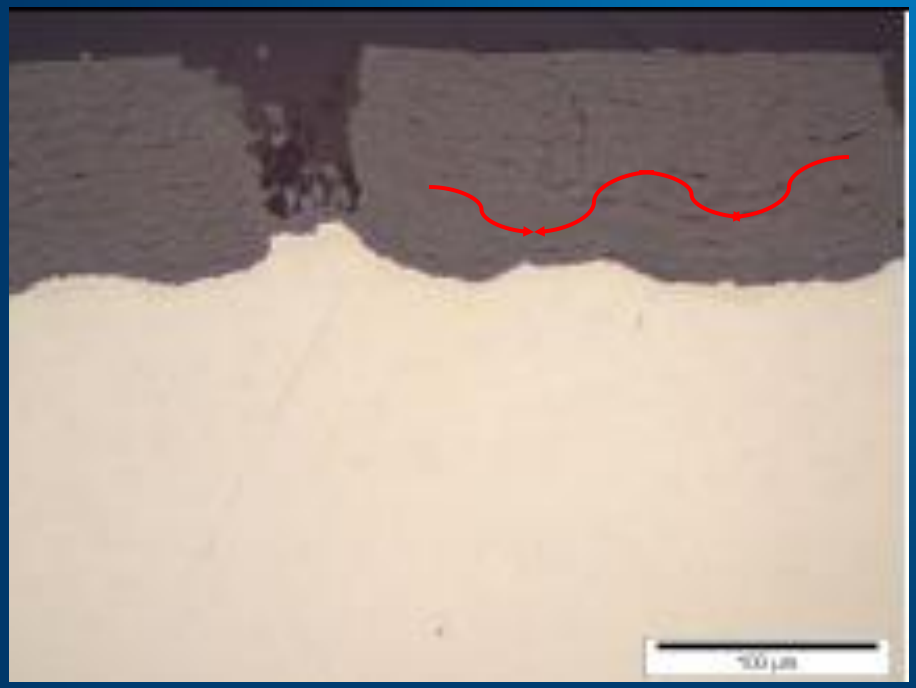
Platt P., Polatidis E., Frankel P., Klaus M., Gass M., Howells R., Preuss M. A Study into Stress Relaxation in Oxides Formed on Zirconium Alloys. J. Nuclear Materials, 2015, V. 456, pp. 415-425.

Idarraga I., Mermoux M., Duriez C., Crisci A., Mardon J.P. Raman Investigation of Pre- and Post-Breakaway Oxide Scales Formed on Zircaloy-4 amd M5 in Air at High Temperatures. J. Nuclear Materials, 2012, V. 421, pp. 160-171.

Sun Y., Li J., Zhang W., Wang T.J. Local Stress Evolution in Thermal Barrier Coating System during Isothermal Growth of Irregular Oxide Layer. Surface and Coatings Technology, 2013, V. 216, pp. 237-240.

Undulated Surface

800°C, 30%O_2, 70%N_2



Characteristic wavelength is less for thin critical oxide thickness

Comparison with Separate Effects Tests. T=600°C

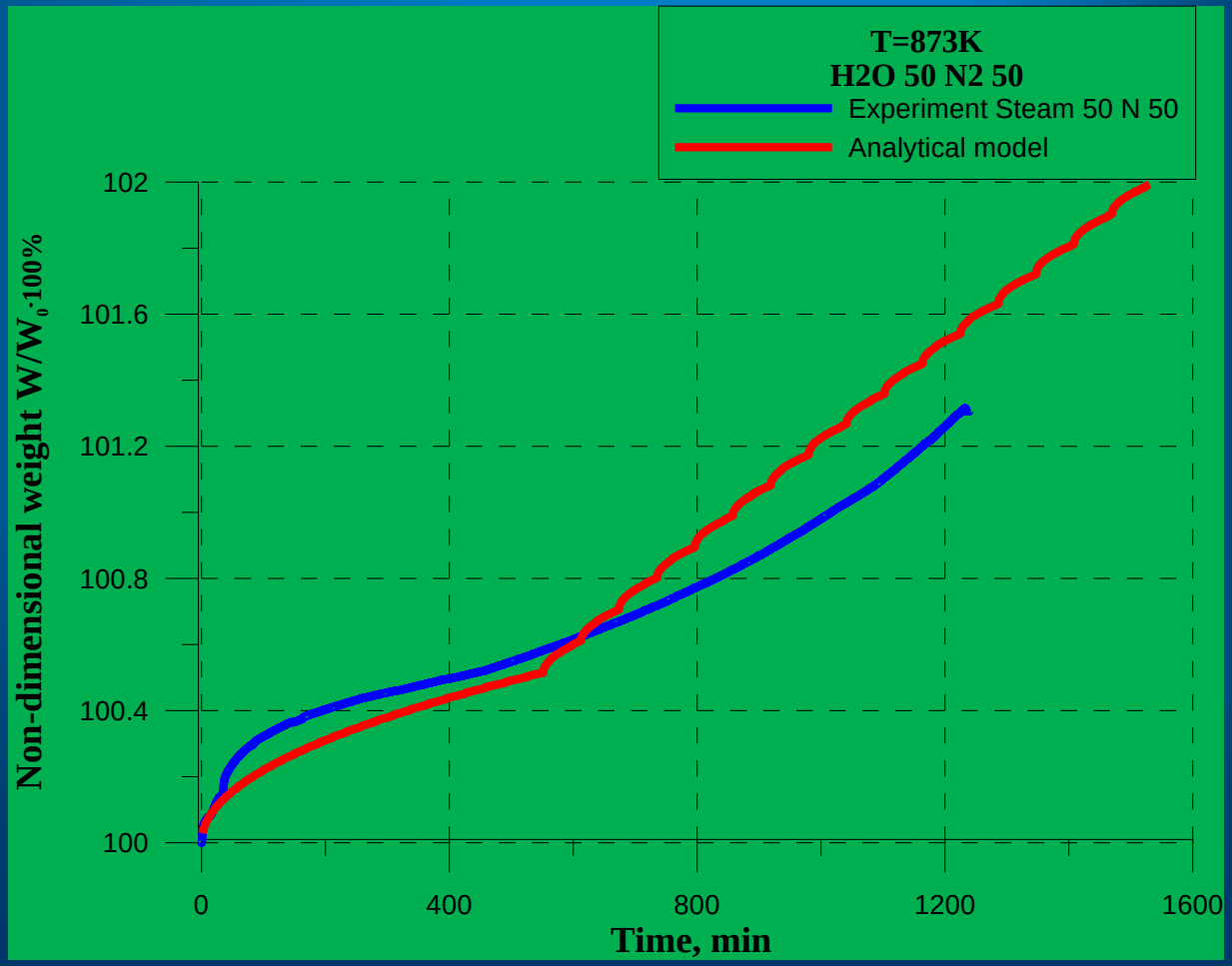
$D_d = 5.1 \cdot 10^{-14}$ m^2/s

$D_a = 1.7 \cdot 10^{-15}$ m^2/s

$K_d = 0.039 \cdot (4D_d)^{1/2}$

$K_d = 1.2 \cdot 10^{-8}$ m/s$^{1/2}$

V≅0.0091 μm/min



Comparison with Separate Effects Tests. T=800°C

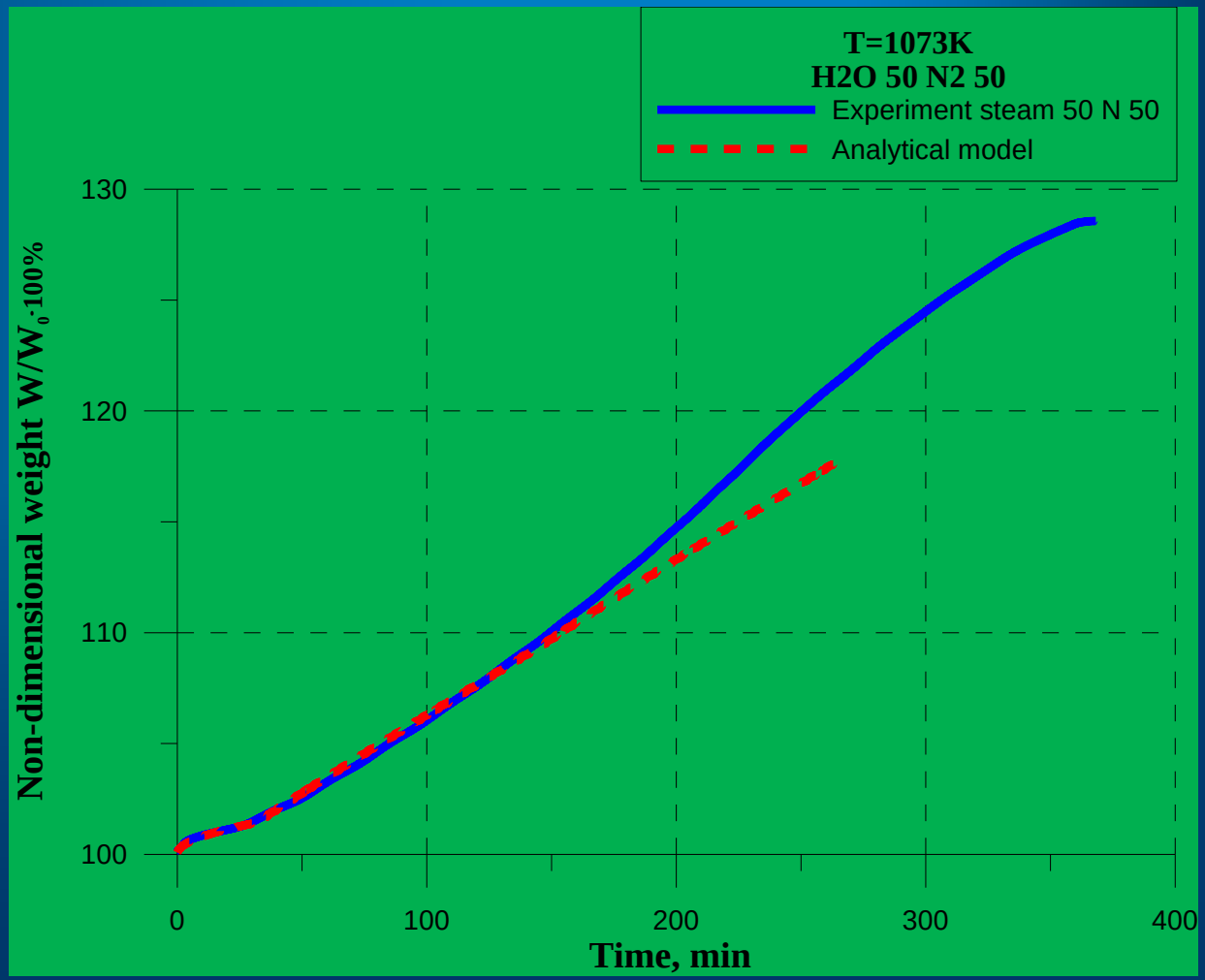
$D_d = 4.4 \cdot 10^{-12}$ m^2/s

$D_a = 1.1 \cdot 10^{-13}$ m^2/s

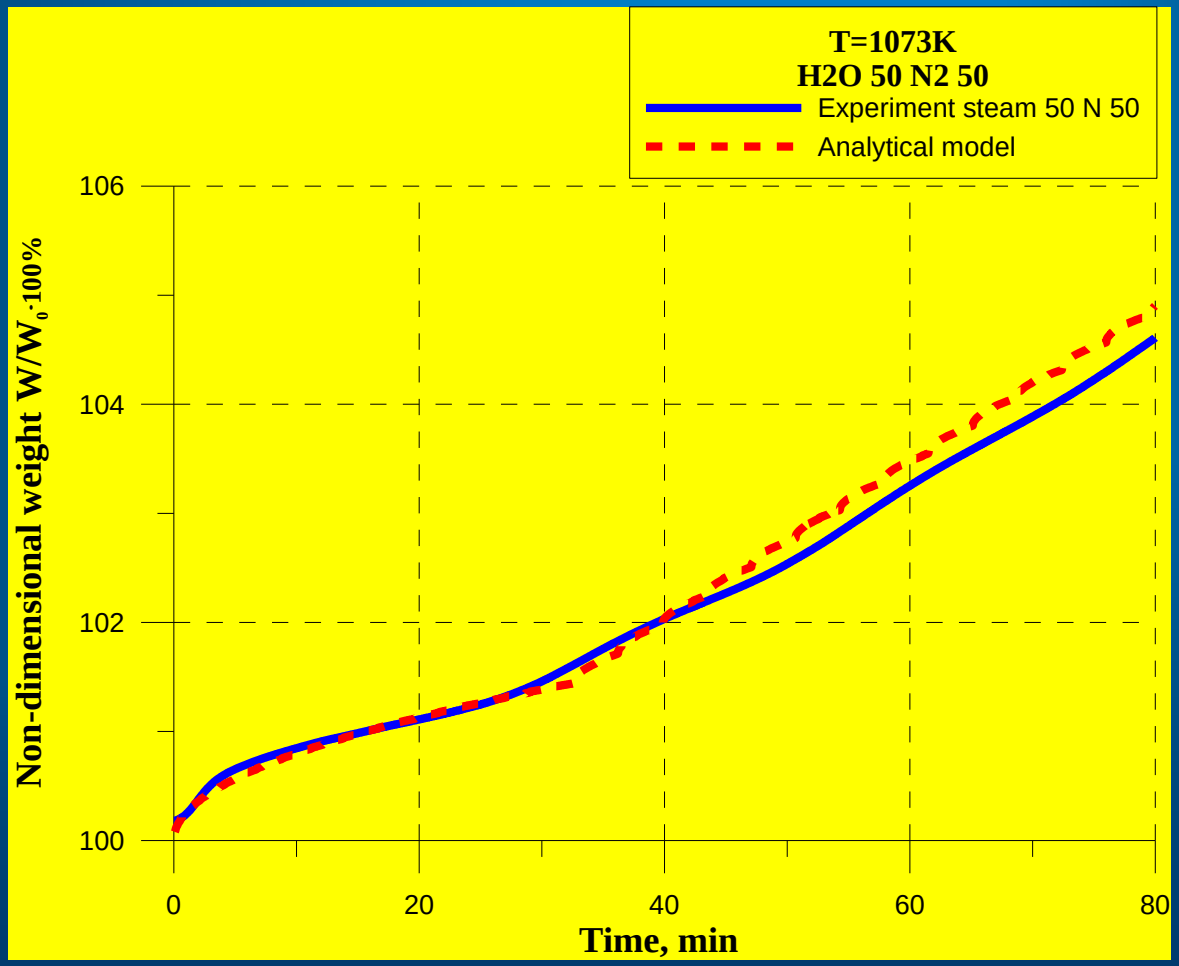
$K_d = 0.027 \cdot (4D_d)^{1/2}$

$K_d = 4.9 \cdot 10^{-8}$ m/s$^{1/2}$

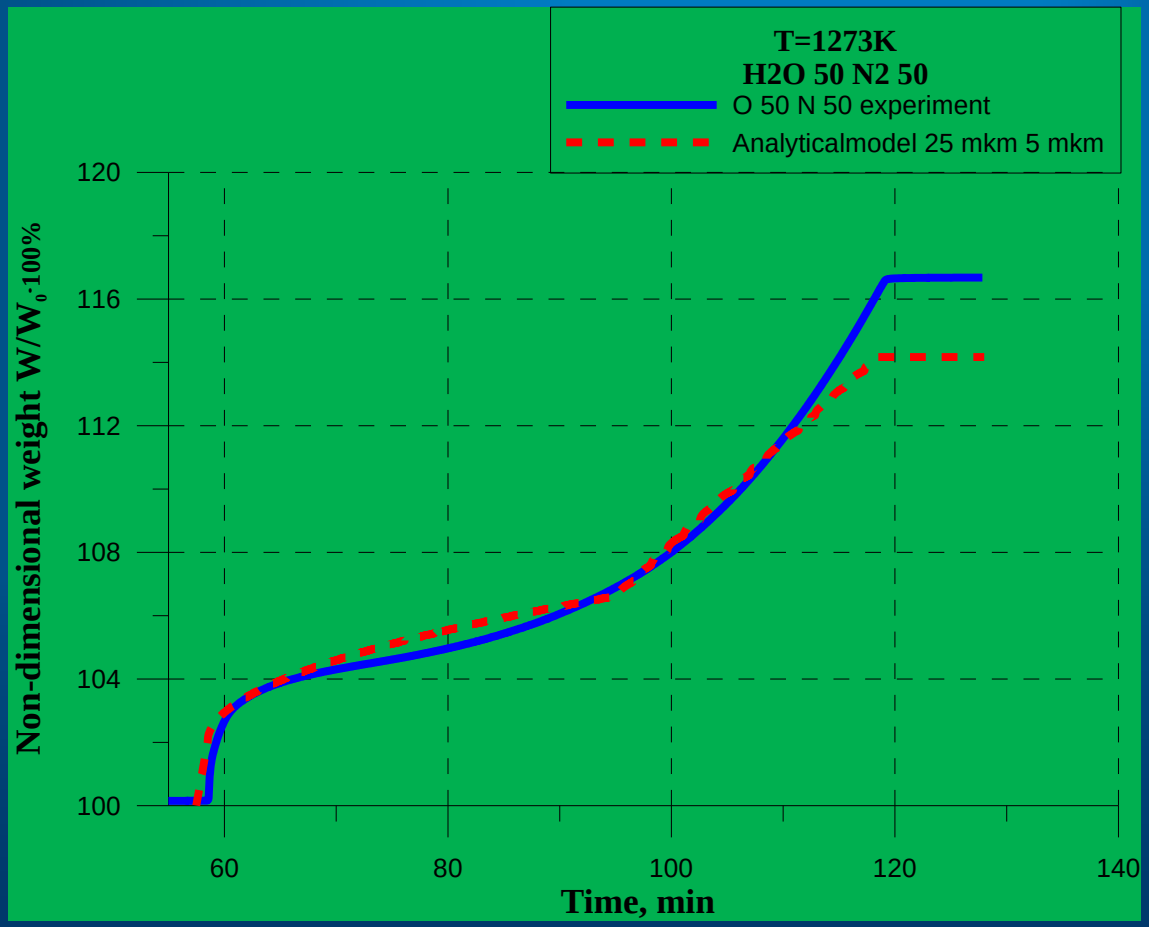
V≅0.45 μm/min



Comparison with Separate Effects Tests. T=800°C Big Scale



Comparison with Separate Effects Tests. T=1000°C

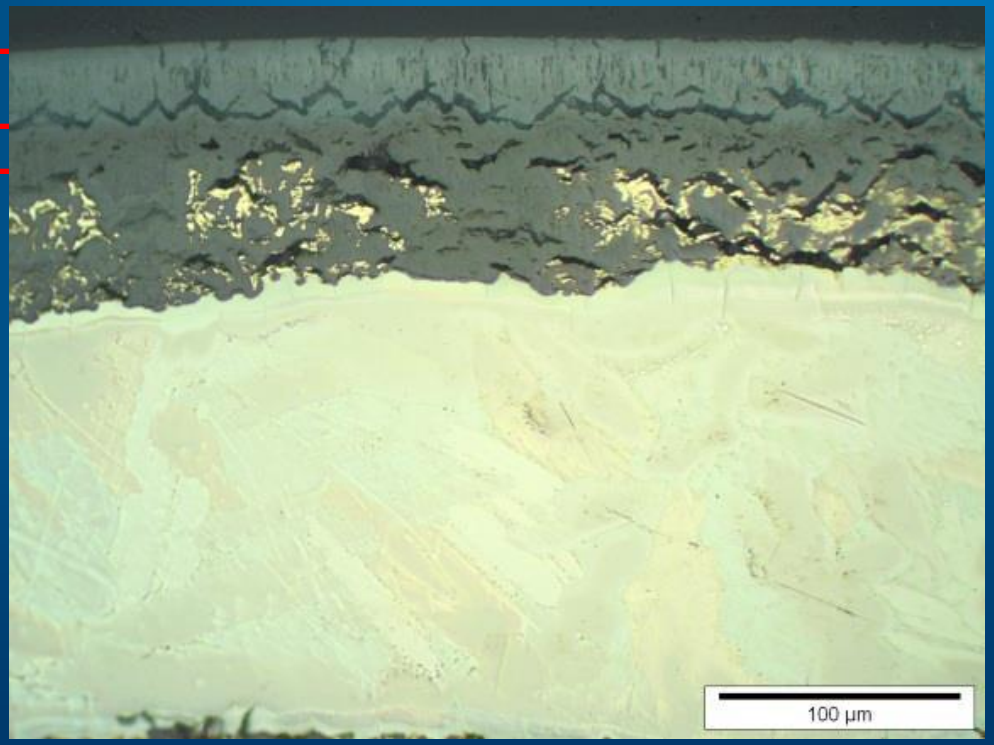


Parameter δ_0 and δ_{cr}

1000°C, 70%O_2, 30%N_2

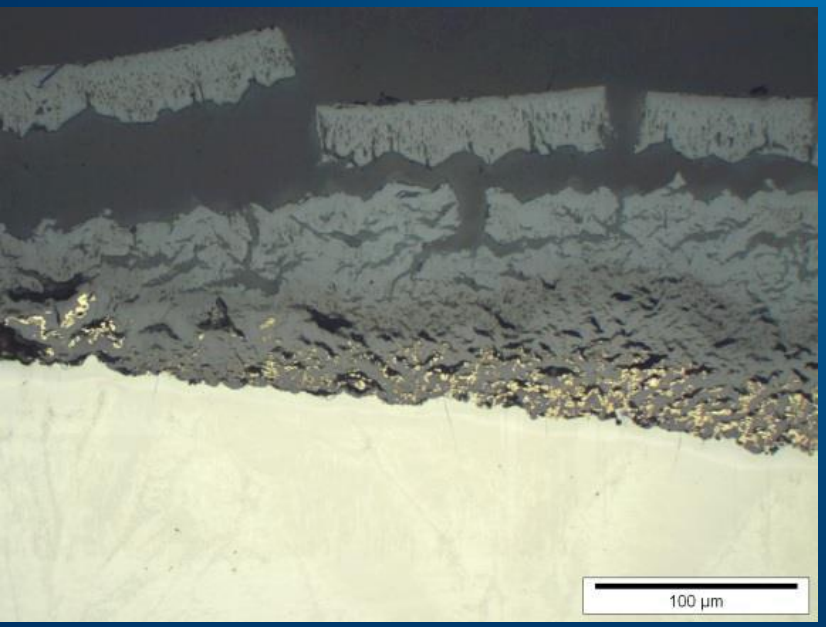
δ_0=30 mkm

δ_{cr}=12 mkm



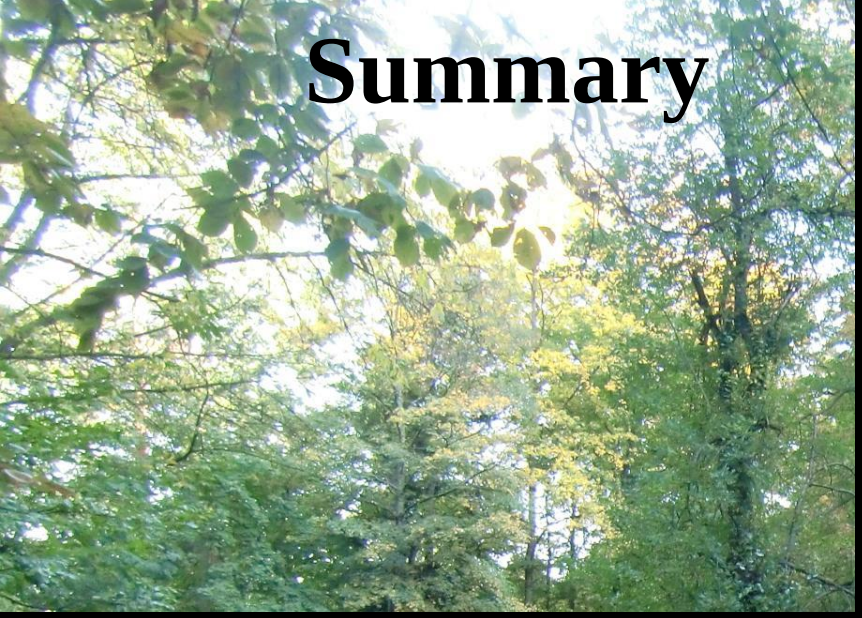
Breakaway in Different Scales

1000°C, 30%O_2, 70%N_2



0°C, 21%O_2, 79%N_2





Summary

The analytical model to calculate Zr-based claddings breakaway oxidation in oxygen-nitrogen and steam-nitrogen mixtures is developed for the temperature range T=673÷1323K.

The role of different compressive and tensile stresses in stress evolution is identified.

At those temperatures the oxidation kinetics is determined by oxide/metal interface renewal (breakaway) mechanism.

The model includes the following:

1. The enhancement of effective oxygen diffusion coefficient (by factor K_{diff}) in ZrO_2+ZrN layer;

2. The quantity K_{diff} (basically, of the order of several times) is a value depending on nitrogen content in a mixture;

3. Treating the mechanism of breakaway by a coupled solution of thermo-mechanical problem , the details of this complicated behaviour were discussed in the presentations;

4. The model developed is able to get important practical consequences;

The predictions of the model are compared with recent SETs. Basic experimental dependencies of key variables are adequately calculated using parameters of the model.

Validation of the SOCRAT code on the integral experiment CORA-15

A.E. Kiselev, T.A. Yudina, D.Yu. Tomashchik, N.I. Ryzhov, A.E. Tarasov, R.V. Chalyy

Nuclear Safety Institute (IBRAE RAN), Moscow, Russia

ABSTRACT

Modeling of the CORA-15 test was perfomed, in which the behavior of the PWR type fuel assembly under the LOCA conditions was investigated. An important feature of the experiments was a high overpressure of fuel rods (prototypical for reactor conditions). As a result of all rods underwent ballooning and rupture. The new results of modeling the CORA-15 test are presented in the paper. It is shown that the SOCRAT code adequately reproduces the history of fuel element temperatures, ballooning and rupture of claddings, oxidation of claddings, dissolution of UO_2 by the zircaloy cladding, the melt formation and relocation, the blockage formation.


parameters,
hydrogen
blockage

РОССИЙСКАЯ АКАДЕМИЯ НАУК
Институт проблем безопасного развития атомной энергетики
RUSSIAN ACADEMY OF SCIENCES
Nuclear Safety Institute
ИБРАЭ

Validation of the SOCRAT code on the integral experiment CORA-15

A.E. Tarasov, A.E. Kiselev, T.A. Yudina,
D.Yu. Tomashchik, N.I. Ryzhov, R.V. Chalyy

23d Quench Workshop, KIT, Karlsruhe, 17-19 October 2017

Motivation

- CORA-15 test was offered to participants of the on-going IAEA FUMAC CRP to check codes capability to model cladding ballooning and burst parameters of fuel rods in bundle
- The test features: 25 fuel rod bundle, pressurized fuel rods (6.0 MPa), UO_2 as a pellet material, 2 Ag/In/Cd - steel absorber rods
- The aim of CORA-15 modeling by integral SOCRAT code:
  - ✓ Investigation of the ballooning effect on the bundle degradation;
  - ✓ Validation of CROX model to predict cladding ballooning and burst parameters, PROF and LIQF models to predict Zr oxidation and hydrogen release, DROG model to predict melt relocation and blockage formation, and their interaction

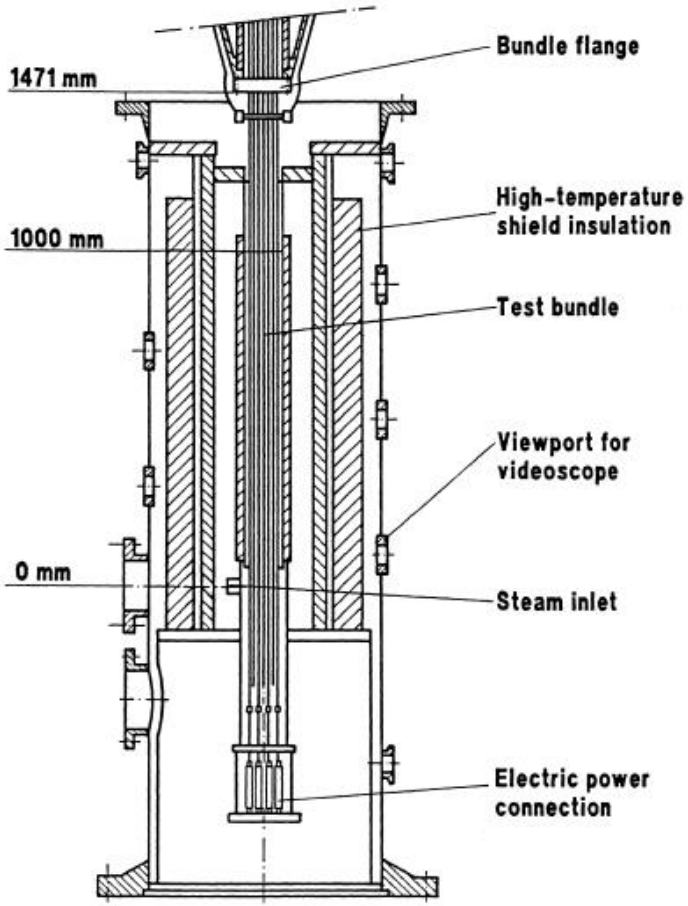
SOCRAT code – main modules

SVECHA	– core degradation (CROX, PROF, LIQF, DROG models)
RATEG	– primary and secondary thermal hydraulics
RADIATION	– radiation heat exchange between elements
HEFEST	– thermal physics of corium and thermal mechanics of RPV
ANGAR, **KUPOL**	– thermal hydraulics in containment
TOCHKA	– point neutron kinetics
BONUS	– FP accumulation in fuel
RELEASE	– FP release from fuel in gas gap
MFPR_MELT	– FP release from the corium
GAPREL	– FP release from the gas gap
PROFIT	– FP behavior in the primary circuit
CONTFP	– FP behavior in containment volumes
RACHIM	– activity and heat generation by FP
VAPEX-M	– fuel-coolant interaction

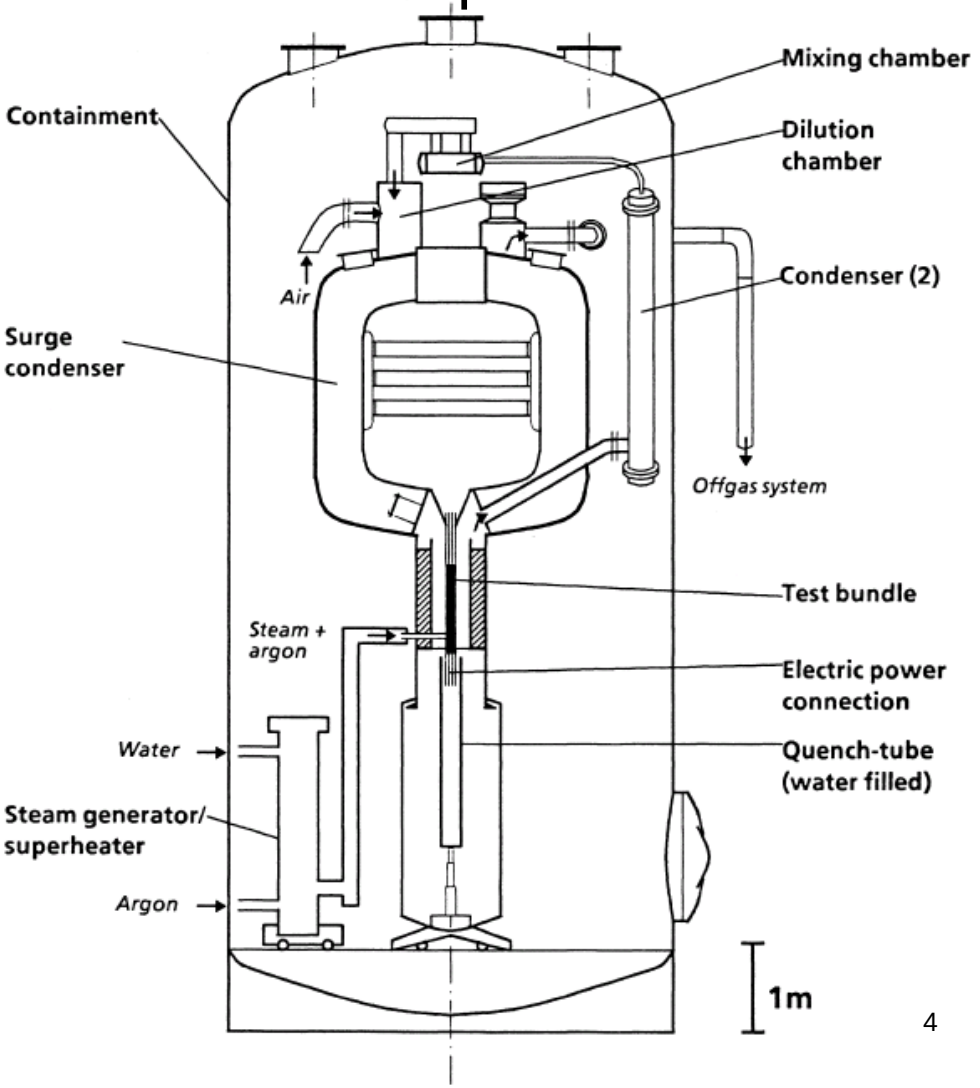
+ **material properties database**

CORA facility

CORA test section



Severe Fuel Damage (SFD) test facility CORA, main components

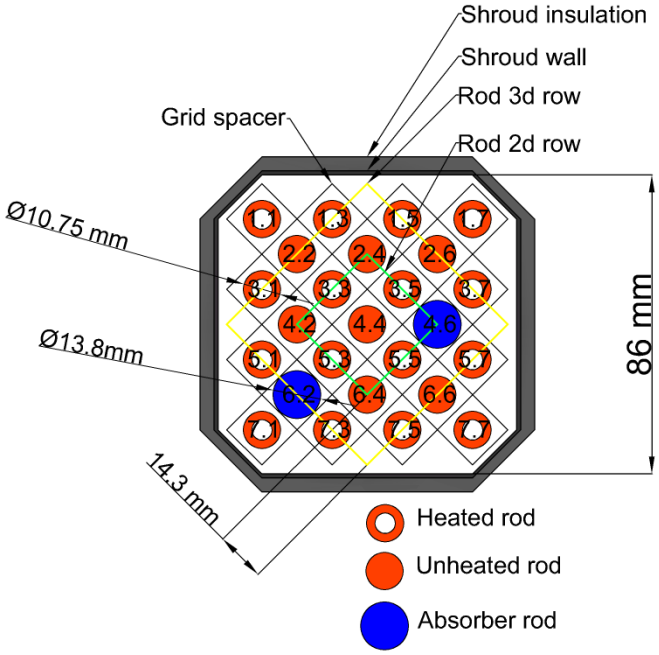


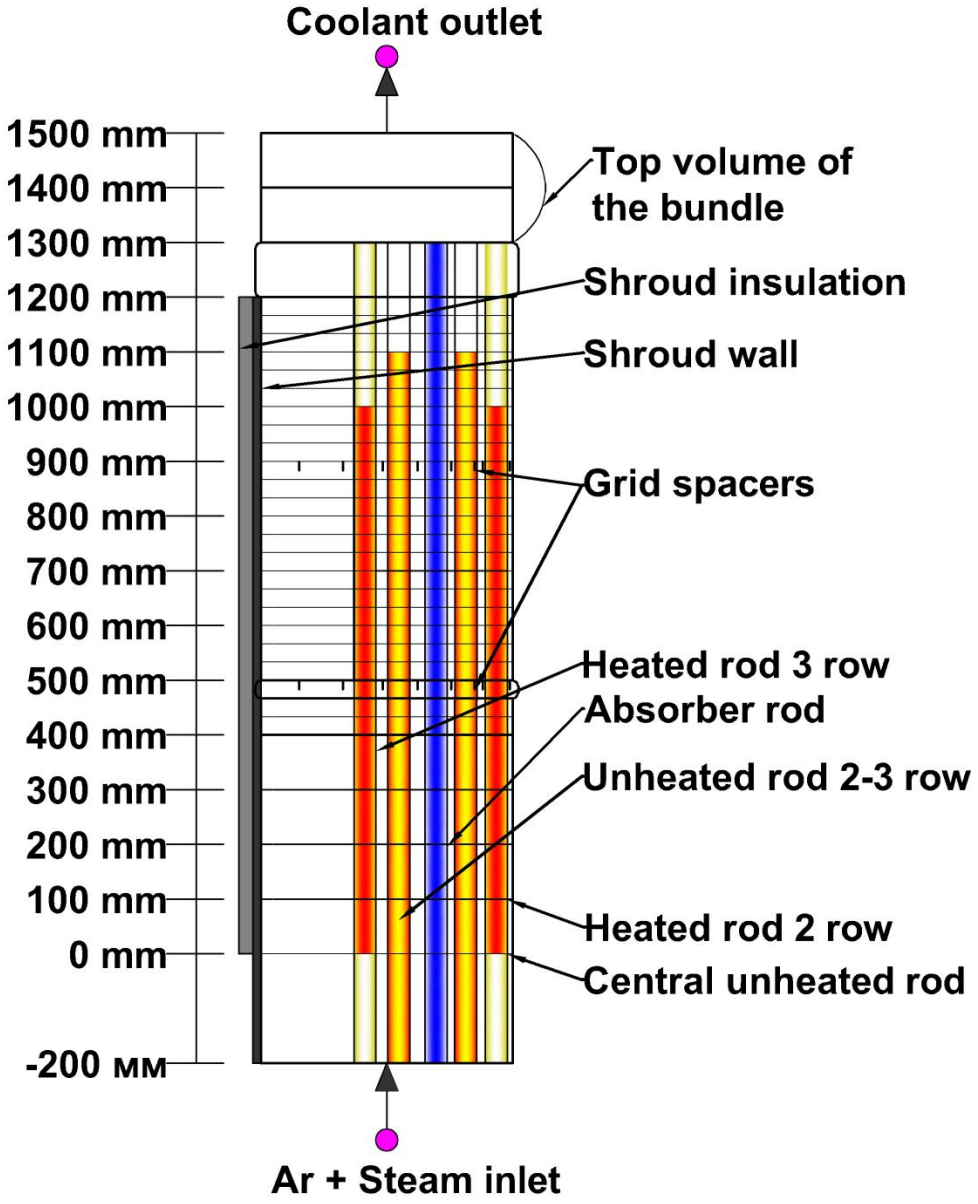
Nodalization scheme of CORA-15 bundle

– Modeled structure:

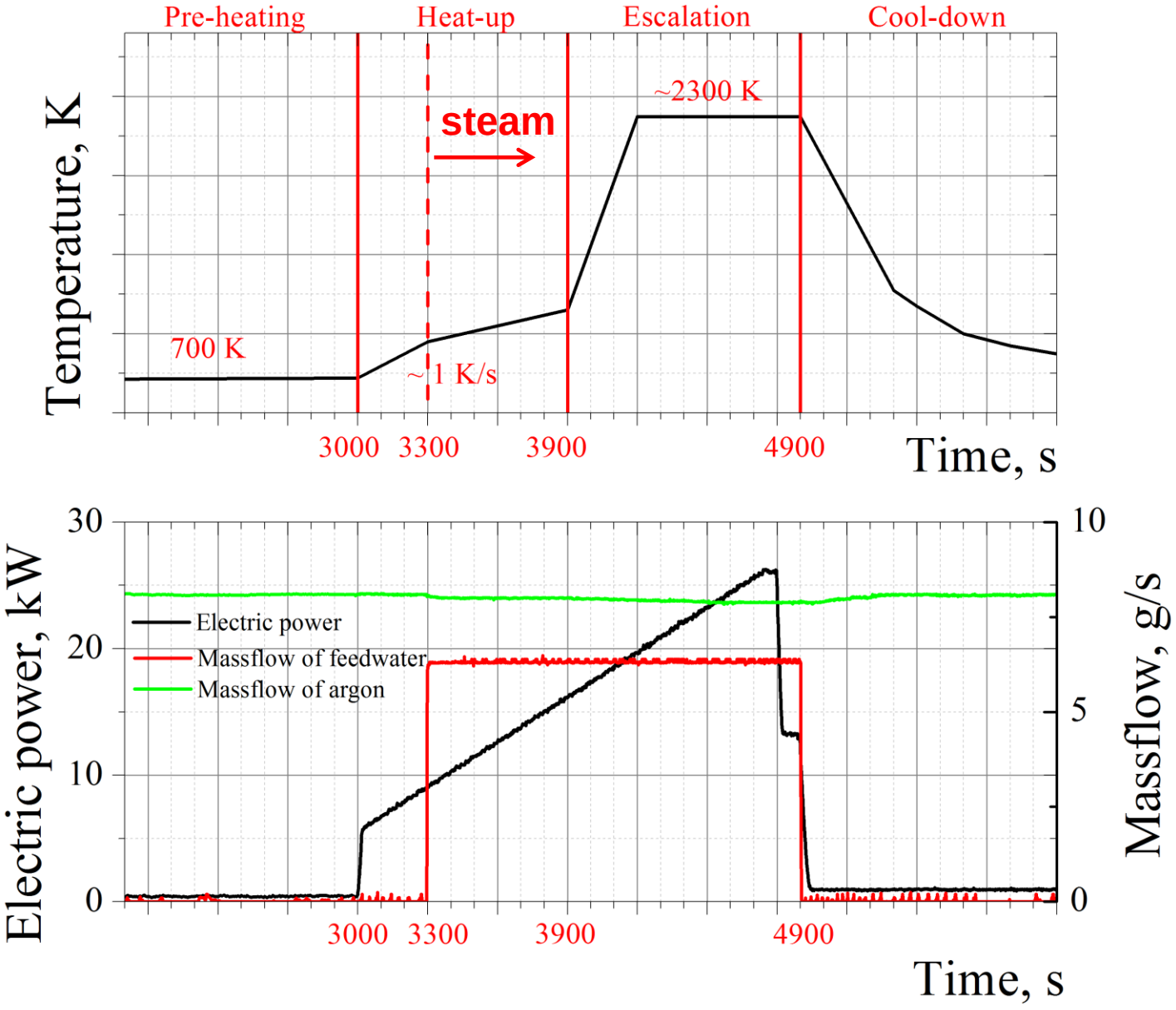
- central rod
- 6 unheated rods in inner (2–3)row
- 4 heated rods in inner (2) row
- 2 absorber rods in inner (2) row
- 12 heated rods in outer (3) row
- shroud
- two spacer grids

–Two hydraulic channels



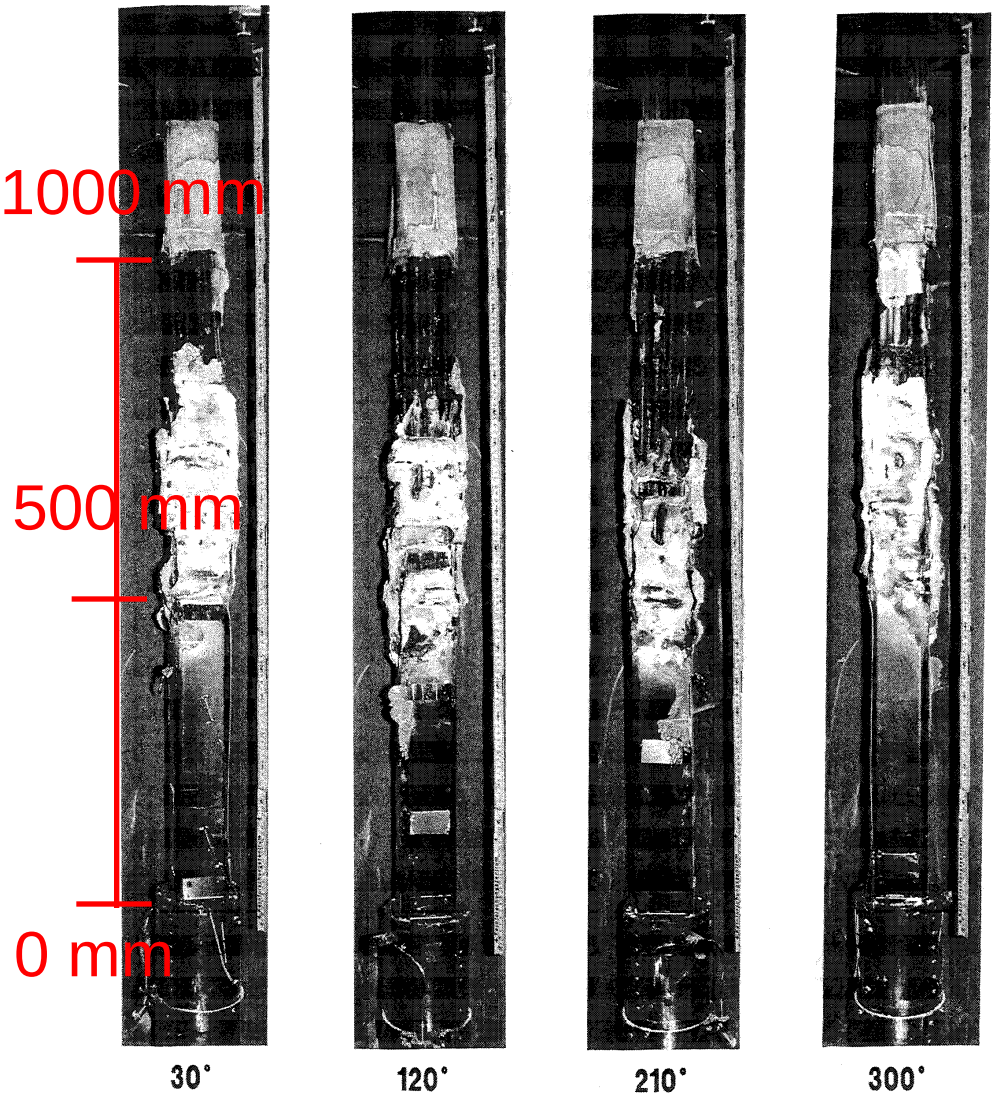


Experiment scenario and boundary conditions



Main results

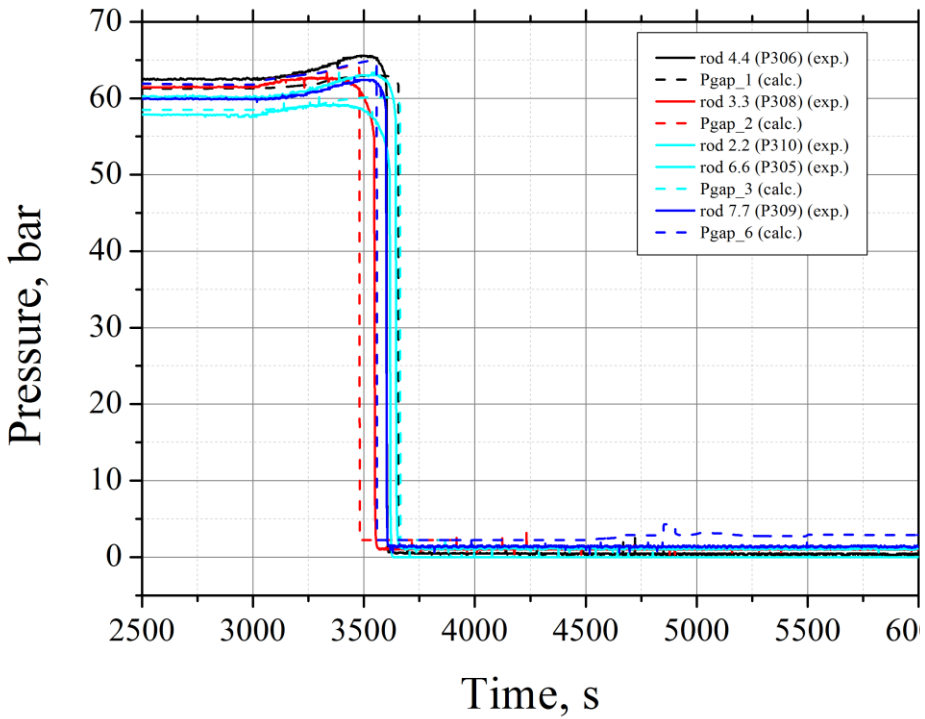
Post-test view

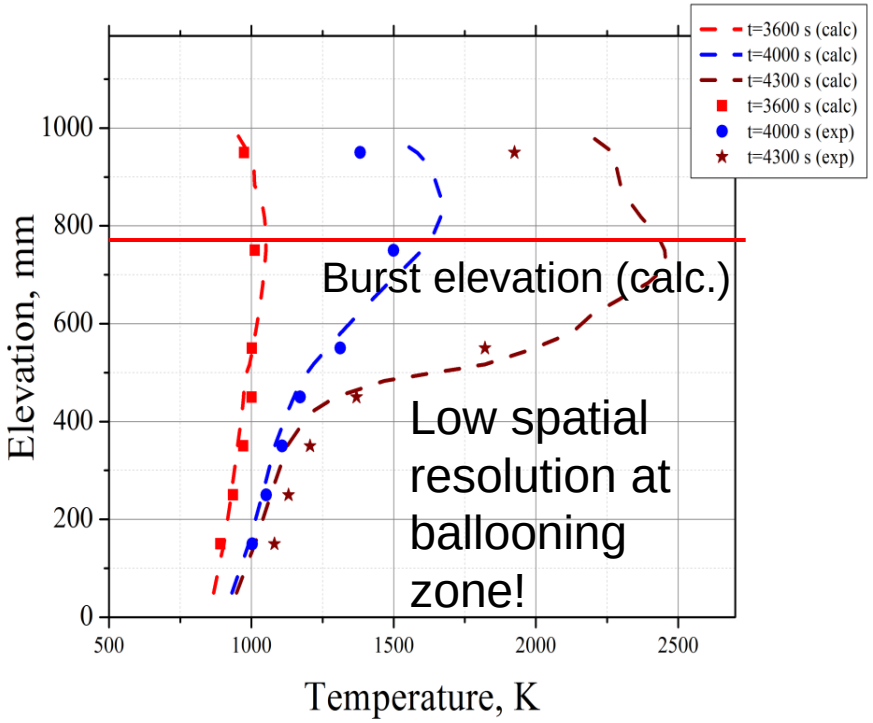


Calculation

- rupture of all claddings as in the test
- severe bundle degradation at the upper part of the bundle and intact bundle in the lower part of the assembly as in the test
- a massive blockage formation at an elevation of 500 mm as in the test

Internal pressure in rods

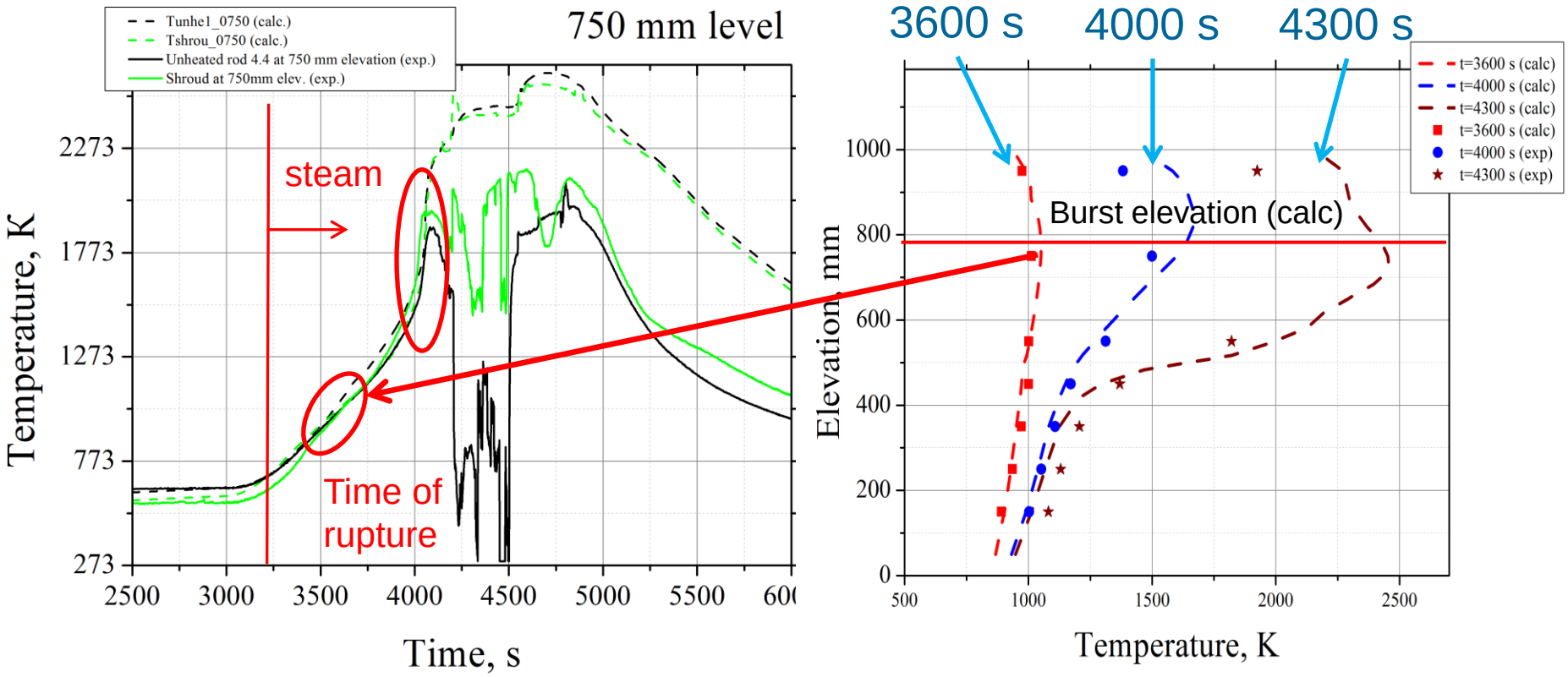




Good agreement on cladding rupture:

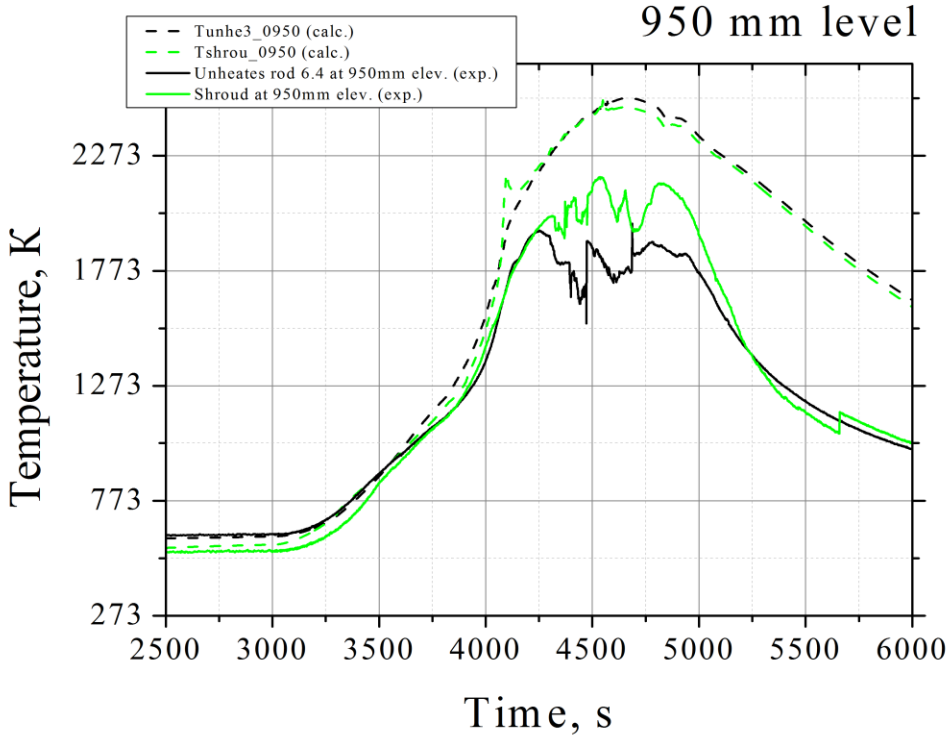
- time:
  - 3493 s – 3645 s (measured)
  - 3480 s – 3663 s (predicted)
- elevation:
  - 800 mm (the hottest zone at the time of absorber rod failure as reported in FZKA 7448)
  - 766-800 mm (predicted)
- temperature :
  - > 923 – 1023 K (TC at 750 mm elevation)
  - 1088 - 1129 K (predicted)

Cladding and shroud temperatures at 750 mm



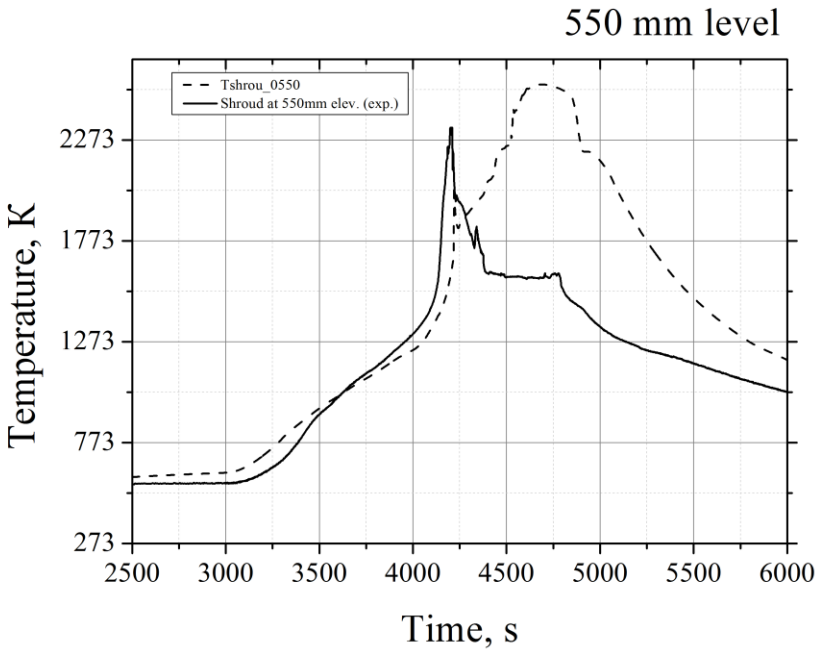
- Predicted burst elevation – 766-800 mm, but the nearest TC location - at 750 mm
- A good agreement on temperature behavior at pre-heating, heat-up and escalation phases at 750 mm, tendency to overestimation at transient phase
- Temperature runaway at 4050 s is due to melt relocation from the upper hottest elevation (850 mm)

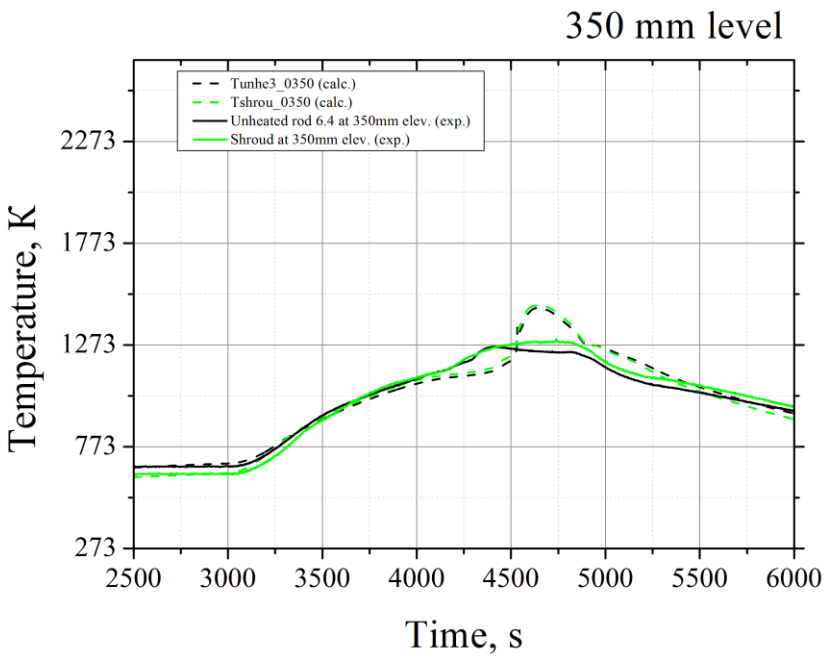
Cladding and shroud temperatures above the hottest zone

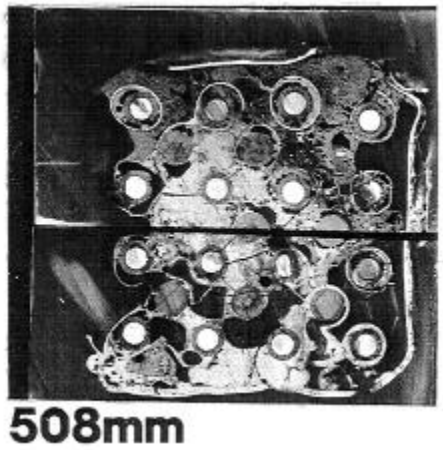


- Temperatures at transient phase are overestimated above the hottest zone (based on TC at 950 mm), but temperature trend is reproduced well – temperature escalation starts at around 4000 s;
- Calculated temperature reaches the melting point; melt formation, its oxidation and relocation downwards occurs

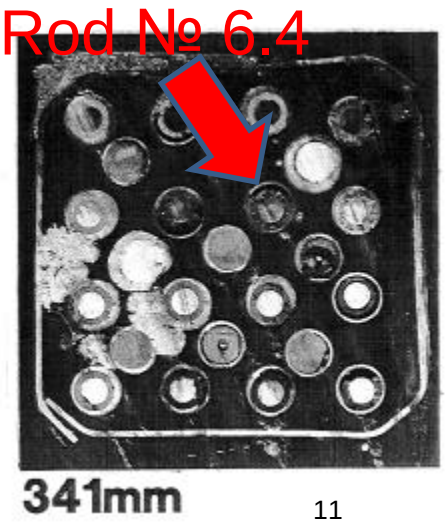
Cladding and shroud temperatures at blockage zone and below



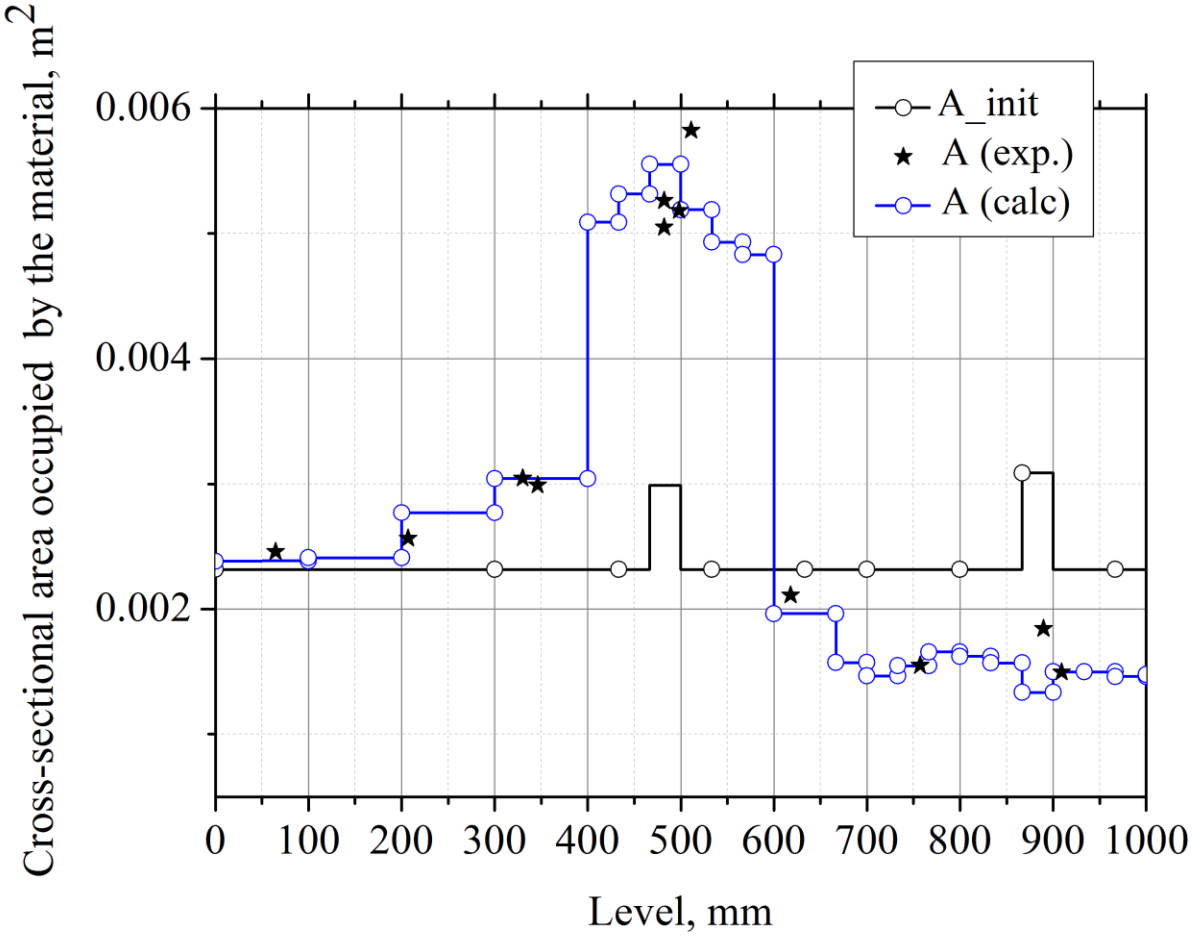




- Predicted temperature escalation at 550 mm and heating up at 350 mm due to melt relocation are in a good agreement with posttest investigations



Blockage profile

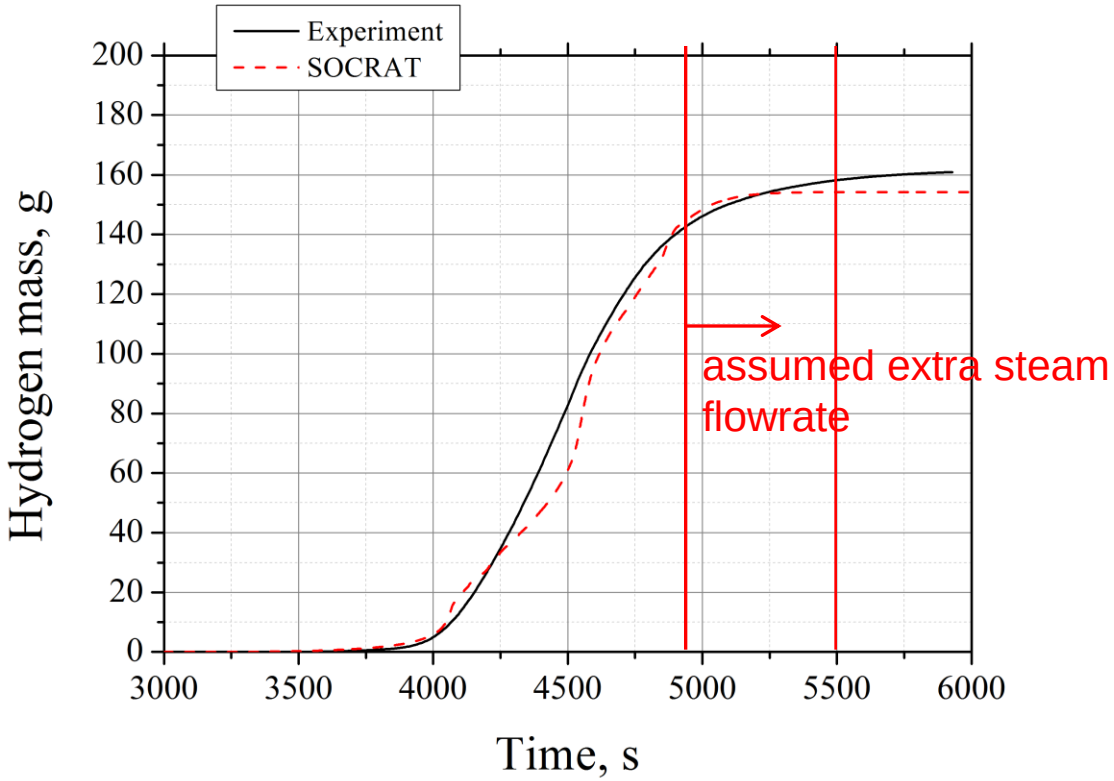


- Good agreement on blockage profile
- Elevation of maximum blockage:

✓ 512 mm – measured

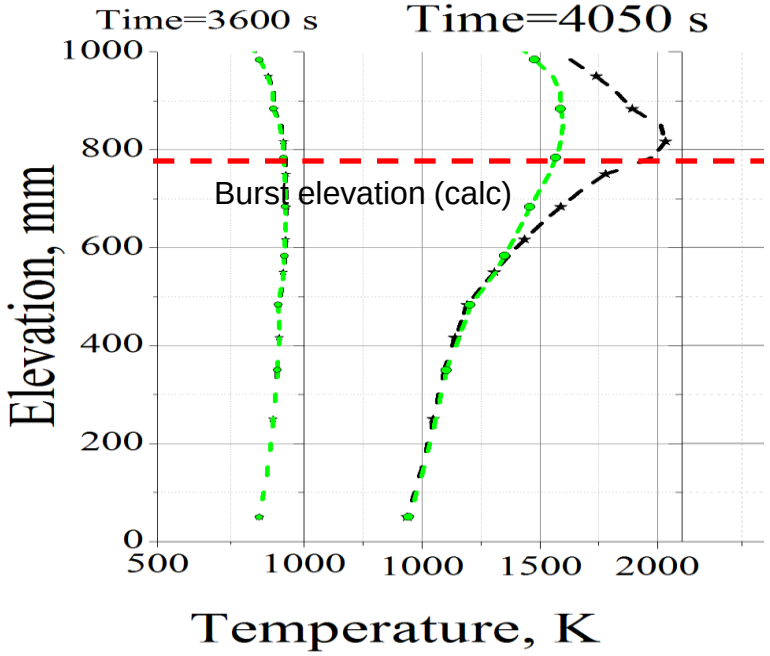
✓ 466-500 mm – predicted

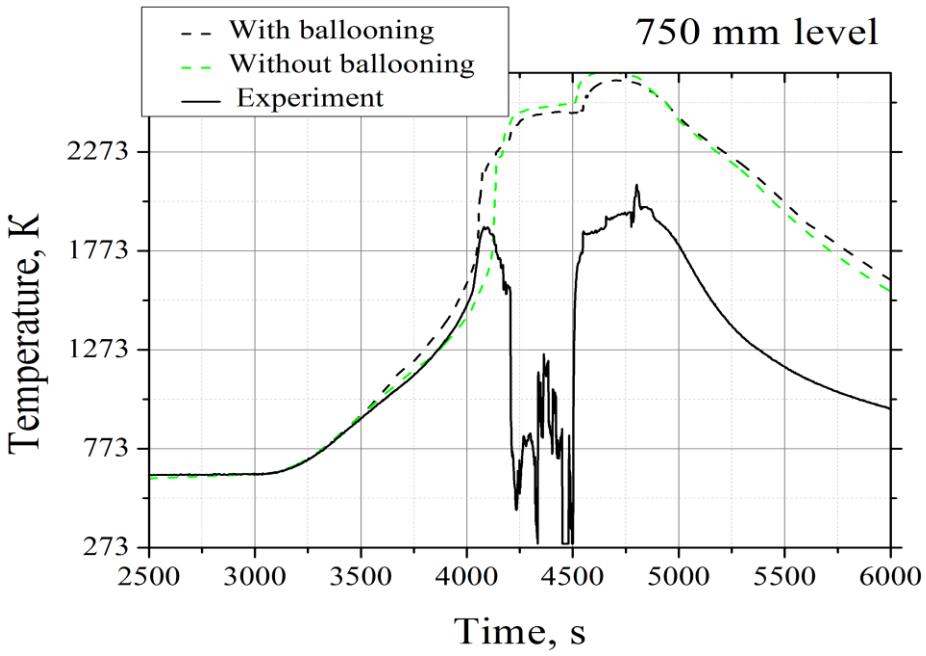
Hydrogen release



- Calculated hydrogen production matches data well while feedwater is injected into steam generator (i.e. till 4900 s)
- The measured extra hydrogen generation for 1000 s after feedwater switching off (4900 s) could be explained by continuation of steam supply with smaller flowrate until the remaining water in the steam generator evaporates. Extra steam flowrate is simulated with linear decreasing from 0.9 g/s to 0 g for 600 seconds

Ballooning influence





- ✓ Higher heat up rate at heat-up phase and earlier onset of temperature escalation (at t=4050 s vs t=4150 s without ballooning)

Ballooning of claddings → increase of gap width → local heat exchange reduction → increase of local fuel and heater temperatures → higher local electrical resistance → higher local Joule heat power → higher local heat up rate → earlier start of temperature escalation

Conclusions

- SOCRAT code adequately reproduces the cladding temperatures evolutions, its ballooning and rupture, hydrogen production, the blockage formation due to melt relocation and solidification (within the uncertainties of the measured data)
- Ballooning effect was found based on the comparison of the calculated results with and without ballooning. It could be referred to the features of the facility (heating up by Joule heat power, small scale of the bundle in radial direction)

Acknowledgements

- The participation in 23d Quench Workshop was supported financially by IAEA
- The numerical modeling was performed in the framework of IAEA FUMAC CRP.

QUENCH-06 Experiment Post-test Calculations and Integrated Uncertainty Analysis with RELAP/SCDAPSIM/MOD3.4 and MOD3.5

Chris Allison and Marina Perez – Innovative Systems Software
Olga Dutkiewicz (KIT), Krzysztof Marcinkiewicz (KIT), Siarhei Shumski - Warsaw Technical University
Hiroshi Madokoro – University of Tokyo (KIT)

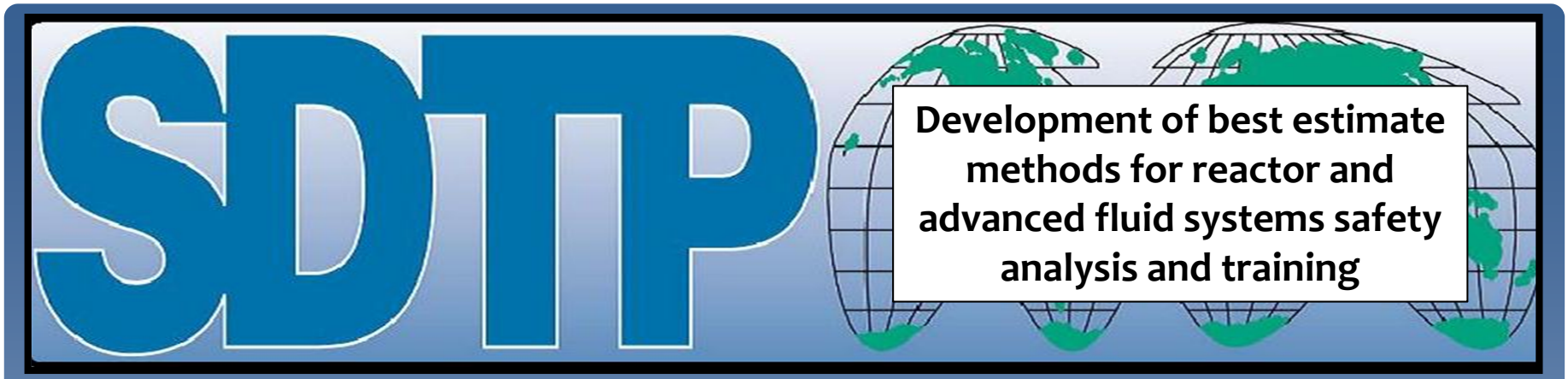
The Quench-06 experiment (ISP-45) has been used as a benchmark and training aid for Innovative Systems Software and our users/students since it was completed in the early 2000s. The experiment was first analyzed by several international organizations using RELAP/SCDAPSIM/MOD3.2. These results were submitted to the "blind" and "open" phases of the ISP. The experiment was subsequently used for basic user training for experimental analysis for our RELAP/SCDAPSIM/MOD3.4 and MOD3.5 users. Consequently, the experiment has been analyzed and discussed in four Master's theses from students in the United States, Canada, Japan, and Bulgaria. It is also used extensively in our university support and training internships.

This presentation provides one of two first examples at this meeting of an integrated uncertainty analysis of German CORA and QUENCH electrically heated experiments, looking at both the influence of uncertainties in experimental conditions and important models/correlations. The QUENCH calculations demonstrated the use of the new IUA, "Integrated Uncertainty Analysis", option introduced into RELAP/SCDAPSIM/MOD3.4 in the summer of 2017 and MOD3.5 in the fall of 2017. Both versions were used in the results provided in this presentation. The MOD3.4 analysis used a MOD3.2 input model that was specifically prepared for the ISP submittal by H. Plank from ANP-Framatome. H. Plank, using this model, provided a very accurate prediction of the experimental results in the "blind" phase of the ISP. It was subsequently improved during the open phase and then provided to ISS for use in training. The MOD3.5 analysis used two new input models. One was the original model prepared by H. Madokoro for his Master's thesis at the University of Tokyo. His model, which was a derivative of H. Plank's base model, provided a more accurate description of the bundle end fittings and took advantage of improvements that Madokoro had made to the SCDAP electrically heat fuel simulator model. The second input model was a derivative of H. Madokoro's input and was prepared by the team from Warsaw Technical University during a training internship in 2016.

The uncertainty analysis provided in this presentation looked at the influence of uncertainties in (a) parabolic equations for Zircaloy oxidation, (b) tungsten heater element resistances, (c) convective heat transfer coefficients, (d) contact resistance of the heater elements, and (e) thermal conductivity of porous zirconia used in the shroud. The uncertainty analysis demonstrated very clearly a bias introduced into the original MOD3.5 input model associated with the modeling of the shroud thermal properties. This bias has been eliminated in subsequent versions of the MOD3.5 input model and will be further refined as additional QUENCH experiments are analyzed with the new version of the code.

The uncertainty analysipresentation included comparisons between calculated and measured bundle temperatures for the CORA-17 and CORA-18 and the results of the integrated uncertainty analysis for the CORA-18 experiment.


KIT
Karlsruher Institut für Technologie



QUENCH-06 Experiment Post-test Calculations and Integrated Uncertainty Analysis with RELAP/SCDAPSIM/MOD3.4 and MOD3.5 (*with IUA*)

Analysis and contributions by
Hiroshi Madokoro – University of Tokyo (KIT)
Warsaw Technical University -
Olga Dutkiewicz (KIT), Krzysztof Marcinkiewicz (KIT), Siarhei Shumski
Dr. Marina Perez – ISS (UPC)
Gergana Gerova, Ivan Spasov – Technical University of Sofia
Hasan Celikkaya - Hacettepe University

Dr. Chris Allison
Innovative Systems Software (ISS)

Outline

* **Brief history** - Importance of Quench 6 (and ISP) for user training and model improvement
* **Application of MOD3.5 with IUA** (integrated uncertainty analysis)

Additional slides

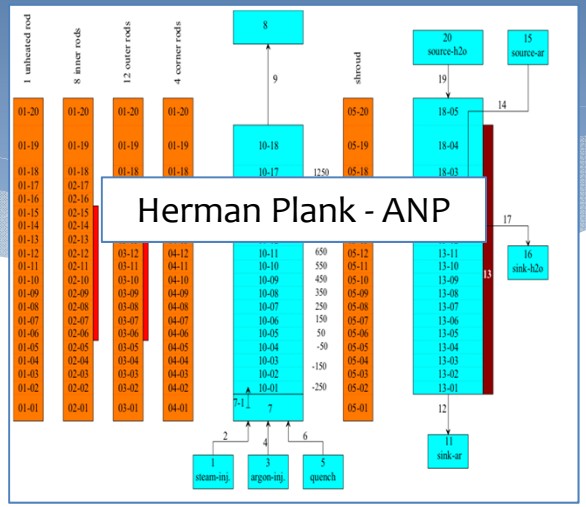
* Brief overview of IUA
* Lessons learned from historic assessment of code using QUENCH 6
* RELAP/SCDAPSIM/MOD3.5
  * Improvements to simulator component (H. Madokoro)
  * Improvements to shroud component (ISS)
* References

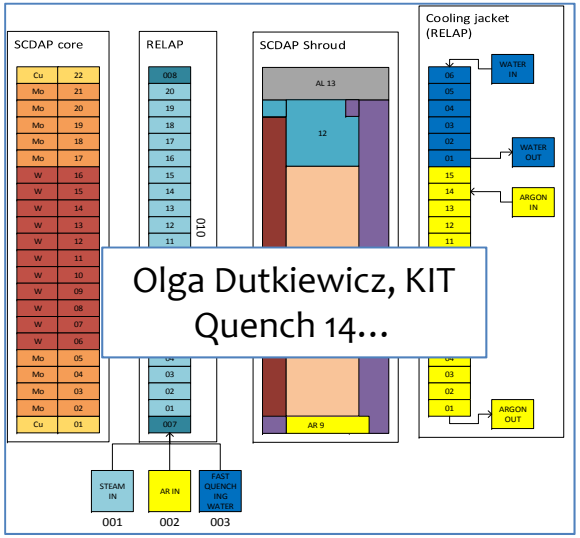
Base calculations for QUENCH-6 (ISP-45) performed using several versions of RELAP/SCDAPSIM

* **RELAP/SCDAPSIM/MOD3.2** – Models and correlations (M&C) based upon SCDAP/RELAP/MOD3.2 with improvements in CORA simulator model
  * **ISP submittals**
    * University of Zagreb
    * University of Pisa
    * NSI of RRC "Kurchatov Institute
    * CNSNS (Mexican Reg. Auth.)
    * Hacettepe University
    * ANP-Framatome (H. Plank)
    * University of Florida (ISS)
  * **Master's Thesis** – E. Honaiser (University of Florida)
* **RELAP/SCDAPSIM/MOD3.4** - MOD3.2 M&C with improved numerics, added user features, improved shroud model) - Quench 6 used for basic user training for experimental analysis
* **RELAP/SCDAPSIM/MOD3.5** - Modeling improvements based on German and other European experimental programs, incorporated KIT and PSI improvements developed by W. Herring, J. Birchley, et al., added tantalum heater element option (Russian Parameter experiments)
  * **International SDTP-sponsored university and young researcher interns**
  * **Master's Thesis** - J. Spencer, McMaster University – Performed initial assessment of MOD3.5 modeling improvements, ie - improved shroud options
  * **Master's Thesis** - H. Madokoro – University of Tokyo and KIT – **Improved SCDAP electrically heated fuel simulator component**
  * **Master's Thesis** - G. Gerova – Technical University of Sofia – Sensitivity studies on analysis results

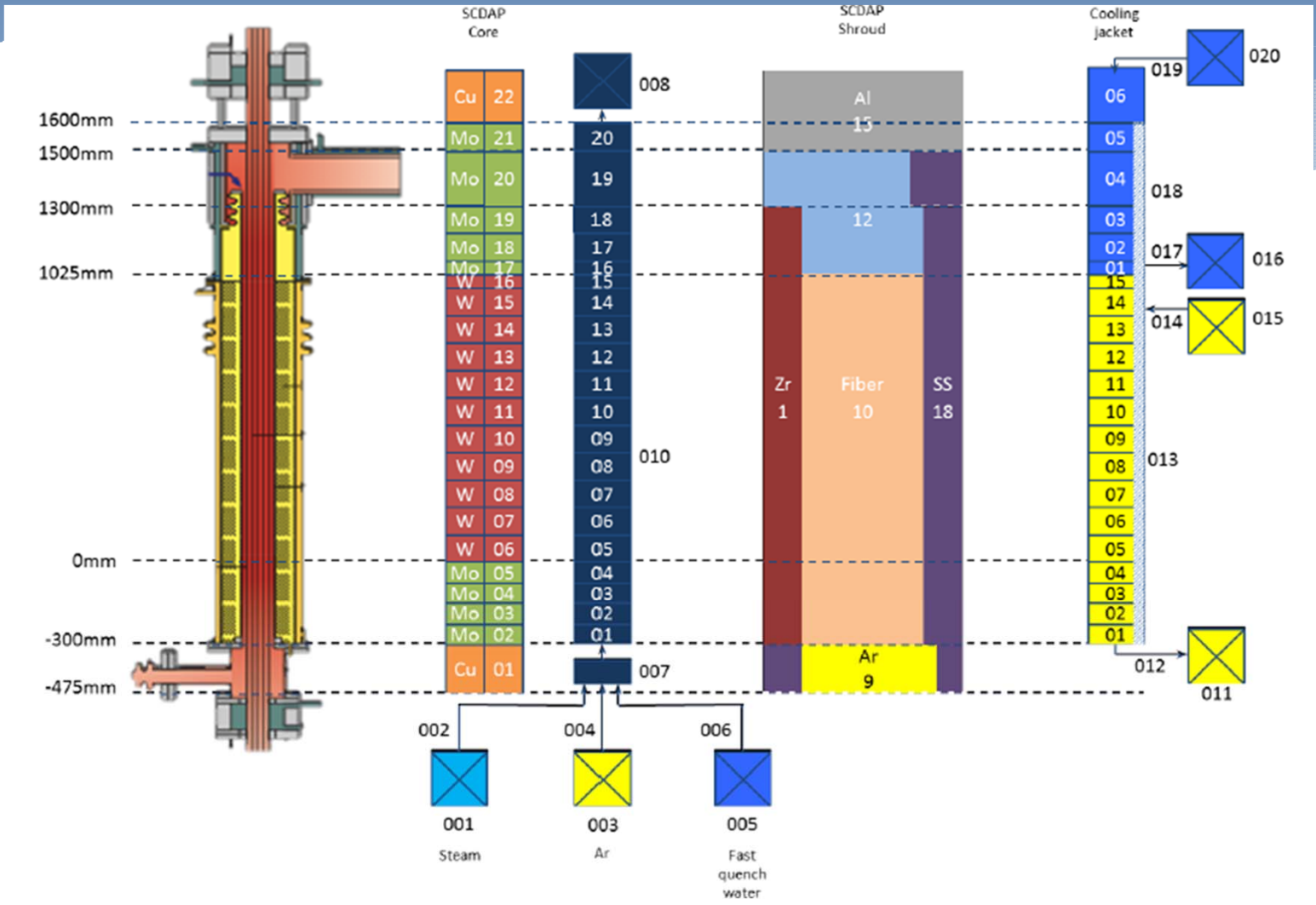
Input model and user guidelines improved for Quench facility

- **Base input model developed by ANP-Framatome (H. Plank) used for MOD3.4, and early releases for MOD3.5 for user training and analysis**
  - **Master's Thesis** - J. Spencer, McMaster University – Performed initial assessment of MOD3.5 modeling improvements, ie - improved shroud options
  - **Master's Thesis** - H. Madokoro – University of Tokyo and KIT – **Improved SCDAP electrically heated fuel simulator component**
- **New MOD3.5 input model for Quench facility developed by H. Madokora (U. Tokyo/KIT) is now recommended as our base MOD3.5 input model**
  - Technical University of Sofia, North Carolina State University, Warsaw Technical University, University of Tokyo…
    - **Standardized input and guidelines for modeling of heater rod and shroud**





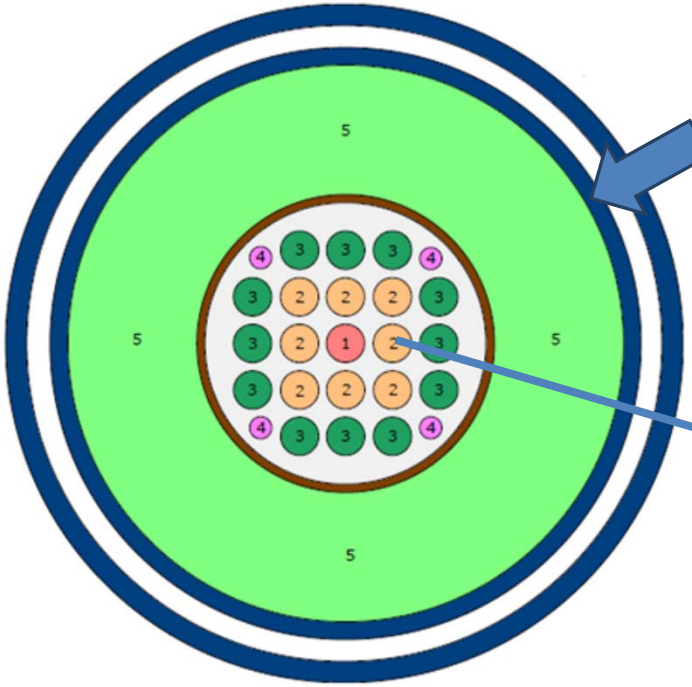
RELAP/SCDAPSIM/MOD3.5dp nodalization – based on H. Madokoro's revisions to MOD3.5 simulator component

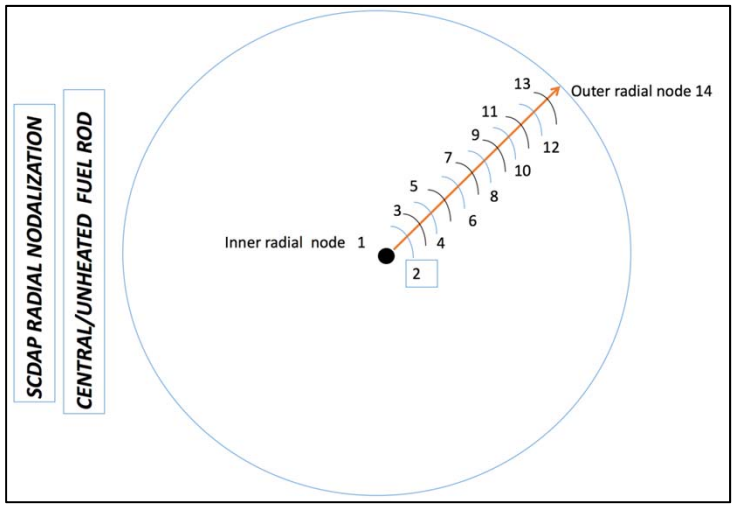


RELAP/SCDAPSIM/MOD3.5dp bundle cross-section and components used

SCDAP components

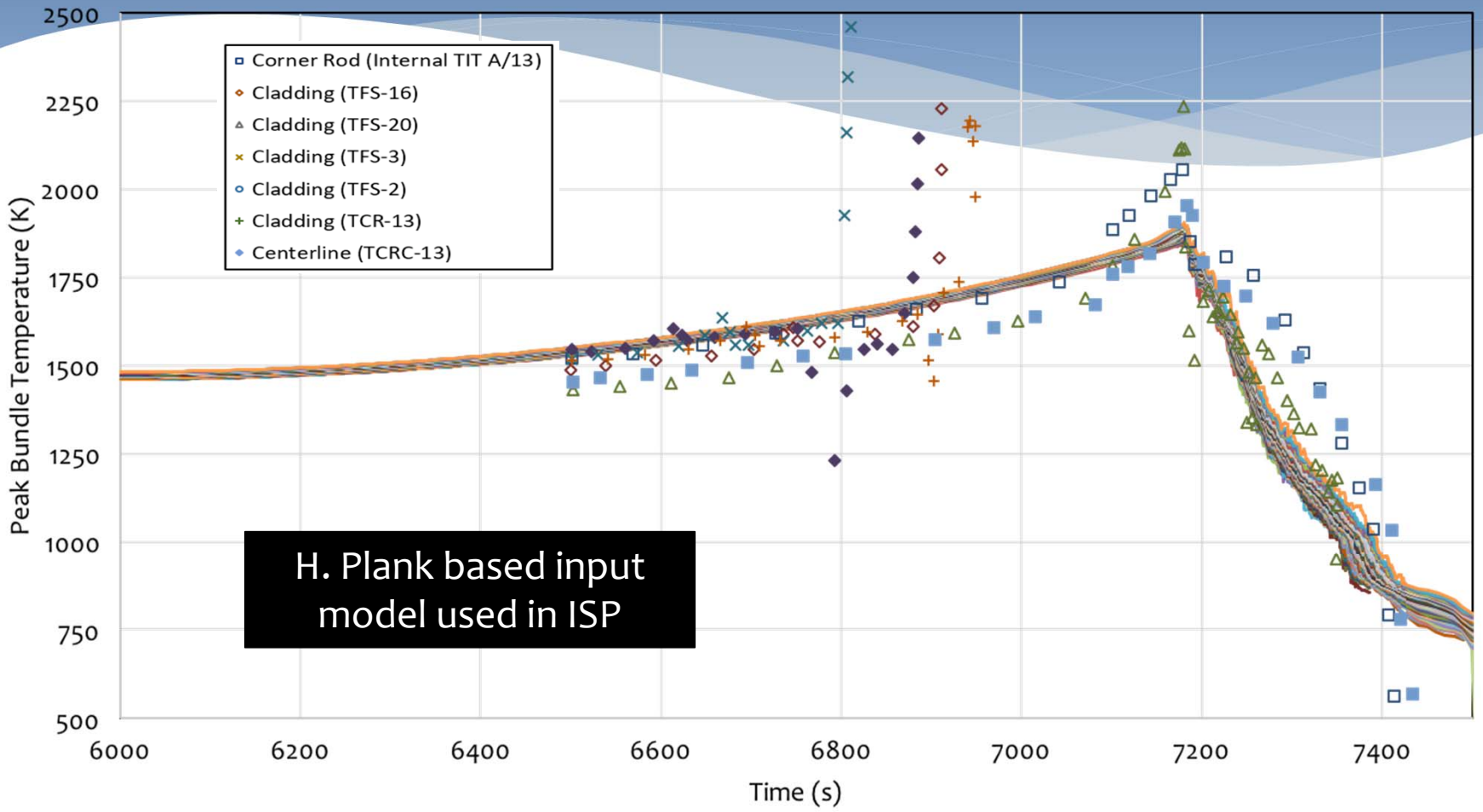
1. Unheated fuel rod
2. Inner simulator rod
3. Outer simulator rod
4. Instrumentation
5. Shroud





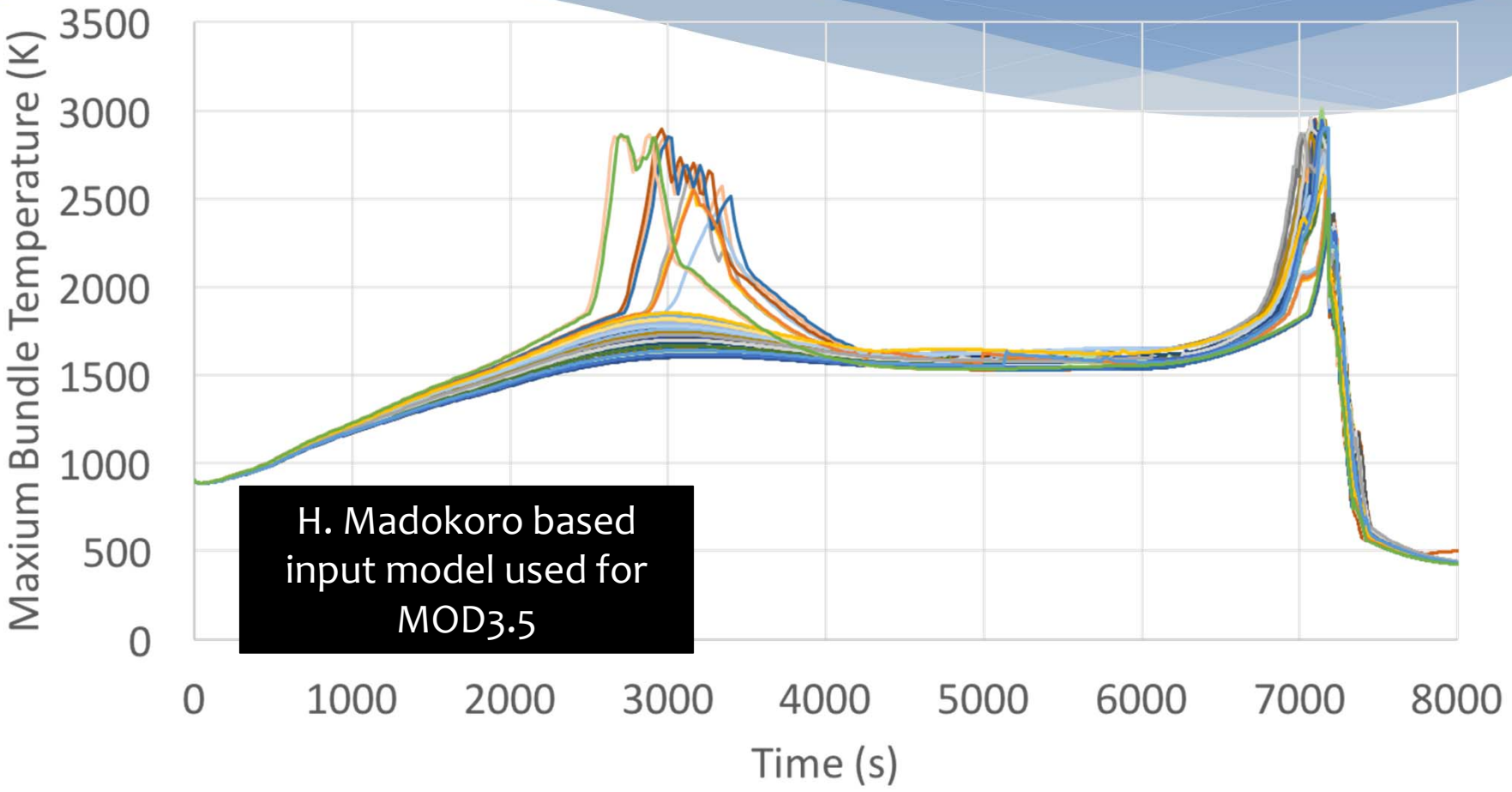
Peak Bundle Temp. – MOD3.4(IUA)

(Comparison with measured temperatures – 950mm)

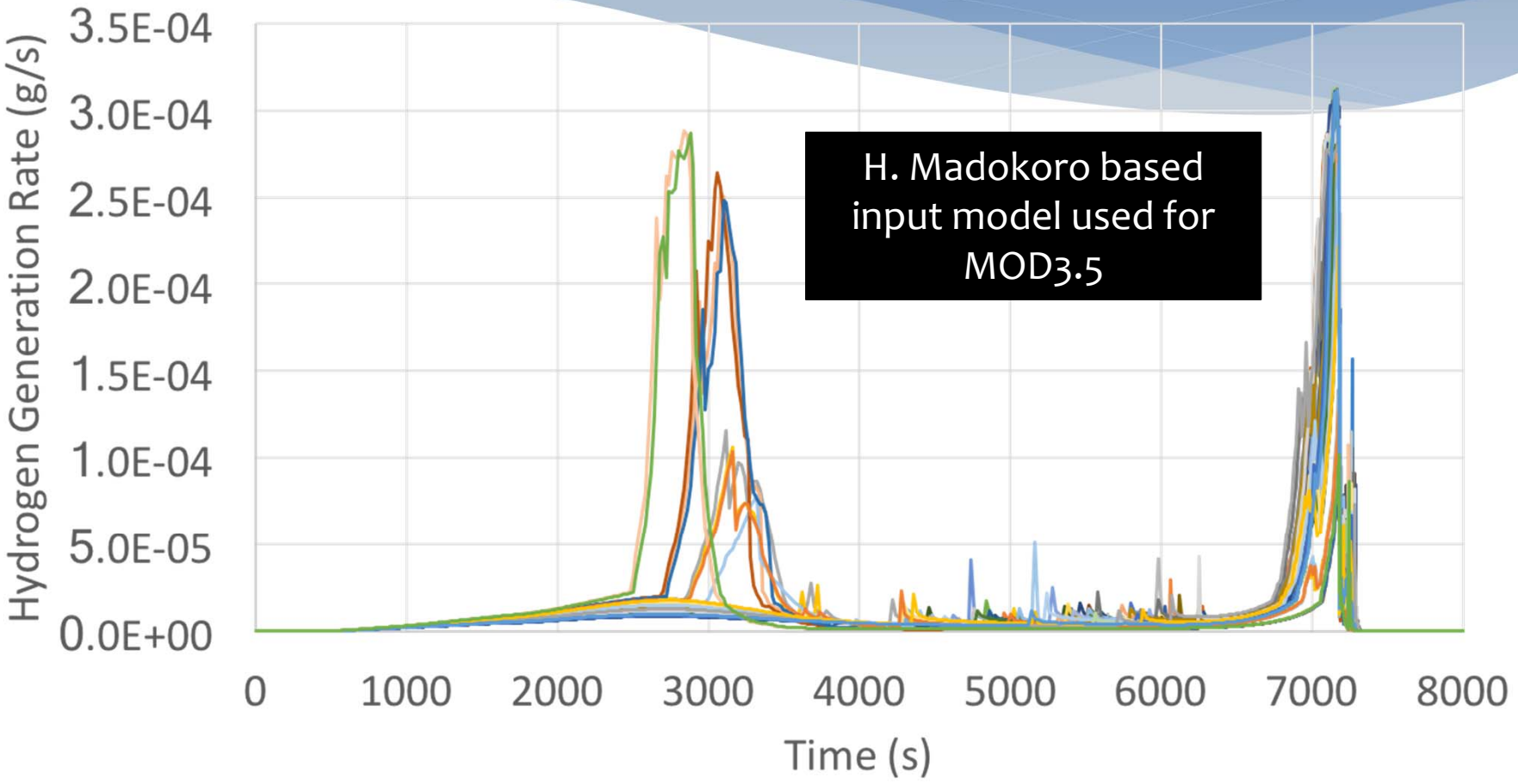


H. Plank based input model used in ISP

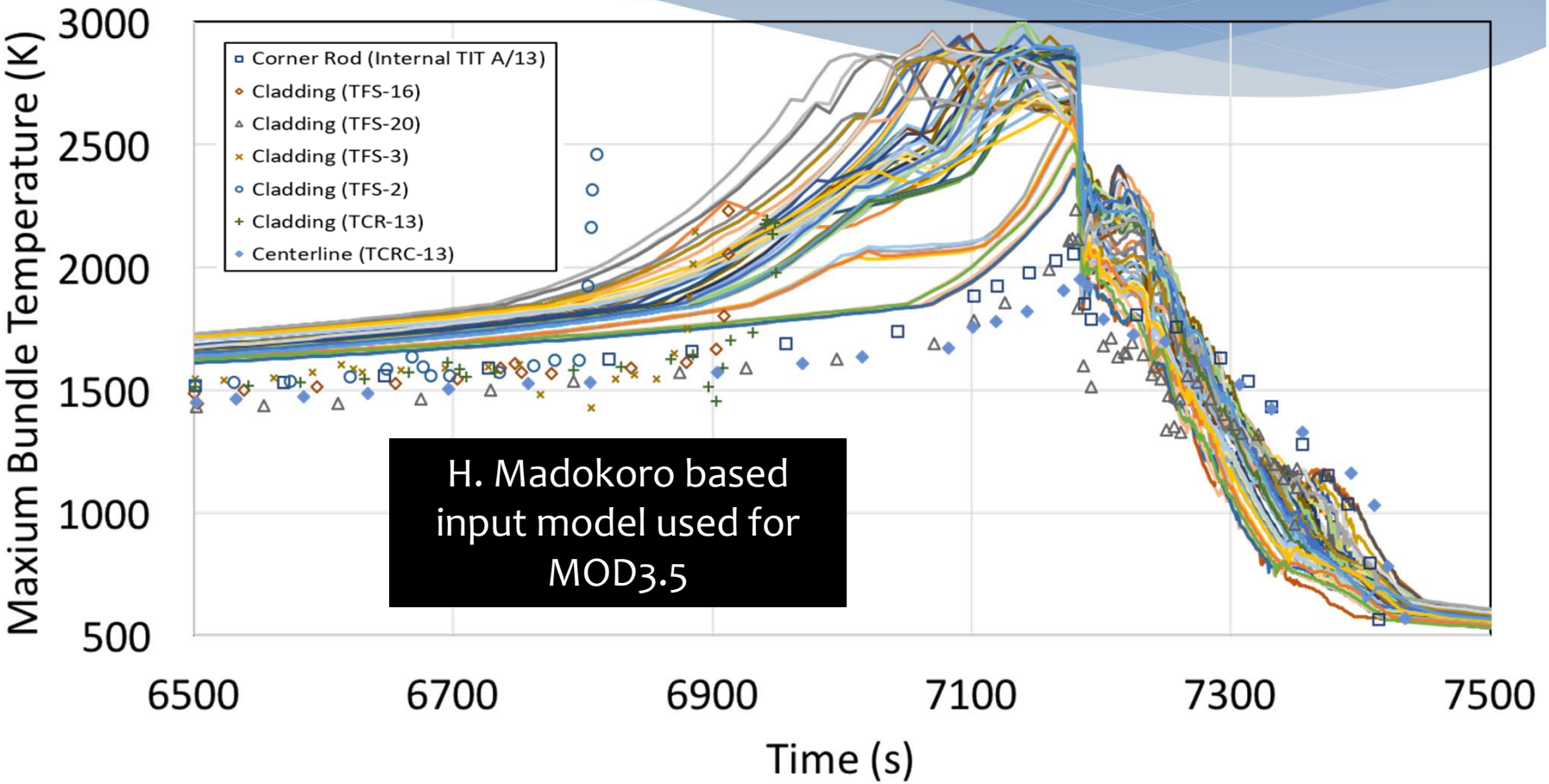
Peak Bundle Temperatures – MOD3.5(IUA)



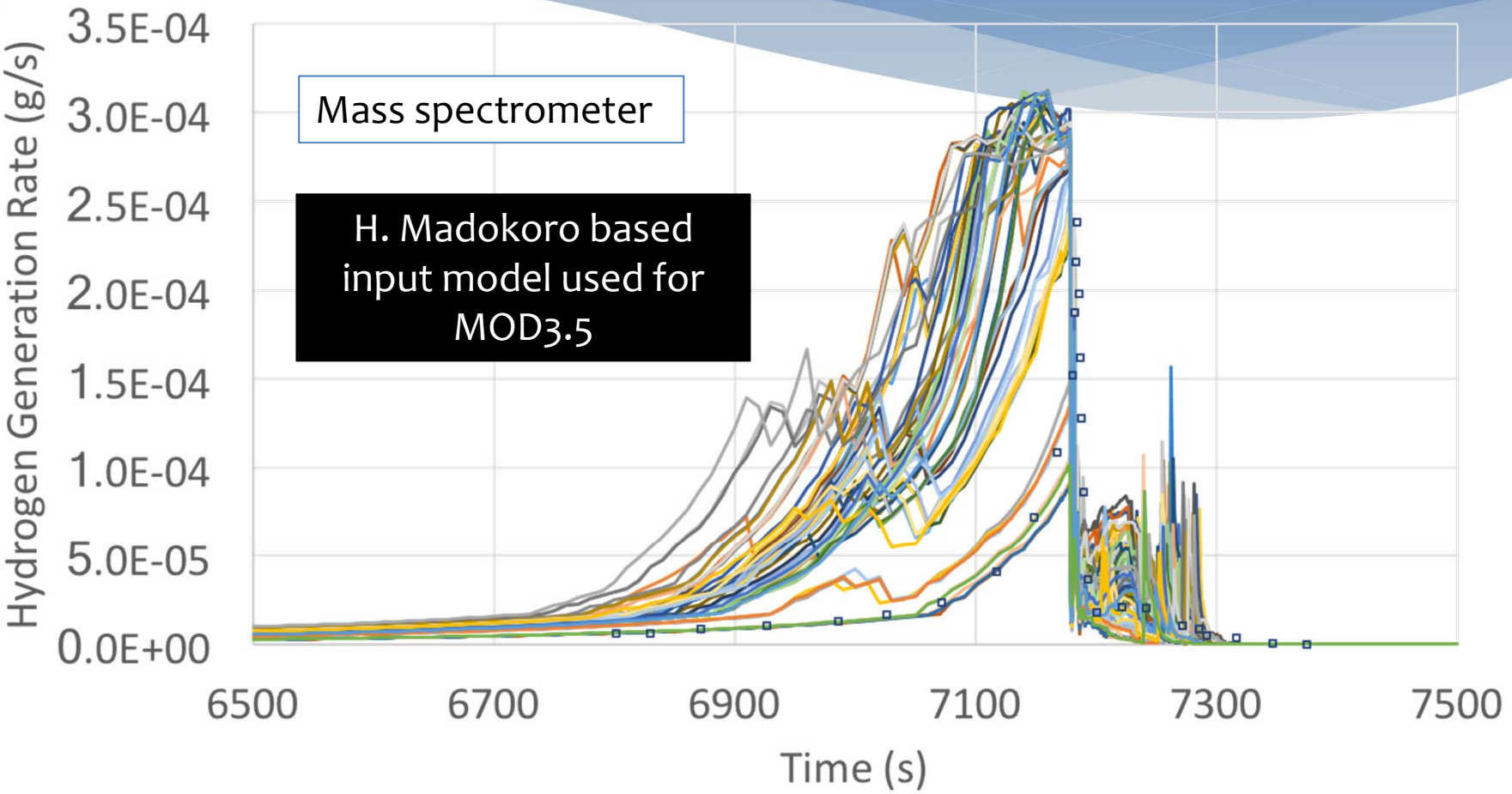
Hydrogen Generation Rate – MOD3.5(IUA)



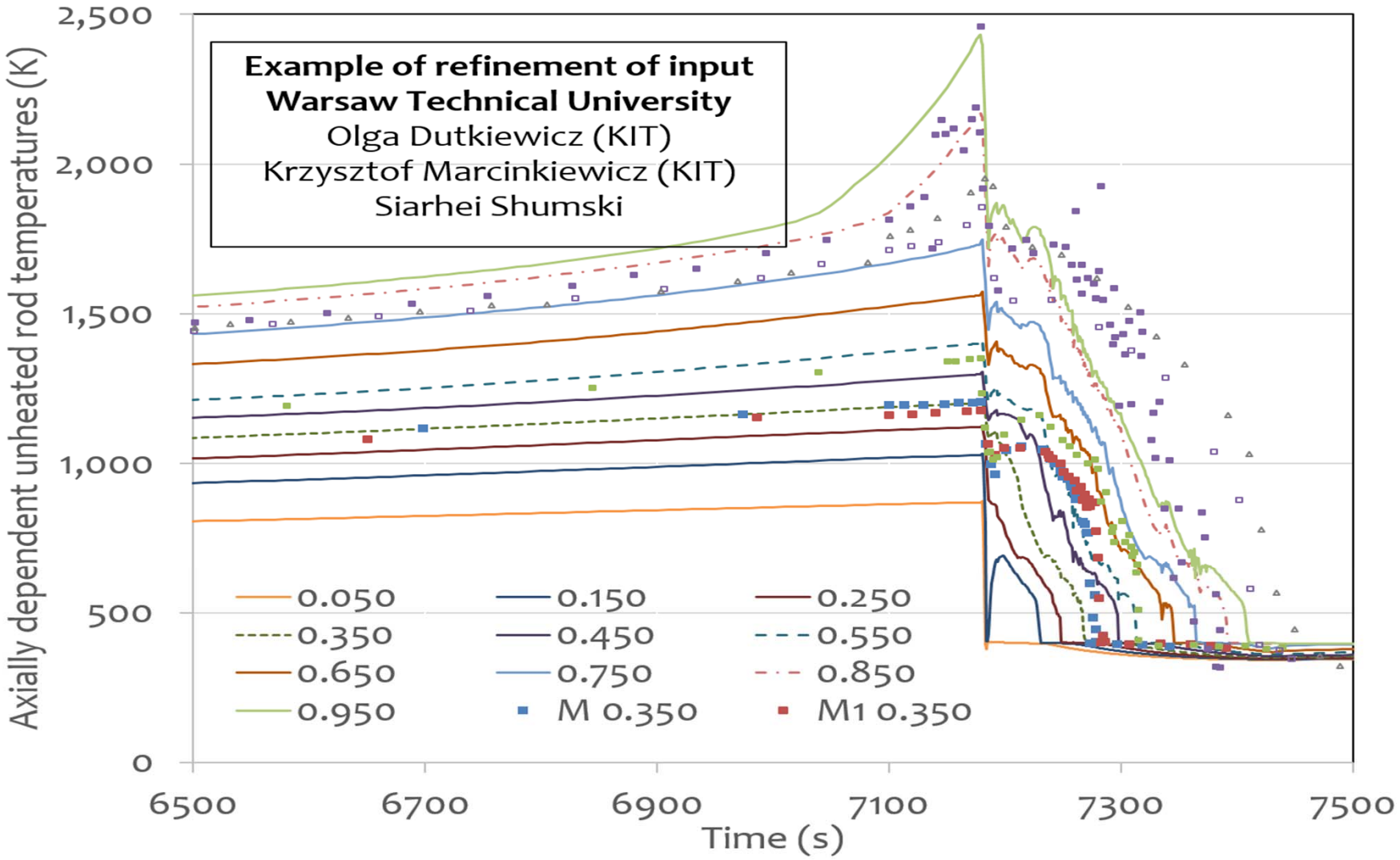
Peak Bundle Temp. – MOD3.5(IUA)
(Comparison with measured temperatures – 950mm)



Hydrogen Gen. Rate – MOD3.5 (IUA) (Comparison with measured rates)



Refinement of base MOD3.5 input improved calculated response (WTU input)



Preliminary conclusions from reassessment – Quench 6

* MOD3.5 with more detailed simulator and shroud models more accurately reflects Quench facility design and conditions
  * New MOD3.5 base Quench input model more detailed and better represents facility, but
    * Needs further refinement to eliminate systematic bias in peak bundle temperatures
    * MOD3.4 still can provide realistic results using ISP based input model (H. Plank – ANP) but may under predict peak bundle temperatures in Quench-06 conditions (T_{peak} ~ 2000K)

Additional slides

Brief overview of IUA (integrated uncertainty analysis)

Lessons learned from historic assessment of code using QUENCH 6

RELAP/SCDAPSIM/MOD3.5 Improvements

References

Brief overview of IUA (integrated uncertainty analysis)

* RELAP/SCDAPSIM/MOD3.X(IUA) allows user to easily compute ranking statistics and uncertainty distributions using base input model
  * User defines parameters to include plus uncertainty distributions for defined parameters
    * Input parameters
    * Source code parameters
  * New "setup" option builds input models and uncertainty files for uncertainty analysis
  * New "post processing" option computes uncertainty distributions and ranking statistics for user selected quantities

QUENCH-06 uncertainty analysis … RELAP5 SETUP input(1)

100 uncsetup

29000000 0 0 0.95 0.95 1 0 1 200 298341

* Source correlations parameters

*Heat transfer coef. (Wall to fluid heat transfer - vapor)

29000002 HTC.09.110 UD 0.70 1.30 … *Trapezoidal distribution

*Tungsten resistivity

29000003 RES.COR.TUNG ND 1.00 0.025 … *Normal distribution

*Oxidation rate

29000004 OXI.MATPRO ND 1.00 0.075 … *Normal distribution

QUENCH-06 uncertainty analysis … RELAP5 SETUP input – MOD3.5

*Shroud thermal conductivity

29101000 UD 0.8 1.2 0.0 0.0

29101001 -1.0 20 20100401 2 2 1

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.

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*external heater rod resistance

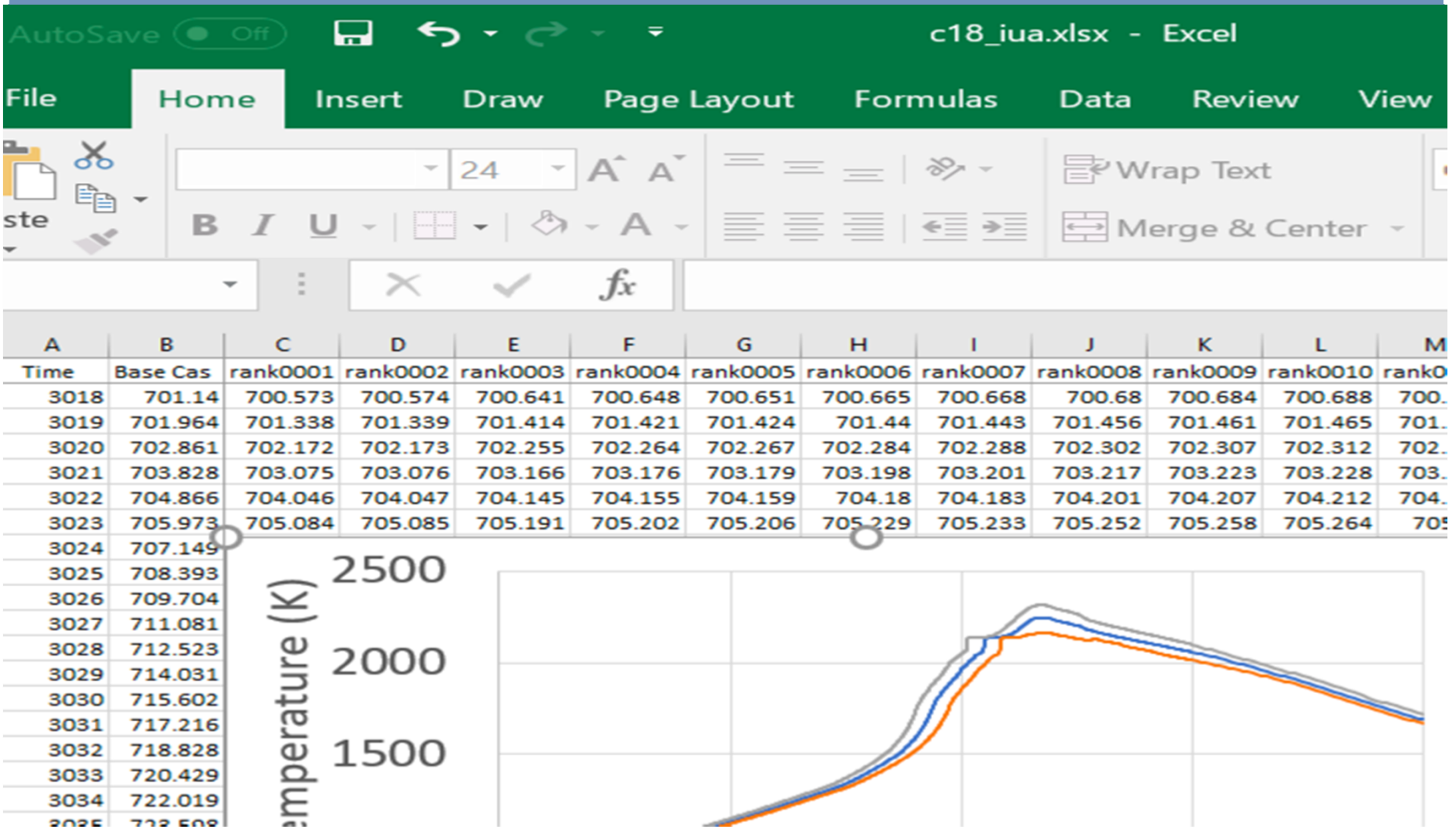
29102000 UD 0.814814815 1.185185185 0.0 0.0

29102001 -1.0 20 40020100 4 4 1

*Base input model

…

QUENCH-06 uncertainty analysis … generating uncertainty distributions and ranking



Time	Base Cas	rank0001	rank0002	rank0003	rank0004	rank0005	rank0006	rank0007	rank0008	rank0009	rank0010	rank0
3018	701.14	700.573	700.574	700.641	700.648	700.651	700.665	700.668	700.68	700.684	700.688	700.
3019	701.964	701.338	701.339	701.414	701.421	701.424	701.44	701.443	701.456	701.461	701.465	701.
3020	702.861	702.172	702.173	702.255	702.264	702.267	702.284	702.288	702.302	702.307	702.312	702.
3021	703.828	703.075	703.076	703.166	703.176	703.179	703.198	703.201	703.217	703.223	703.228	703.
3022	704.866	704.046	704.047	704.145	704.155	704.159	704.18	704.183	704.201	704.207	704.212	704.
3023	705.973	705.084	705.085	705.191	705.202	705.206	705.229	705.233	705.252	705.258	705.264	705
3024	707.149											
3025	708.393											
3026	709.704											
3027	711.081											
3028	712.523											
3029	714.031											
3030	715.602											
3031	717.216											
3032	718.828											
3033	720.429											
3034	722.019											

Lessons learned from Quench 6 ISP – Presentation Quench meeting

RELAP/SCDAPSIM/MOD3.2(bd) used by ISP participants

- New models added for ISP
  - Generalized tungsten heating element for CORA,Quench, and other electrically heated facilities
    - Based on FzK modeling improvements
  - Improved shroud model with axially varying properties

Original SCDAP/RELAP5/MOD3.2 simulator model could not be used

- Original simulator model hardwired for CORA heater rod design
- Several SCDAP/RELAP5/MOD3.2 coding (and input manual) errors had to be removed
  - Fuel rod simulator model
  - Radiation heat transfer

- Experience and level of effort important factor in best calculations
  - Plank of Framatome-ANP developed most complete input model
    - Modeled full height of simulator rods including electrodes
    - Included radiation heat losses in upper shroud
  - Experienced users able to overcome code problems and modeling weaknesses
    - Initial calculations impacted by error in original SCDAP simulator model
    - Error in shroud model impacted radial heat transfer boundary condition to bypass region

Influence of radial heat losses - Quench 6 ISP – Presentation Quench meeting

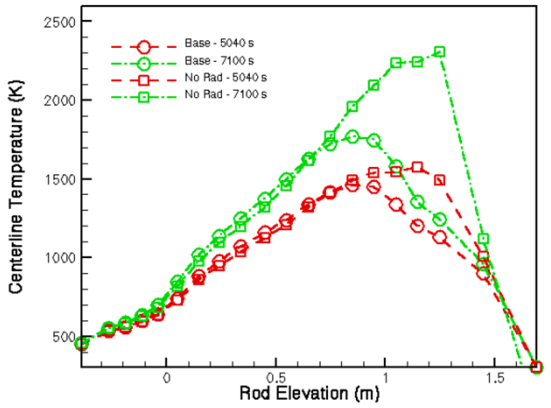
Study used input model developed by H. Plank

- Model was most detailed of ISP submittals
  - Entire electrode of simulator modeled
  - Axial variation in shroud modeled
    - **Radiation in upper shroud gap considered**
- Modeling defaults used except
  - U-Zr-O relocation temperature = 2200 K
  - Ballooning parameters

Prime Objective of Study

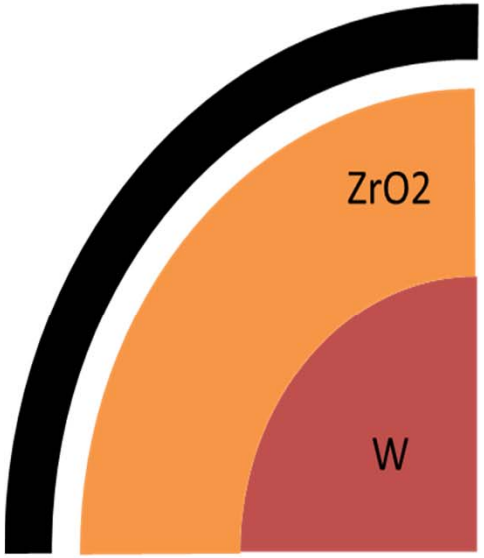
- Quantify impact of radial heat losses in upper electrode region
  - Argon thermal conductivity varied from 0% to 100% of base conductivity
    - 0 % case represents argon thermal conductivity ignoring convection and radiation

Neglecting radiation/convection in argon results in unrealistic heating above heated zone



RELAP/SCDAPSIM/MOD3.5dp – Improvements to simulator component (H. Madokoro)

* **Fuel gap: Inner gap between the fuel and the heater is also considered**

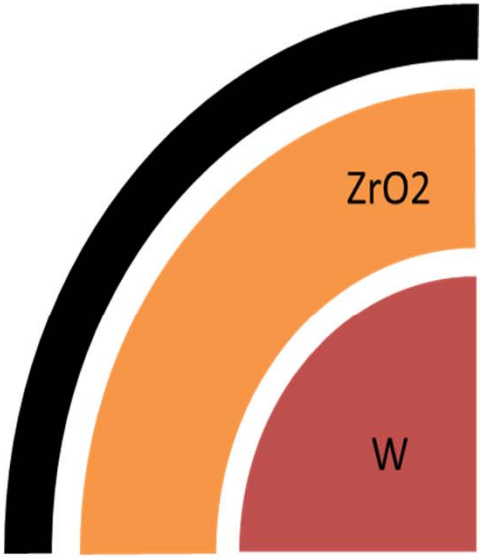


◂ Original model

- Gap between heater and pellet is not considered

New model ▸

- Gap between heater and pellet is considered. Radiation heat transfer as well as conduction.



RELAP/SCDAPSIM/MOD3.5dp – Improvements to simulator/shroud component

H. Madokoro - simulator

* Improve axial resistivity and power distribution
  * More detailed description of heater element including end fittings can be included in input option
* Option to include measured total heater resistance in resistivity calculation

ISS -SCDAP components

* Improve radial heat losses through shroud in upper test train
  * Axially dependent material layers
* Improve radial heat conduction in gaps
  * Option to automatically include radiation exchange in gas-filled gaps

Historical References

* S. Sadek, "RELAP/SCDAPSIM (MOD3.2-bht) Analysis of the QUENCH-06 Experiment, University of Zagreb Report FER-ZVNE/SA/DA-ir01/03-0, 2003.
* L. Sepold, W. Hering, et al. "Experimental and Computational Results of the QUENCH-06 Test (OECD ISP-45)", Forschungszentrum Karlsruhe (KIT) report FZKA 664, 2004.
* C.M. Allison, "Quench-06 Analysis Using RELAP/SCDAPSIM/MOD3.2 – Impact of Radial Heat Losses", International Quench Meeting, Karlsruhe, December 2001.
* C.M. Allison, "Quench-ISP Analysis Using RELAP/SCDAPSIM/MOD3.2(BD) – Lessons Learned", International Standard Problem Comparison Meeting, Karlsruhe, December 2001.

MOD3.4/MOD3.5 References

* J. K. Hohorst, C.M. Allison, L. J. Siefken, Assessment of New Modeling in RELAP/SCDAPSIM/MOD3.5 Using Experimental Results from the Quench Program, Proceedings of ICAPP '09, Tokyo, Japan, May 2009.
* J. H. Spencer, C. M. Allison, and J. K. Hohorst, "Assessment of new modeling in RELAP/SCDAPSIM using experimental results from the Quench program," in *ICAPP 2011*, 2011.
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* H. Madokoro, "Assessment of RELAP/SCDAPSIM with QUENCH and CORA Analysis", Master Thesis, Department of Nuclear Engineering and Management, School of Engineering, University of Tokyo, 37-126808, September 2014.

MOD3.4/MOD3.5 References

* T. Kaliatka, A. Kaliatka, V. Vileiniškis, and E. Ušpuras, Modelling of QUENCH-03 and QUENCH-06 Experiments Using RELAP/SCDAPSIM and ASTEC Codes, Hindawi Publishing Corporation Science and Technology of Nuclear Installations Volume 2014, Article ID 849480.
* G. Gerova, C. Allison, "Quench-06 Analysis Using RELAP/SCDAPSIM/MOD3.5", Bulgarian Nuclear Society Meeting, September 2016.

Title:

QUENCH-14, QUENCH-16 and QUENCH-ALISA analysis with RELAP5/SCDAPSIM MOD3.5. and MOD3.5+

Presenters:

Olga Dutkiewicz, Krzysztof Marcinkiewicz

Abstract:

QUENCH experiments, conducted in Karlsruher Institut für Technologie, investigate the behavior of materials, such as cladding materials or absorber materials used in nuclear reactors, under accidental conditions. The focus is on important aspects such as hydrogen production, oxidation of the materials, coolability, and lately, on air ingress, which can enhance the risk of accidental scenario in a nuclear power plant. The QUENCH tests use electrically heated rods to simulate nuclear fuel behavior, and RELAP5/SCDAPSIM has a well-established electrically heated rod model. Both QUENCH-14 and QUENCH-ALISA use M5® cladding material; QUENCH-16 and QUENCH-ALISA are both air ingress experiments. Oxidation rate of M5® cladding material has been investigated by KIT and their results are used in these analyses. Air ingress is an issue over which research has begun relatively recently and it demands a lot of further work, regarding zircaloy nitridation phenomena. The analyses of QUENCH-14, QUENCH-16 and QUENCH-ALISA experiments use RELAP5/SCDAPSIM in MOD3.5. and in MOD3.5+, a version that has been available for a small group of users in a process of testing. RELAP5/SCDAPSIM MOD3.5. has no problem with reflecting the bundle behavior in experiments such as QUENCH-14, however the distinct nature of QUENCH-16 and QUENCH-ALISA, being air ingress tests and having different electrical power history trend, creates a need to introduce new reaction kinetics and oxidation rates to the code's library.


KIT

QUENCH-14, QUENCH-16 and QUENCH-ALISA analysis with RELAP5/SCDAPSIM MOD3.5(KIT) and MOD3.5+(KIT)

***Presenting authors**:*

Olga Dutkiewicz, Krzysztof Marcinkiewicz

***Co-authors**:*

Hiroshi Madokoro, dr Heinrich Muscher, dr Juri Stuckert

***Special thanks to**:*

dr Martin Steinbrück, dr Chris Allison



Outline

1. Introduction: title, outline, overview.
2. QUENCH-14 analysis with RELAP5/SCDAPSIM MOD3.5. and MOD3.5+ both using *M5* oxidation rate*.
3. QUENCH-16 analysis with MOD3.5.
4. Pre-test calculations for QUENCH-ALISA with MOD3.5. and MOD3.5+
5. Post-test calculations for QUENCH-18 / QUENCH-ALISA in MOD3.5+
6. Summary and conclusions.
7. Future plans regarding RELAP5/SCDAPSIM MOD3.6.
8. References.
9. The end.

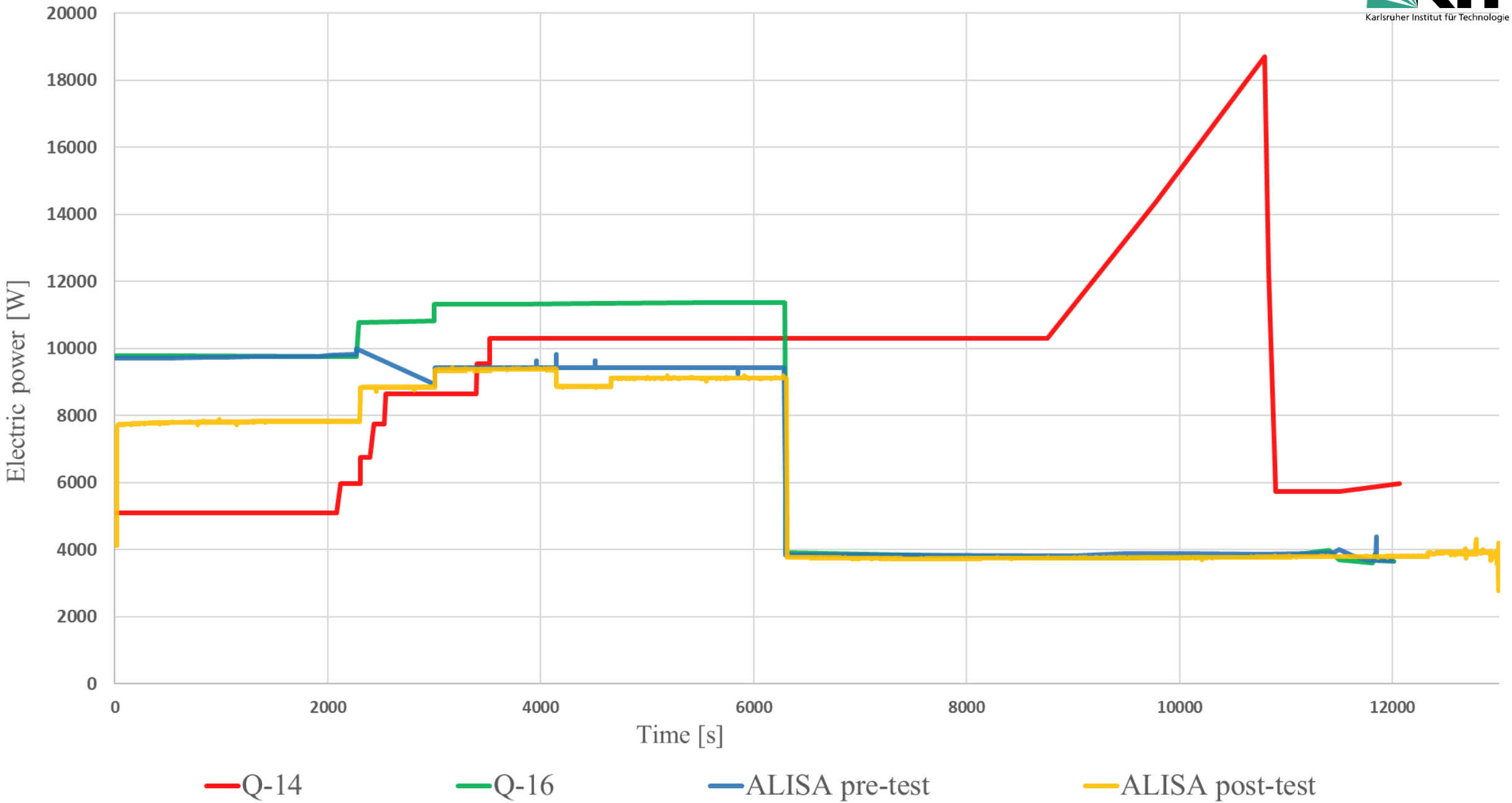


Overview

- KIT's QUENCH – experiments, using electrically heated simulator rods, and investigating the behavior of materials, for example cladding or absorber, under accidental conditions;
- QUENCH focuses on oxidation, hydrogen production, melting, coolability;
- QUENCH-14 and QUENCH-ALISA / 18 – both have M5® cladding;
- QUENCH-16 and QUENCH-ALISA – both are air ingress tests.

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Comparison of power histories in QUENCH-14, QUENCH-16 and QUENCH-ALISA



QUENCH-14 with M5*

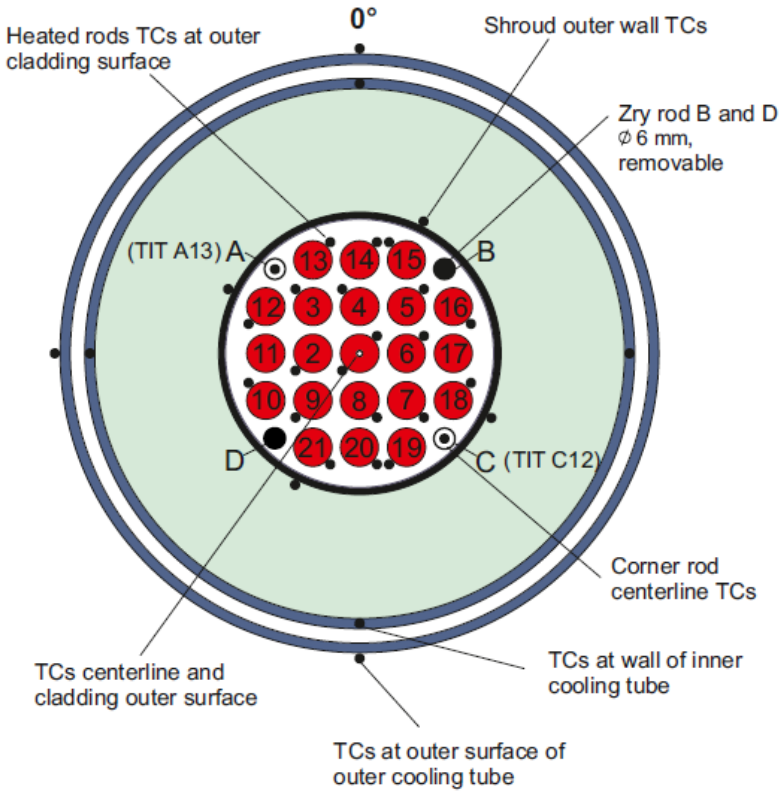


Fig. QUENCH-14 test bundle. [1]

QUENCH-14 used M5. In the analysis we use M5 oxidation rates (according to [2]) instead of standard MATPRO Zircaloy oxidation rates.

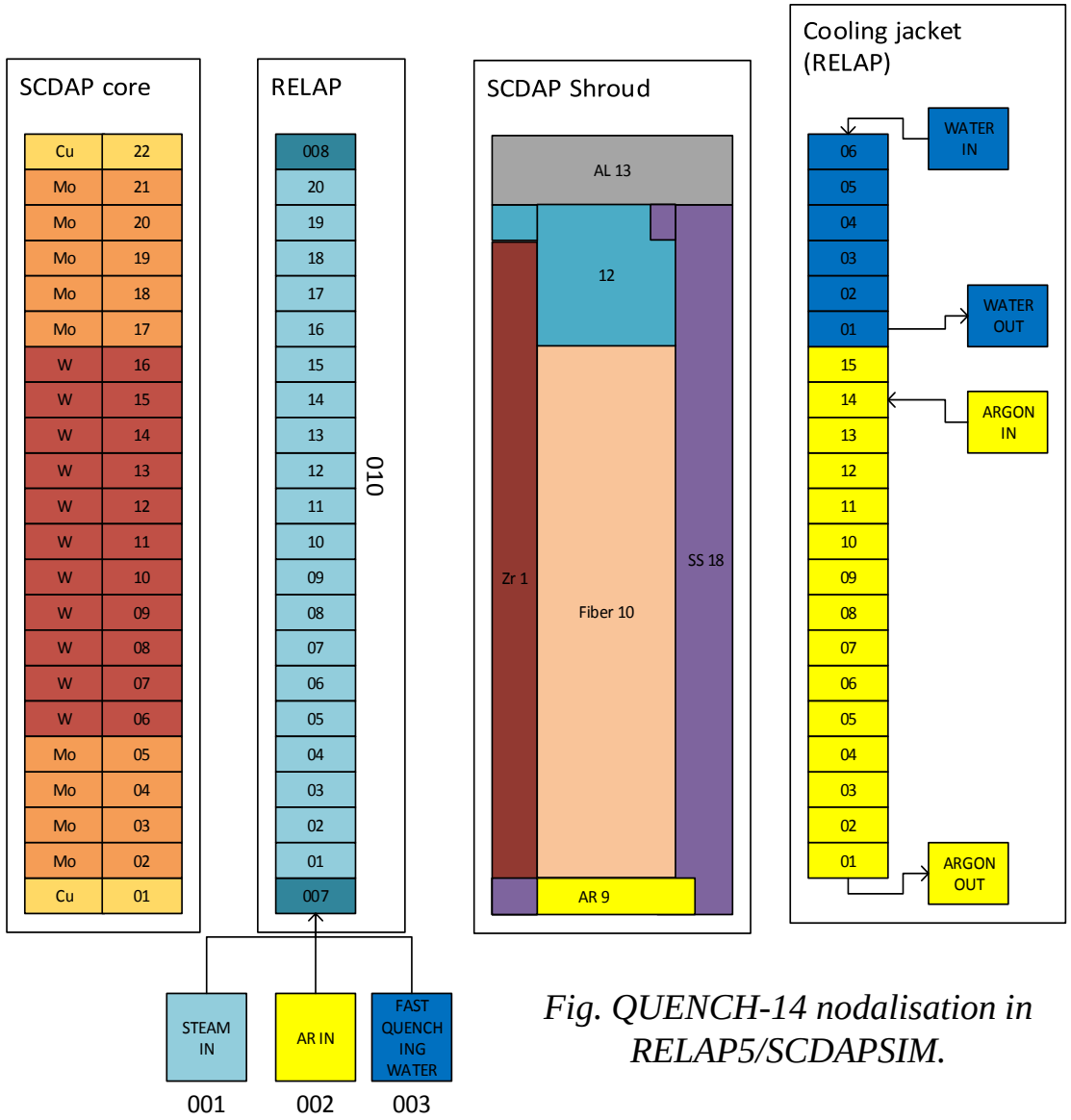
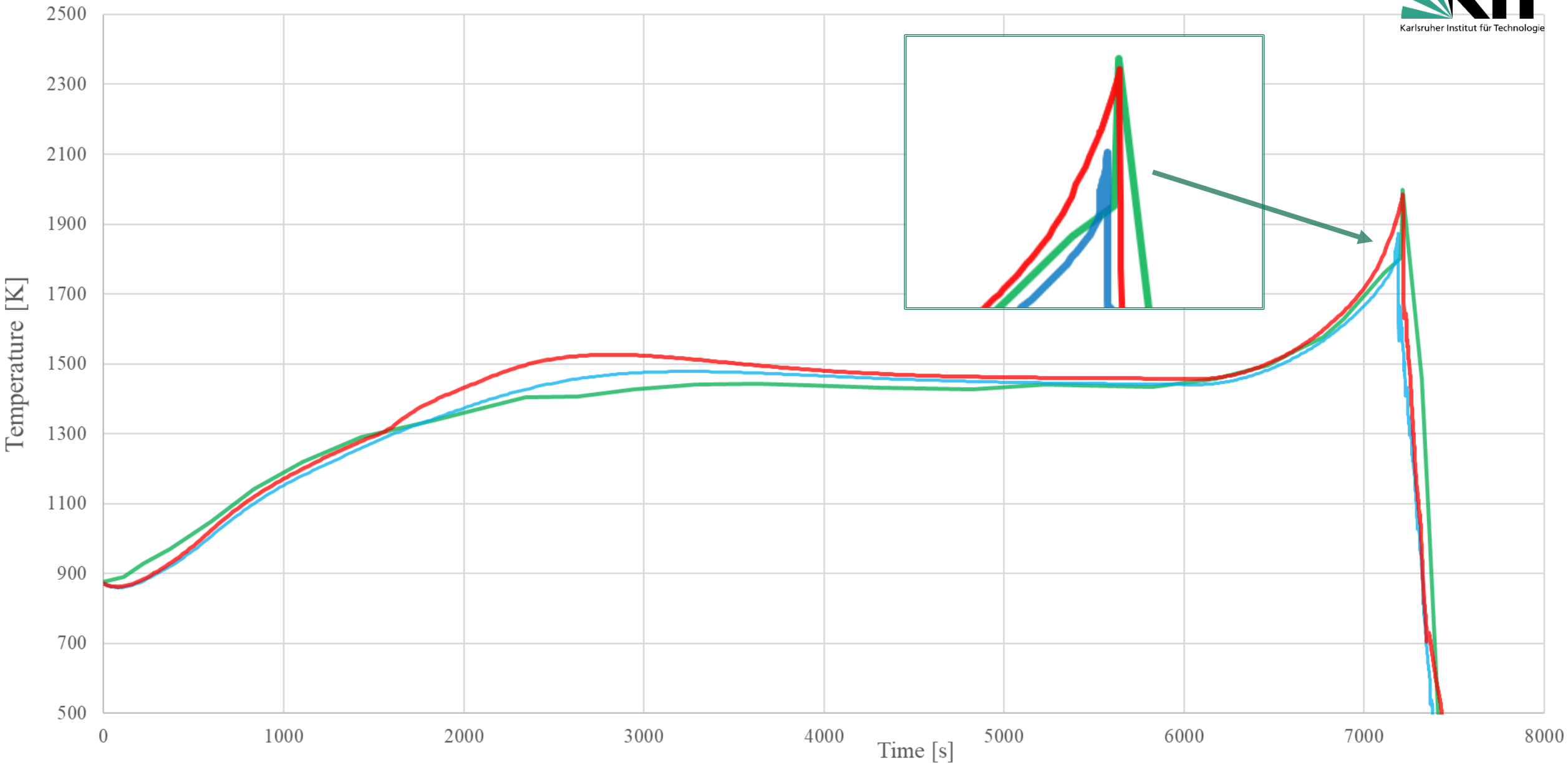
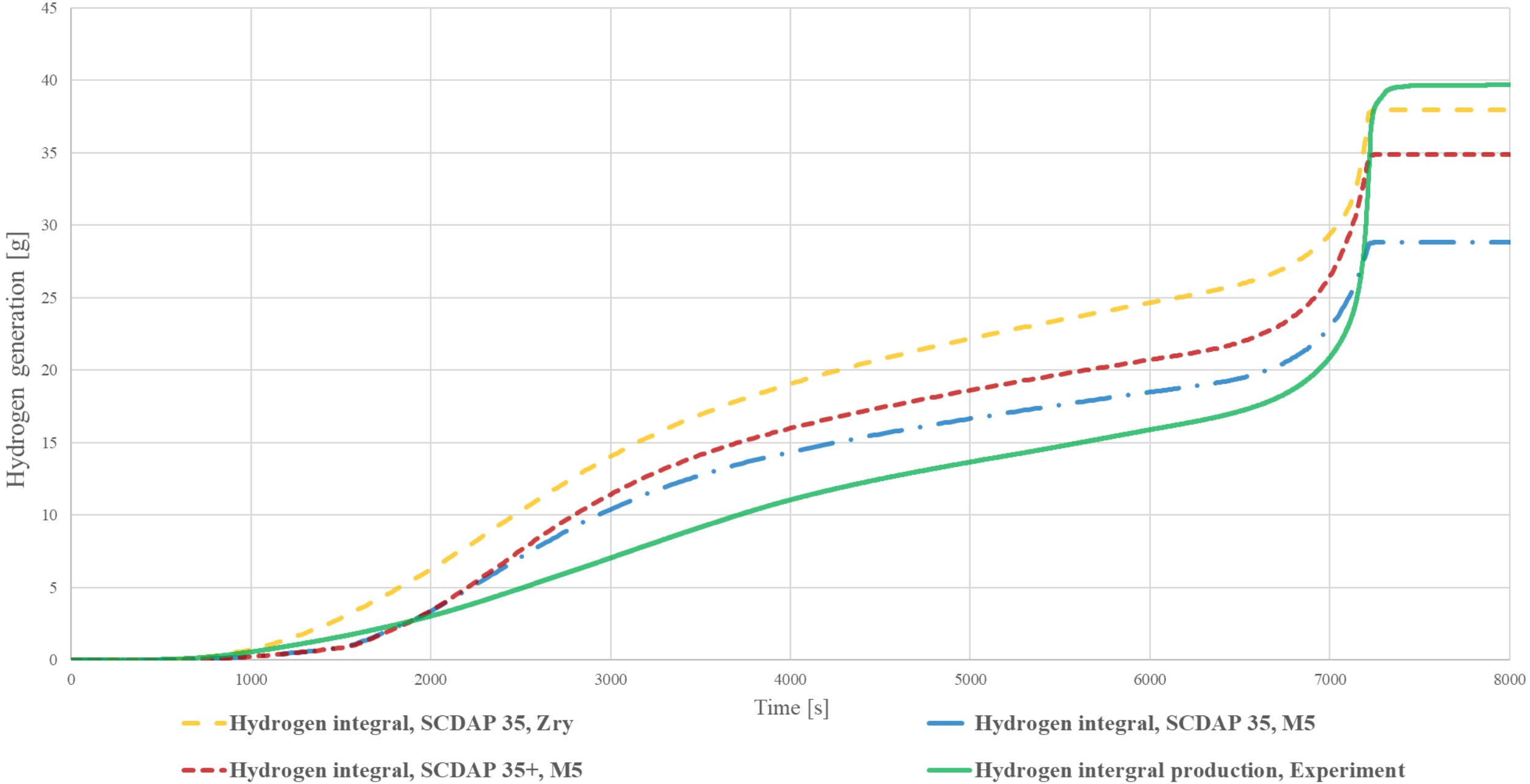


Fig. QUENCH-14 nodalisation in RELAP5/SCDAPSIM.

Temperatures at elevation 850 mm in Q-14 experiment and from calculations in RELAP5/SCDAPSIM



Total hydrogen production [g] in QUENCH-14



QUENCH-16

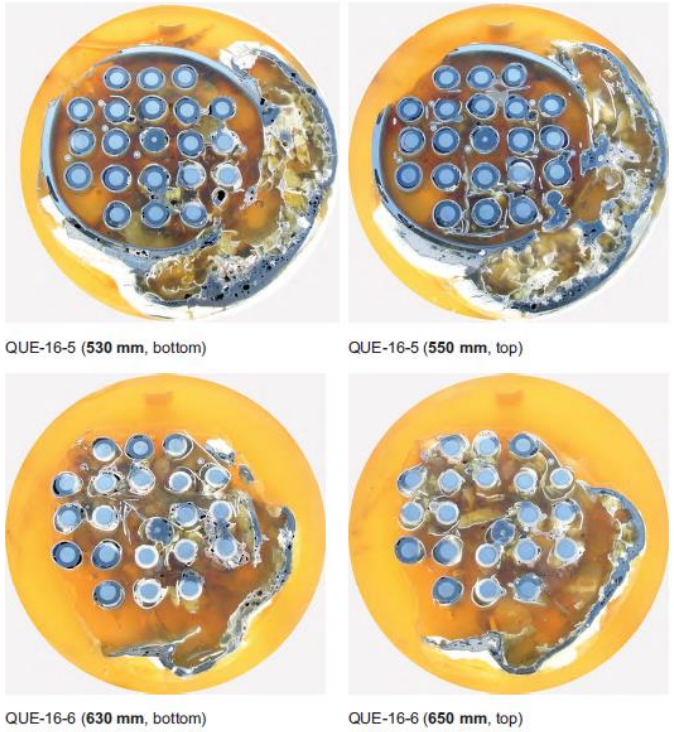


Fig. QUENCH-16 bundle cross sections at 530 mm, 550 mm, 630 mm and 650 mm. [3]

In this experiment there was a significant **shroud melt** at elevations between ~500 and ~700 mm. The shroud failure lead to water leakage in an amount of 7264 g, found post test in the ZrO_2 fiber [3].

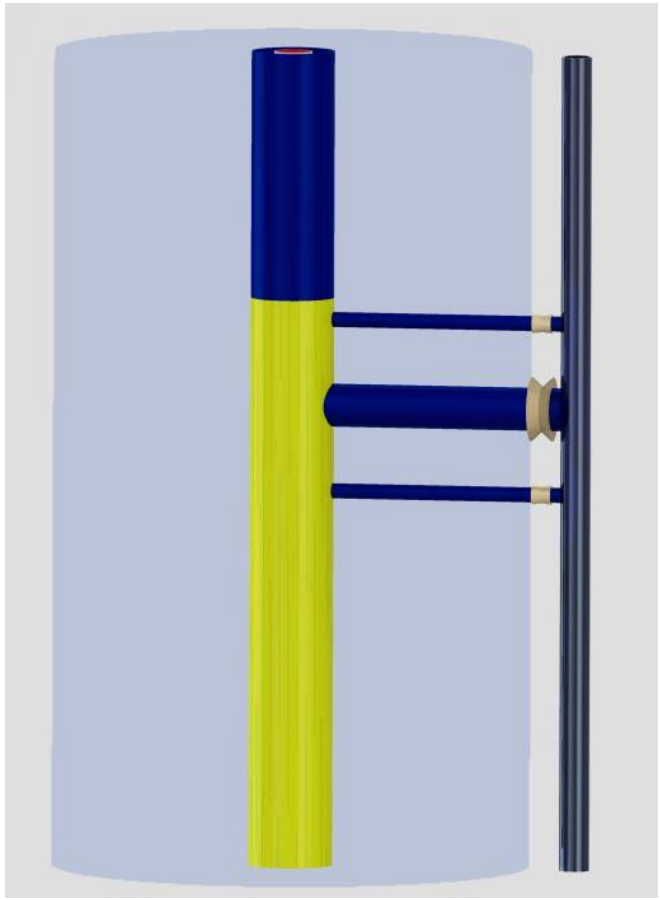


Fig. 3D QUENCH-16 model representation. The shroud failure, with water leaking from the bundle, is simulated using valves and a pipe of a volume equal to the volume available in the ZrO_2 fiber. ***Please note that sources and sinks are not drawn here.***

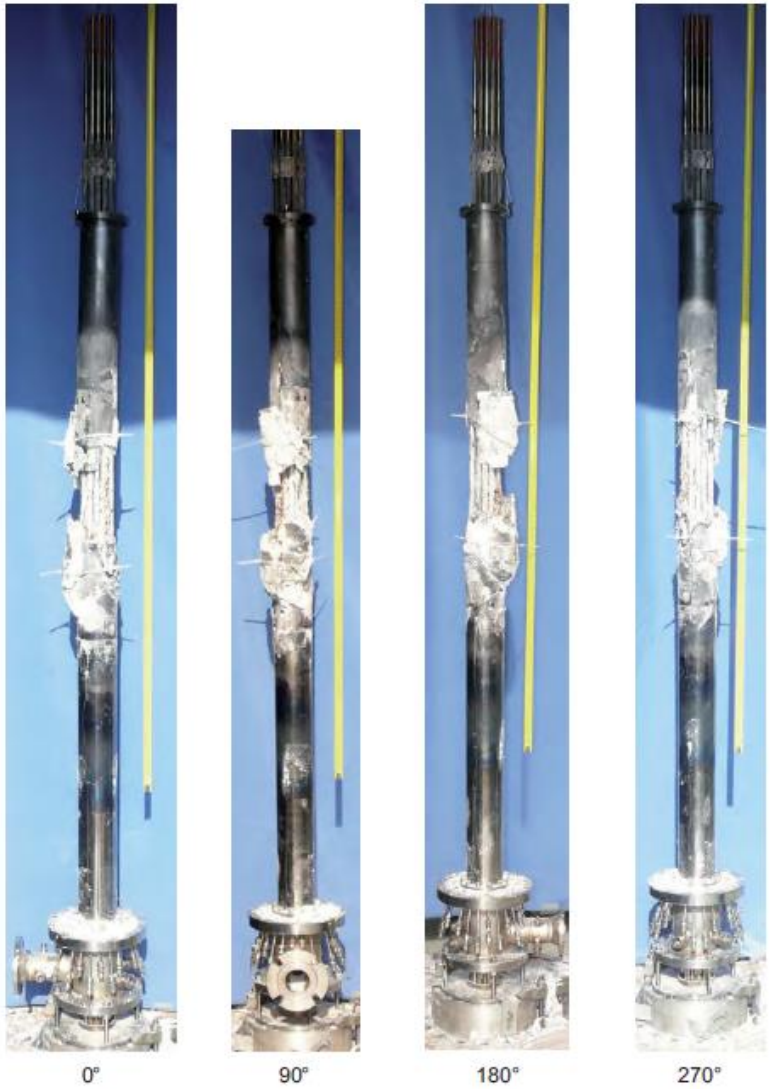
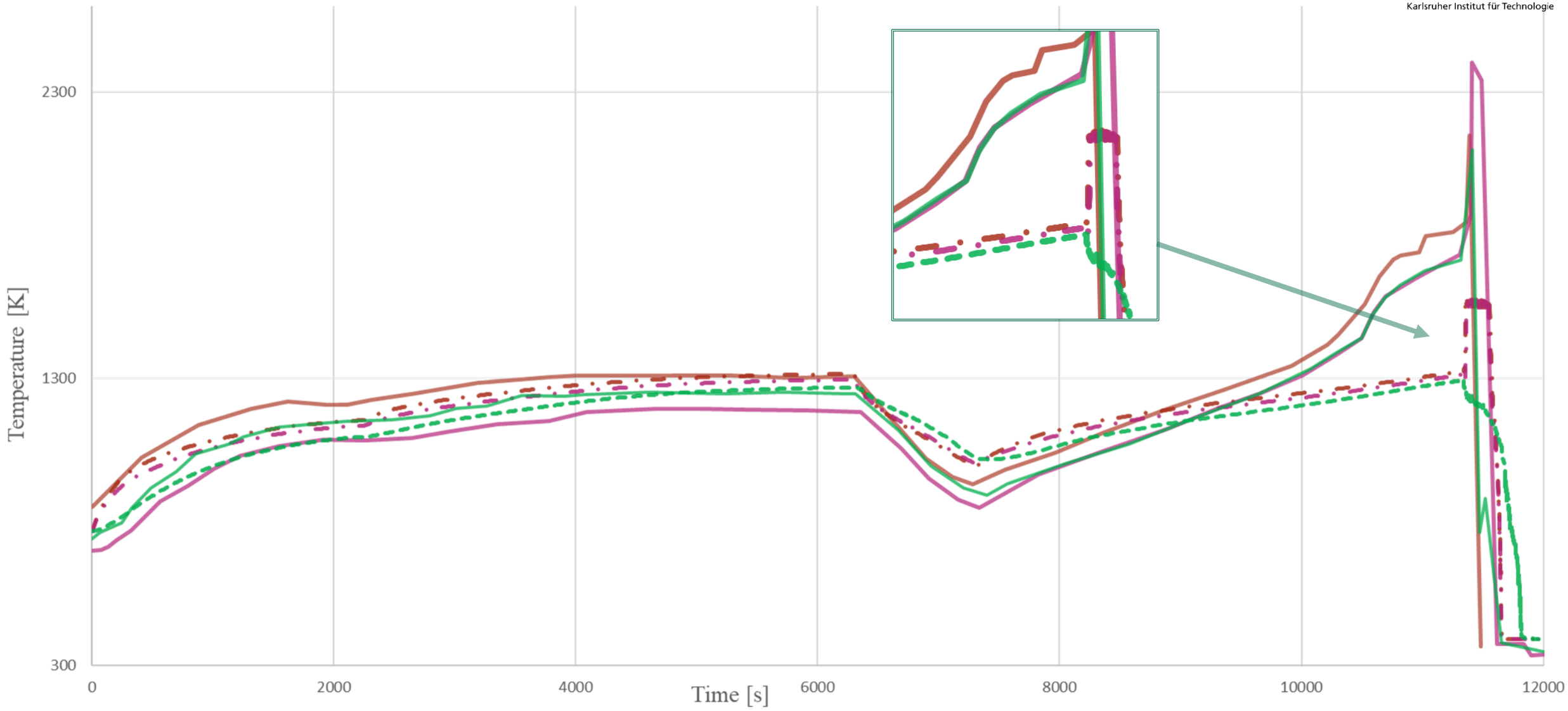
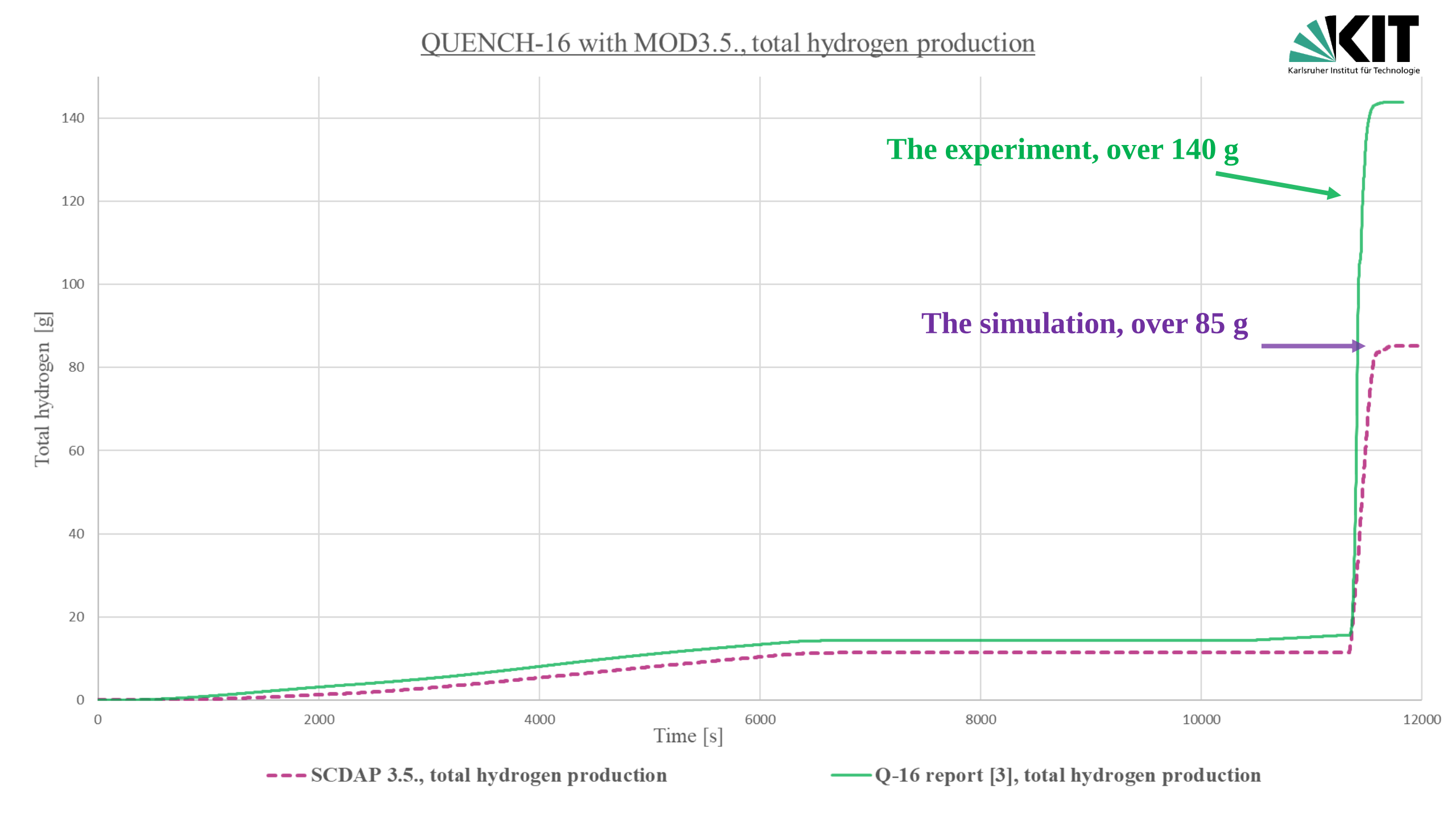


Fig. QUENCH-16 bundle withdrawn from cooling jacket. [3]

QUENCH-16 with MOD3.5., temperatures at 700 mm




QUENCH-16 with MOD3.5., total hydrogen production
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The experiment, over 140 g
The simulation, over 85 g
Total hydrogen [g]
Time [s]
0
20
40
60
80
100
120
140
0
2000
4000
6000
8000
10000
12000
SCDAP 3.5., total hydrogen production
Q-16 report [3], total hydrogen production

ALISA: pre-test

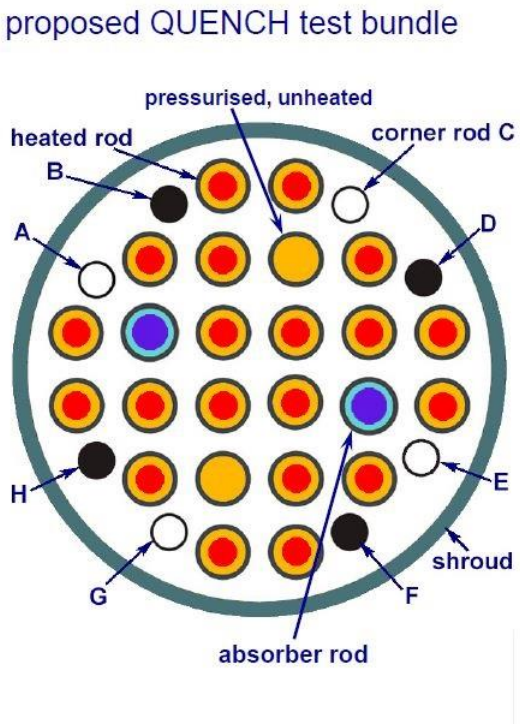


Fig. QUENCH-ALISA bundle composition. [5]

In case of QUENCH-ALISA pre-test calculations we did not assume leakage from the shroud, thus the used **physical model was simpler**, compared to QUENCH-16.

The boundary conditions according to the ***Pre-test simulations of QUENCH-ALISA in the frame of the NUGENIA QUESA project*** [6] presentation from 4th NUGENIA-SARNET TA2.1 Review Meeting.

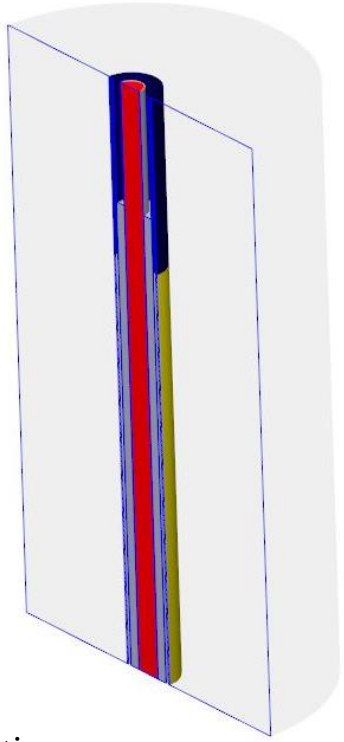


Fig. QUENCH-ALISA model cross-section.

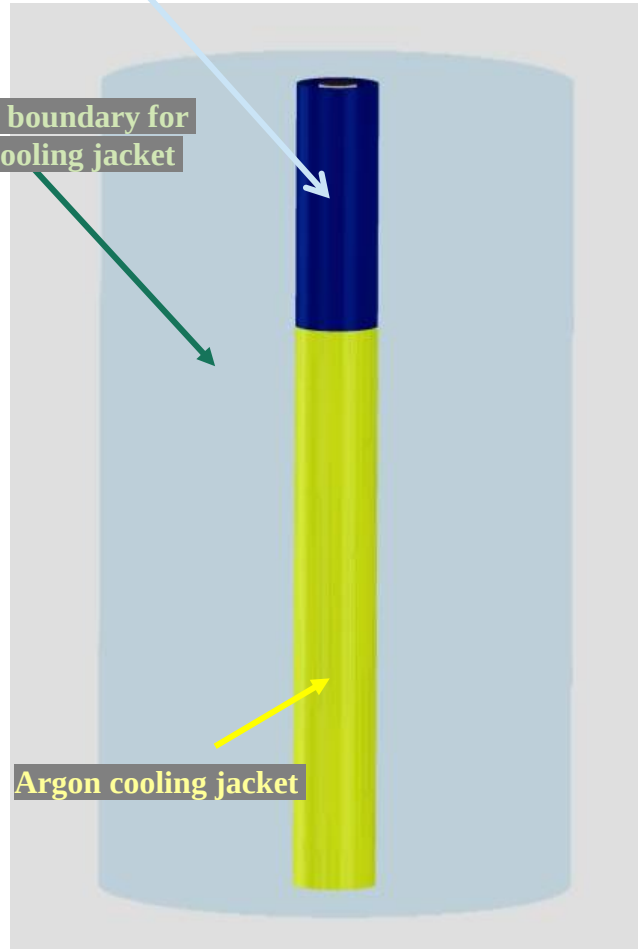
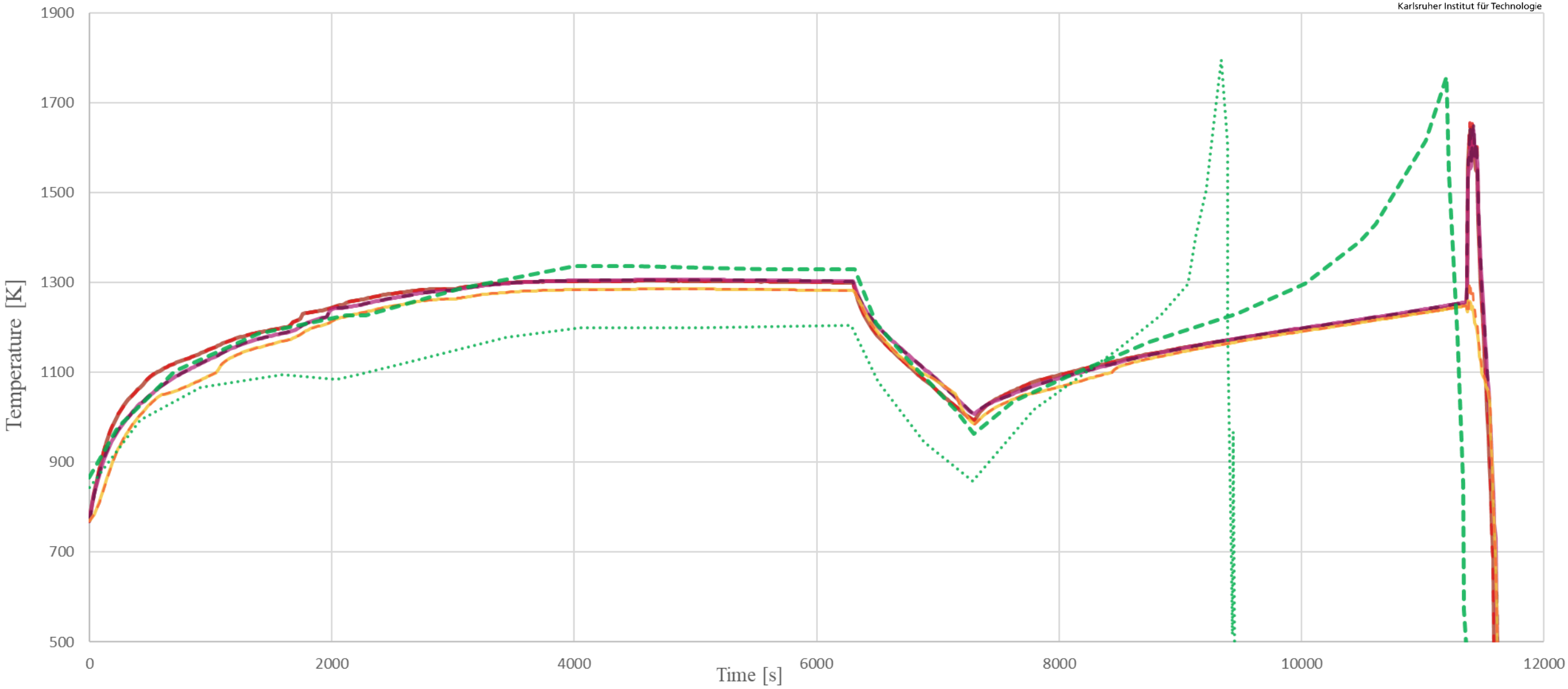


Fig. 3D QUENCH-ALISA model, without sources and sinks.

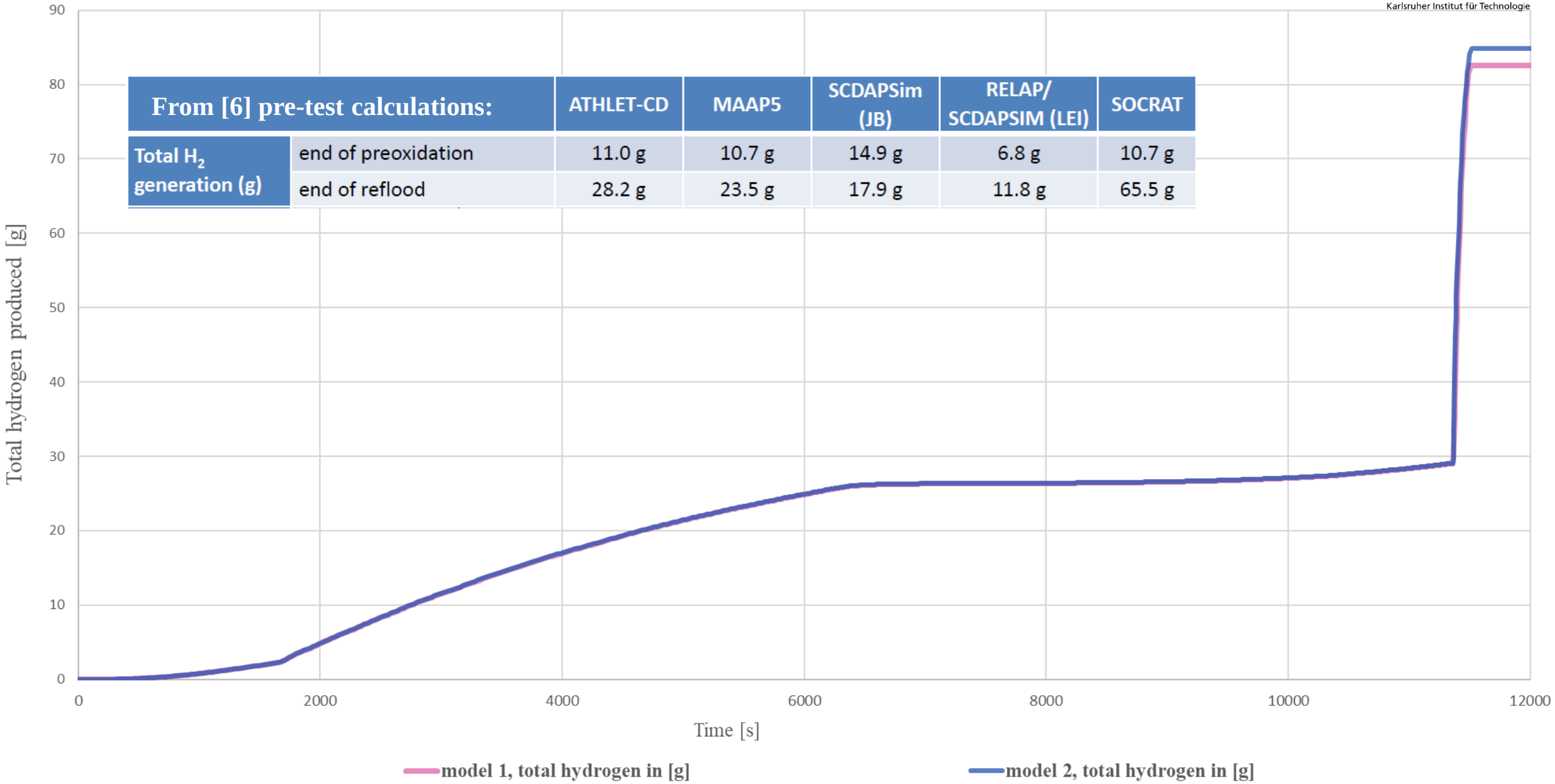
Pre-test ALISA temperatures at 700 mm in MOD3.5. (model 1) and MOD3.5+ (model 2)



Pre-test ALISA in MOD3.5. (model 1) and MOD3.5+ (model 2) - total hydrogen production

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From [6] pre-test calculations:		ATHLET-CD	MAAP5	SCDAPSim (JB)	RELAP/ SCDAPSIM (LEI)	SOCRAT
Total H_2 generation (g)	end of preoxidation	11.0 g	10.7 g	14.9 g	6.8 g	10.7 g
	end of reflood	28.2 g	23.5 g	17.9 g	11.8 g	65.5 g



ALISA: post-test in MOD3.5+

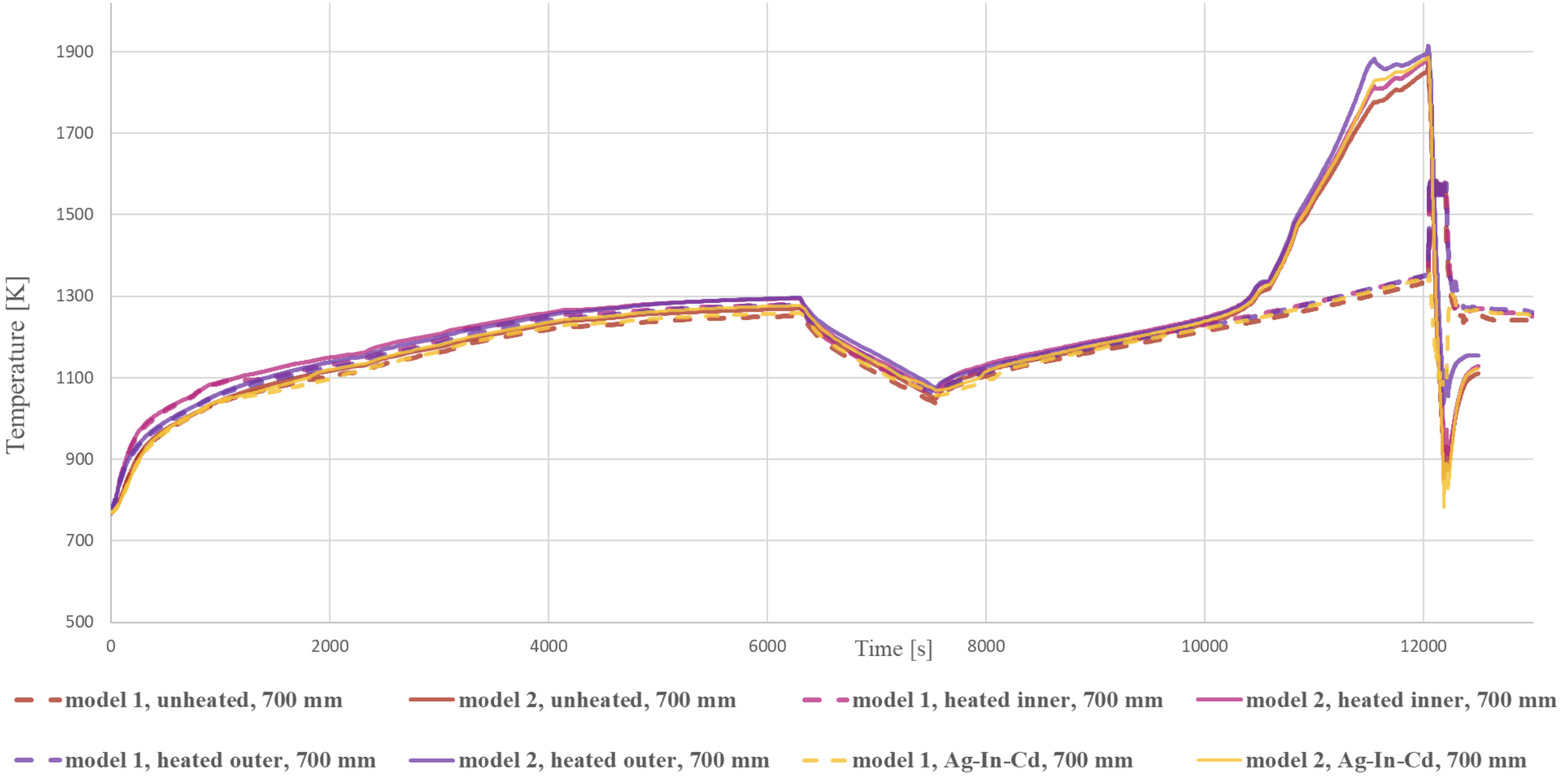
The physical model remains the same, the timeline changes.

The new timeline:

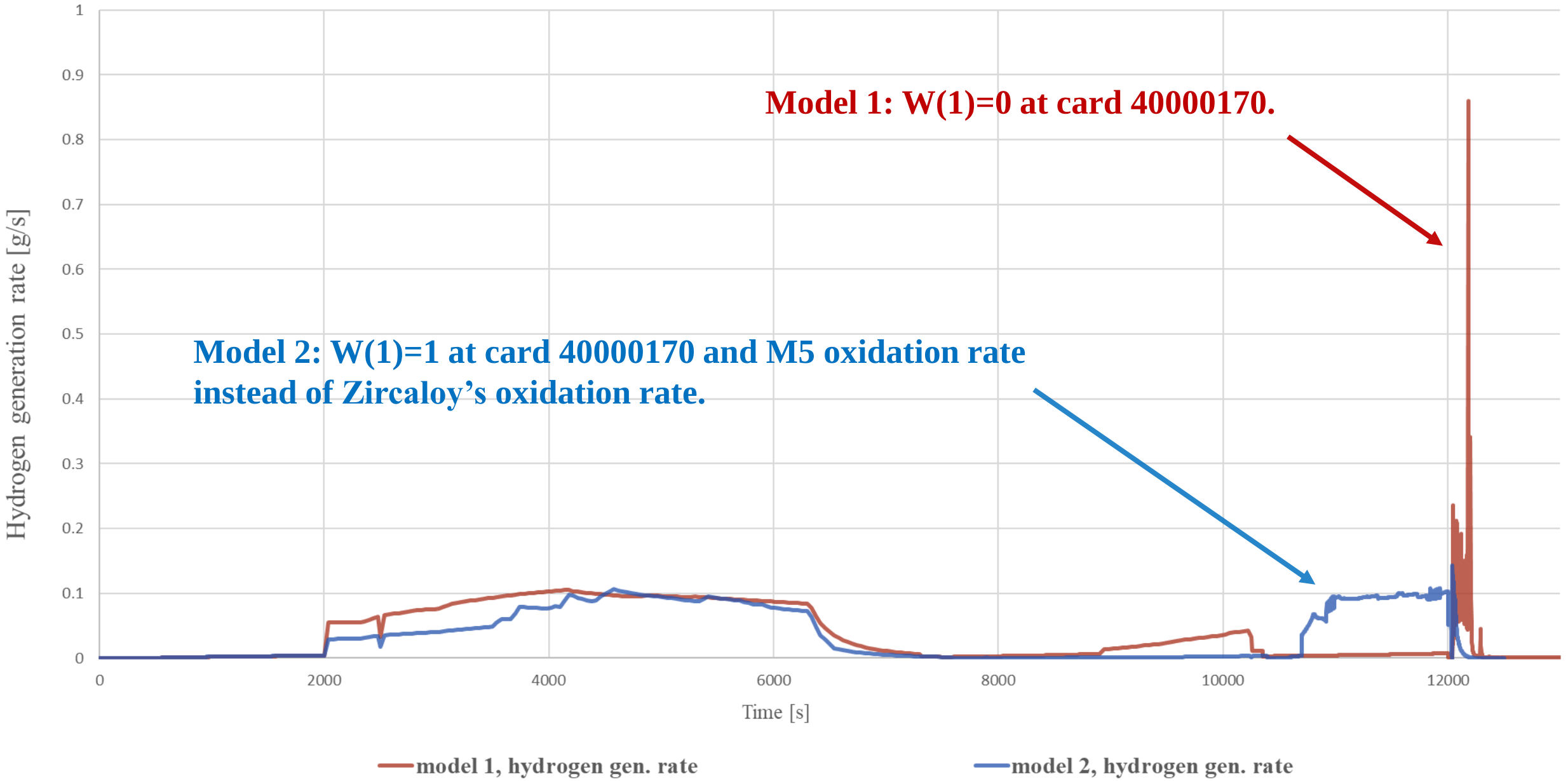
- instead of 7300 s, at 7542 s into the experiment the steam flow is set to 0.3 g/s and argon flow to 1.0 g/s;
- at 7542 s the air flows at 0.2 g/s rate, and thus the air ingress phase begins;
- at 12030 s the quench begins, instead of 11341 s - QUENCH-16 case;
- 12180 s is the end of the quenching phase;
- 12300 s is the end of simulation.

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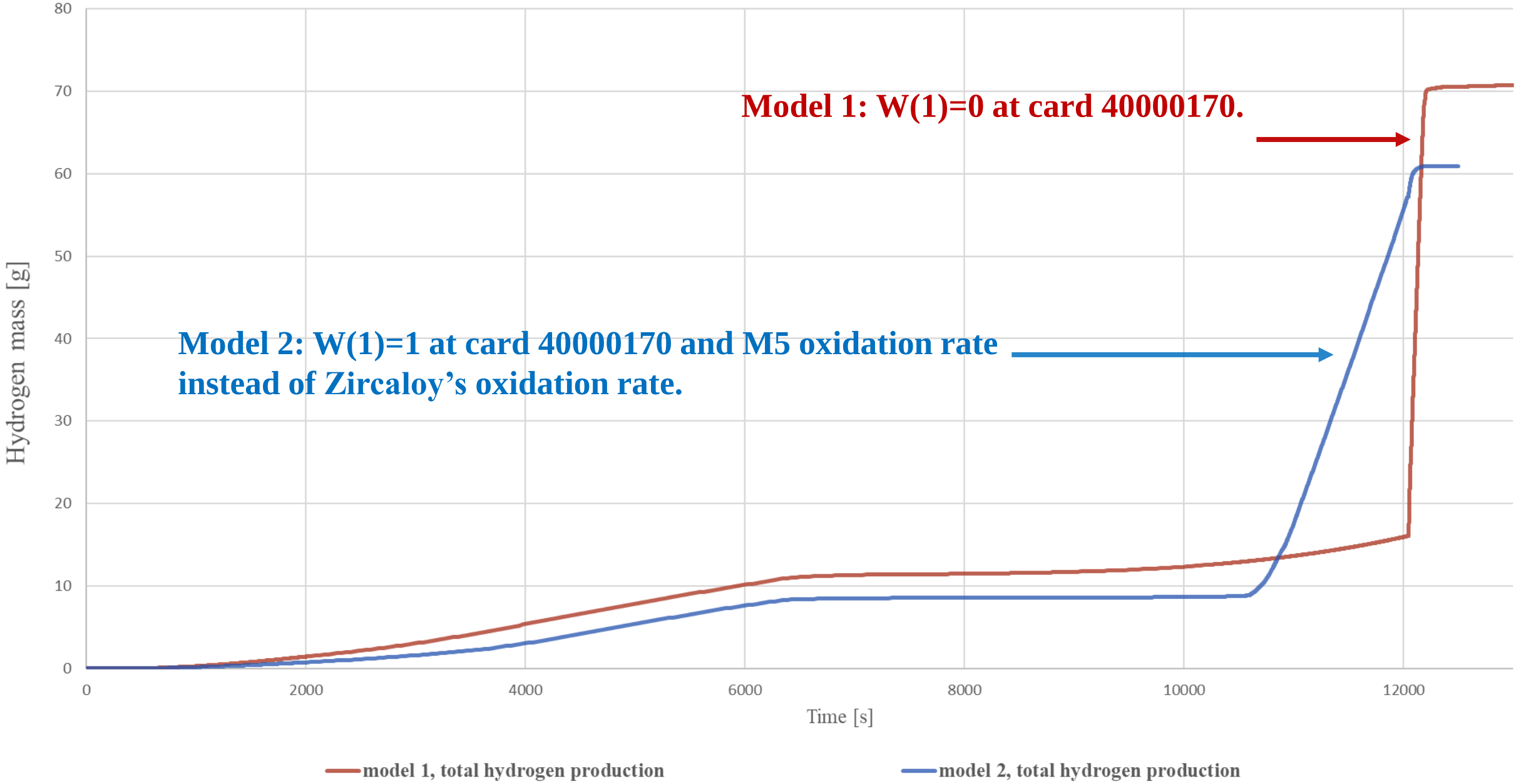
Post-test ALISA in MOD3.5+ at 700 mm, model 1: W(1)=0, model 2: W(1)=1 and M5*



Post-test ALISA in MOD3.5+, model 1: W(1)=0, model 2: W(1)=1 and M5*



Post-test ALISA in MOD3.5+, model 1: W(1)=0, model 2: W(1)=1 and M5*



Summary

- QUENCH-14 simulation results are satisfying. However, it has been the least challenging experiment to model. The M5* oxidation rate in MOD3.5. gives similar peak value of hydrogen generation rate to the experimental value.
- QUENCH-16 is an experiment difficult to model, due to several phenomena. Concerning hydrogen production, the air ingress contributed into 5 g produced in re-oxidation of nitrides in claddings and 2 g in shroud; oxidation of melt has given 19 g of H_2 from claddings and 14 g from shroud. Total contribution of claddings in hydrogen production during reflood was 81g [3], in our simulation we have obtained about 85g.
- The pre-test calculations for QUENCH-ALISA were until 8700s very similar to those presented as "*JB_SCDAPSIM"* in [6]. In our simulation we did not have a temperature escalation during air ingress as in [6]. On the other hand our quench phase has been more pronounced.
- In the post-test calculations for QUENCH-ALISA we have analyzed different oxidation models available in RELAP5/SCDAPSIM. We have used M5* in model 2. The difference between models 1 and 2 is evident. In model 1 quench phase with fast temperature increase may be observed. In model 2 there is a temperature increase in air ingress. The total hydrogen production is bigger for model 1, for model 2 it is about 10 g smaller.

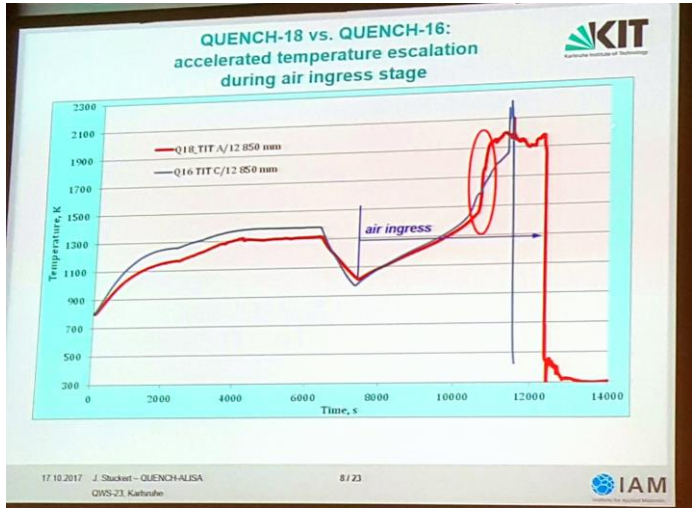


Fig. Photo taken yesterday (17.10.2017) at the Quench Workshop; Presentation by dr Juri Stuckert

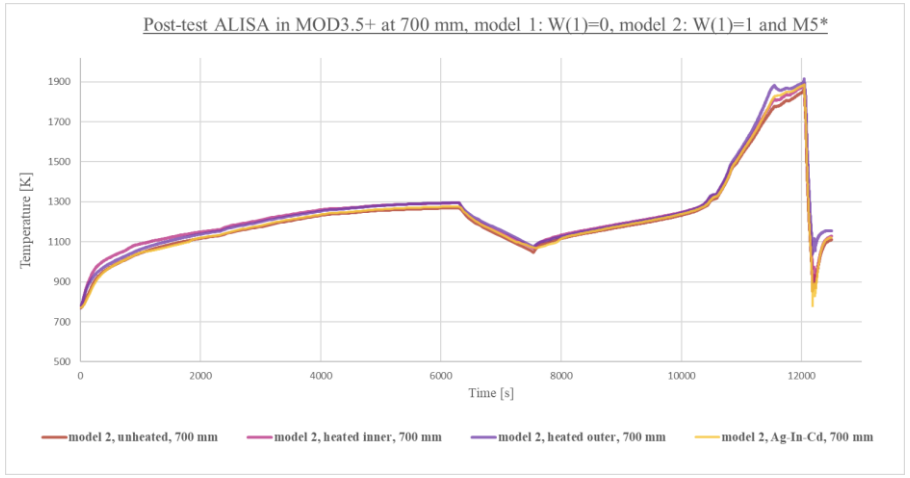


Fig. Our post-test QUENCH-ALISA results with model 2.

Conclusions

- RELAP5/SCDAPSIM model for electrically heated rod is a good tool to model QUENCH experiments. At the very beginning, we have compared the results between simulations using resistance calculated by the code and from the table, and those results were almost identical.
- RELAP5/SCDAPSIM has given very good results for QUENCH-14. In case of air ingress tests: QUENCH-16 and QUENCH-ALISA, the complex reactions between nitrogen and Zircaloy need be introduced to the code, and for that reason nitrogen should be separated from air.
- The chemical reaction of Zircaloy with nitrogen, as described in [8], is an exothermic reaction, where Δh is about 4.056×10^6 J/kg$_{Zr}$. This value translates into such an example: if 6.79% of Zr present in QUENCH-16 bundle reacted with N_2, then the power difference between QUENCH-16 and QUENCH-14, since 8000 s till the quench, would be compensated.

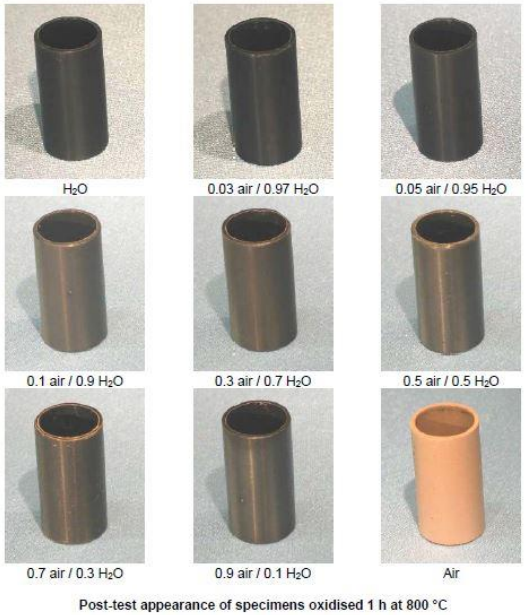


Fig. Source: [4]



Fig. Source: [4]

Conclusions

- There is a need for a quantification of nitride formation during air ingress and energy release due to that process.
- In QUENCH-16 simulation we did not obtain the temperature measured in the experiment on the shroud. Our shroud (Zircaloy) did not melt, thus its contribution to the hydrogen production was smaller. Concerning hydrogen production from claddings, our result (85g) is very close to the one mentioned in the report (81g) [3].
- Modelling of QUENCH-18 seems less challenging than QUENCH-16, considering the quenching phase. It may be a result of steam being injected together with air in air ingress phase in ALISA.
- QUENCH-ALISA post-test and pre-test differ notably, due to different timeline. In post-test calculations we managed to apply M5* oxidation rate (model 2). Temperatures from model 2 reach higher values in air ingress phase than those from model 1.

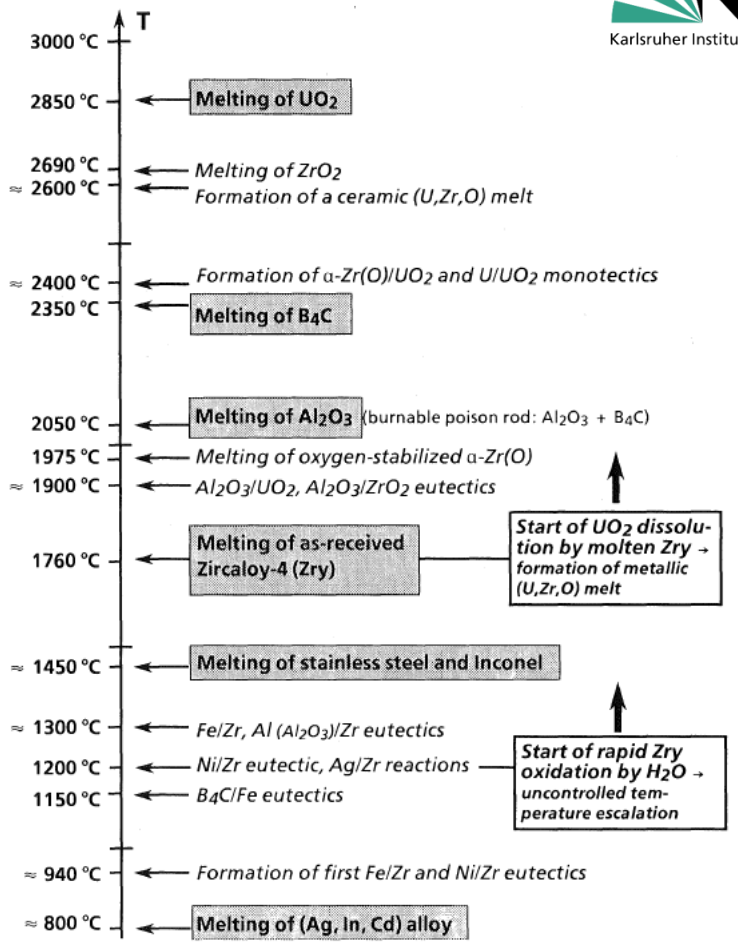


Fig. Chemical interactions and formation of liquid phases in a LWR fuel rod bundle with increasing temperature. [7]

Future plans

Applying all the phenomena observed in QUENCH-16 to RELAP5/SCDAPSIM:

- nitridation;
- re-oxidation of the nitrides;
- melt oxidation.

References:

[1] J. Stuckert, U. Stegmaier, M. Steinbrück, Results of Severe Fuel Damage Experiment QUENCH-14 with Advanced Rod Cladding M5®, KIT Scientific Report 7549, KIT Scientific Publishing, 2010

[2] M. Grosse, Comparison of the high-temperature steam oxidation kinetics of advanced cladding materials, Nuclear Technology vol. 170, 272-279, 2010

[3] J. Stuckert, M. Grosse, Z. Hozer, M. Steinbrück, Results of the QUENCH-16 Bundle Experiment on Air Ingress, KIT Scientific Report 7634, KIT Scientific Publishing, 2013

[4] M. Steinbrück, U. Stegmaier, T. Ziegler, Prototypical Experiments on Air Oxidation of Zircaloy-4 at High Temperatures, FZKA 7257, Forschungszentrum Karlsruhe in der Helmholtz-Gemeinschaft, 2007

References:

[5] J. Stuckert, QUENCH bundle tests in the framework of the SAFEST and ALISA projects, 4th NUGENIA-SARNET TA2.1 Review Meeting, March 2 – 3, 2017

[6] T. Hollands, Pre-test simulations of QUENCH-ALISA in the frame of the NUGENIA QUESA project, 4th NUGENIA-SARNET TA2.1 Review Meeting, March 2 – 3, 2017

[7] P. Hofmann, S. Hagen, G. Schanz, A. Skokan, Chemical Interactions of Reactor Core Materials Up to Very High Temperatures, KfK 4485, Kernforschungszentrum Karlsruhe, January 1989

[8] C. Bals, T. Hollands, New Results of Post-test Calculation of Quench-16 with ATHLET-CD, June 14, 2016, Budapest

Thank You for Your attention.

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Pre-test calculations of CODEX-AIT3 with ATHLET-CD

C. Bals, T. Hollands
Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH, Germany

Abstract

As the behaviour of heated rods in a steam-air environment has been up to now not extensively investigated, the conduct of an experiment with a steam-air mixture is planned in the CODEX facility at MTA EK within the SAFEST project. This future CODEX-AIT3 test, which will be conducted in cooperation with KIT, has the primary objective to investigate the reaction of pre-oxidized Zr-1%Nb (E110G) claddings in a mixed flow of steam and air. It is intended to obtain a similar scenario as in the previous air ingress test QUENCH-ALISA where during the air ingress phase an air and steam mixture with a flow rate defined by pre-test calculations should be injected into the heated bundle up to a maximum temperature of 1600 °C. During the duration of about 1 hour for this phase oxygen and steam starvation should be reached to get information about the reaction of nitrogen with the pre-oxidized E110G-cladding. Different to the test QUENCH ALISA, where the bundle was quenched by water, no further temperature excursion is expected during the cool down by argon.

Concerning geometry and materials the rod bundle is constructed corresponding to the bundle design of VVER-440 reactors and should contain 6 electrical heated rods and 1 central unheated rod with Zr-1%Nb claddings. In the CODEX facility the bundle is surrounded by a hexagonal shroud made of Zr-2.5%Nb and an insulation composed of different materials with defined properties.

To support the experimenters in the definition of boundary conditions pre-test calculations with the code ATHLET-CD were performed. To verify the basic input data deck for the CODEX facility post-test calculations of the tests CODEX-2, LOCA E4 and an additional commissioning test, all having the same 7-rod bundle configuration as the planned test AIT3, were performed previously. The same code version as used for post-test calculations with improved results of the QUENCH-16 experiment and the pre-test calculations of QUENCH-ALISA was applied.

Calculations with variation of flow rates show that a higher flow rate of 1 g/s for argon during the pre-oxidation period gives a better agreement of the aimed 100 µm of maximum oxide layer as pre-condition for the later steam and air ingress phase. The assumption of an intermediate cooling phase with a reduced power resulting in a temperature reduction to ~900 °C helps to get a slow temperature increase after start of air and steam injection. With a constant power of 800 W during the air ingress period and an argon flow of 0.1 g/s, a steam flow rate of 0.02 g/s and an air flow rate of 0.05 g/s can be chosen to meet the aimed conditions

of oxygen and steam starvation. With these conditions the air ingress phase with a temperature increase up to a maximum value of 1600°C has the duration of 3224 s which is close to the aspired period of 1 hour. After power shut-off and stop of steam and air injection the argon flow rate of 2 g/s cools down the bundle to ~400 °C within a period of ~2800 s. However, there is a great influence of the external electrical resistance on the course of the temperatures, so that within the uncertainty of this input value the simulations show a wide band of variation for the temperature behaviour.


Karlsruher Institut für

Pre-test Calculations for Test CODEX-AIT3 with ATHLET-CD

Christine Bals, Thorsten Hollands

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH,
Germany

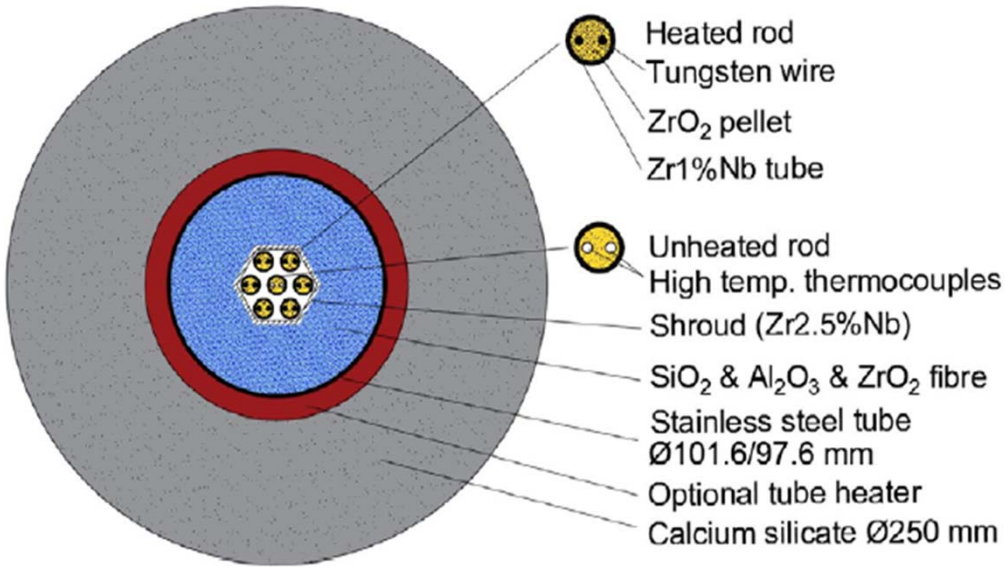
23. International QUENCH-Workshop,
KIT Karlsruhe

Content

- CODEX-AIT3 specification
  - Bundle configuration
  - Experimental parameters
- Modelling with ATHLET-CD
  - Input model
  - Modelling options and used code version
- Verification with CODEX post-test simulations of
  - CODEX-2
  - LOCA E4
  - Commissioning test (AIT3)
- Results of pre-test calculations for test AIT3
- Summary

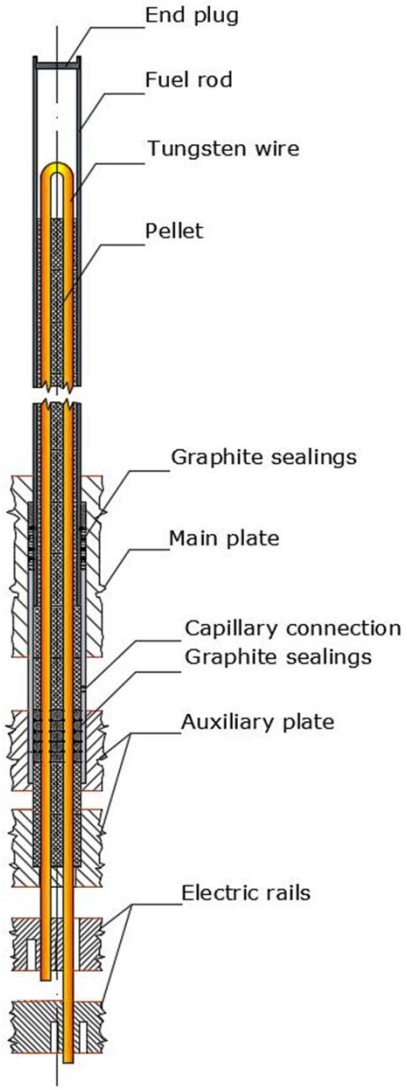
CODEX-AIT3 specification (1)

- **Bundle configuration**
  - 6 heated rods
    with E110G claddings (Zr-1%Nb)
  - 1 central unheated rod
    with E110G cladding (Zr-1%Nb)
  - 3 spacer grids (Zr-1%Nb)
    assumed at 300-600-900 mm
  - Hexagonal shroud (Zr-2.5%Nb)
  - Inner insulation – SS tube –
    outer insulation
    (material properties of insulation
    were defined)



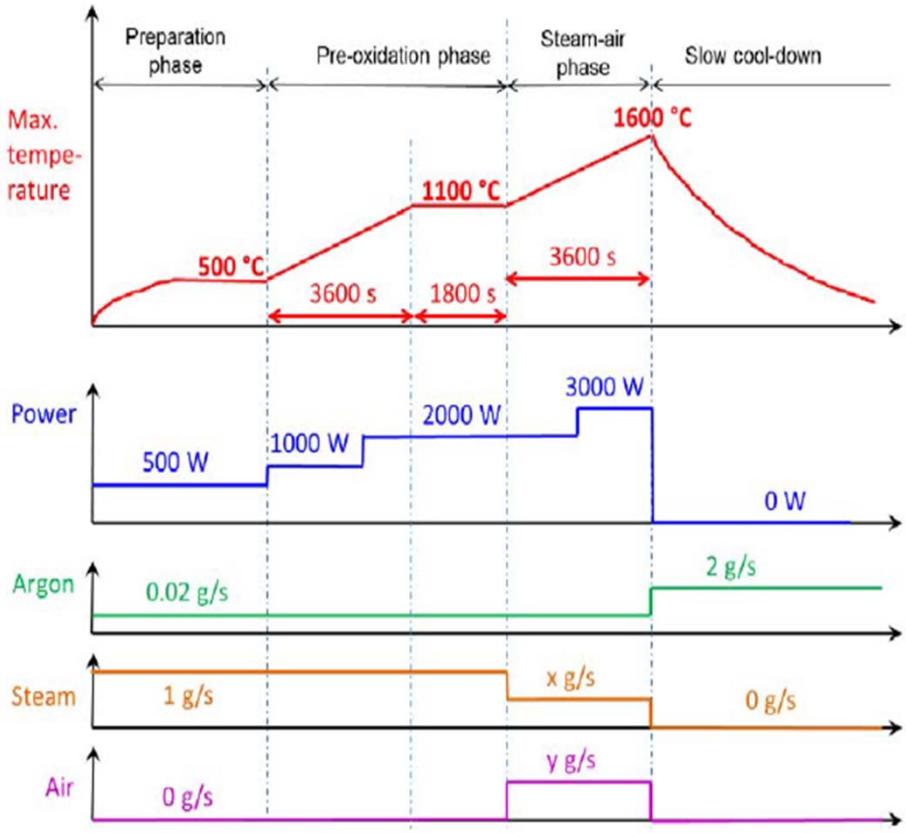
CODEX-AIT3 specification (2)

- **Heated fuel rod**
  - 6 heated rods with tungsten heaters
  - 2 wires with 1.86 mm diameter
  - heated length 2 x 980 mm
  - ZrO_2 pellets
  - total length of rod 1000 mm
    (20 mm unheated length at top of rod)
  - no upper plug
  - lower plug with Cu connections



CODEX-AIT3 specification (3)

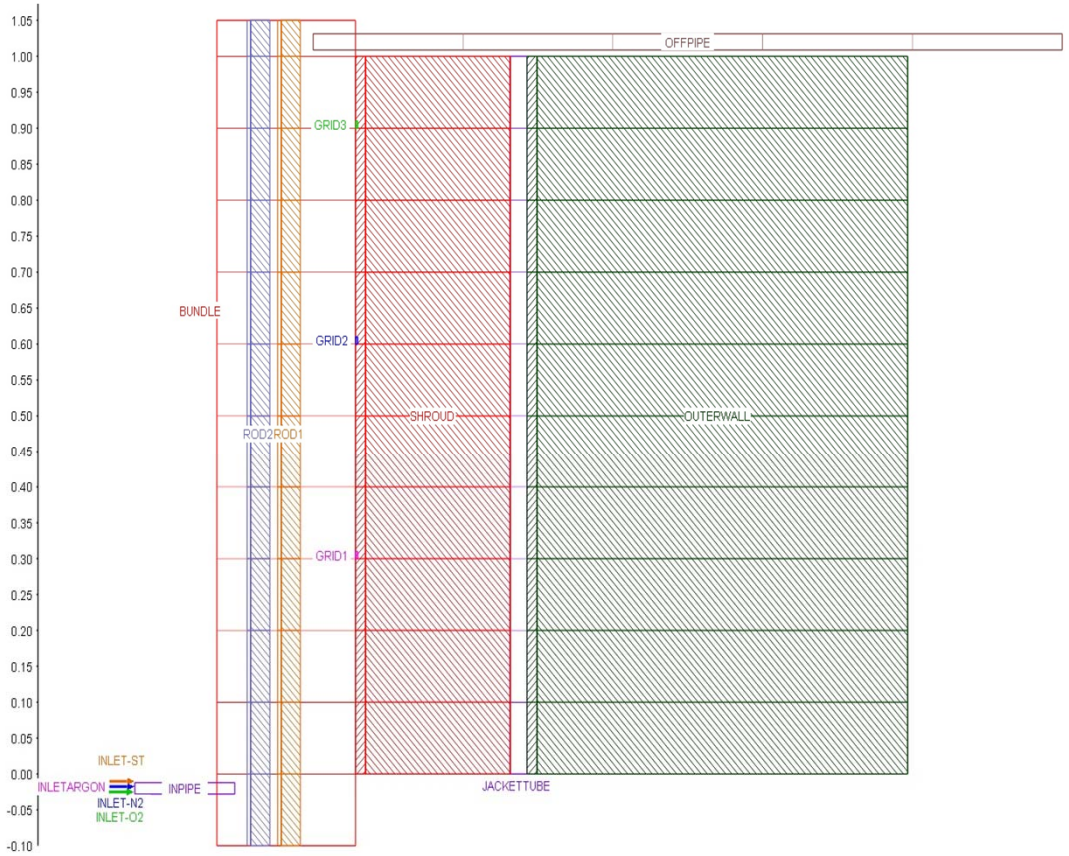
- **Proposed experimental parameters:**
  - Preparation phase:
    heat-up with 500 W power in argon and steam → stabilisation at 500 °C
  - Pre-oxidation phase:
    heat-up with 1000 W – 2000 W power in argon and steam (Δt=3600 s), stabilisation at 1100 °C (Δt=1800 s), max. oxide layer ~ 100 µm
  - Steam-air phase:
    heat-up to max. 1600°C in air + steam flow (with argon);
    aims: duration ~ 3600 s, select flow rates to get oxygen and steam starvation;
  - Cool-down phase:
    argon flow of 2 g/s with stopped power and finished air + steam supply



from
Z. Hózer, et al.,
Specification of CODEX air ingress test in the framework of the EU SAFEST project, MTA EK-FRL-2016-448-1-1-M1

Input model for CODEX-AIT3 pre-test calculation with ATHLET-CD (1)

- **Fluid and heat objects:**
  - BUNDLE with rods,
    12 axial nodes
    heated region 10 nodes
    between 0.0 and 0.98 m
    ROD1: 1 unheated central rod
    ROD2: 6 heated outer rods
  - Inlet and outlet pipe
    (INPIPE, OFFPIPE)
  - JACKETTUBE: gap of 0.3 mm
    to connect object SHROUD
    (shroud - inner insulation) with
    object OUTERWALL
    (SS tube – outer insulation)
- **Fills:**
  - STFILL: injection of steam
  - ARFILL: injection of argon
  - O2FILL, N2FILL, STFILL
    injection of air + steam
  - ARFILLC: cool down with argon

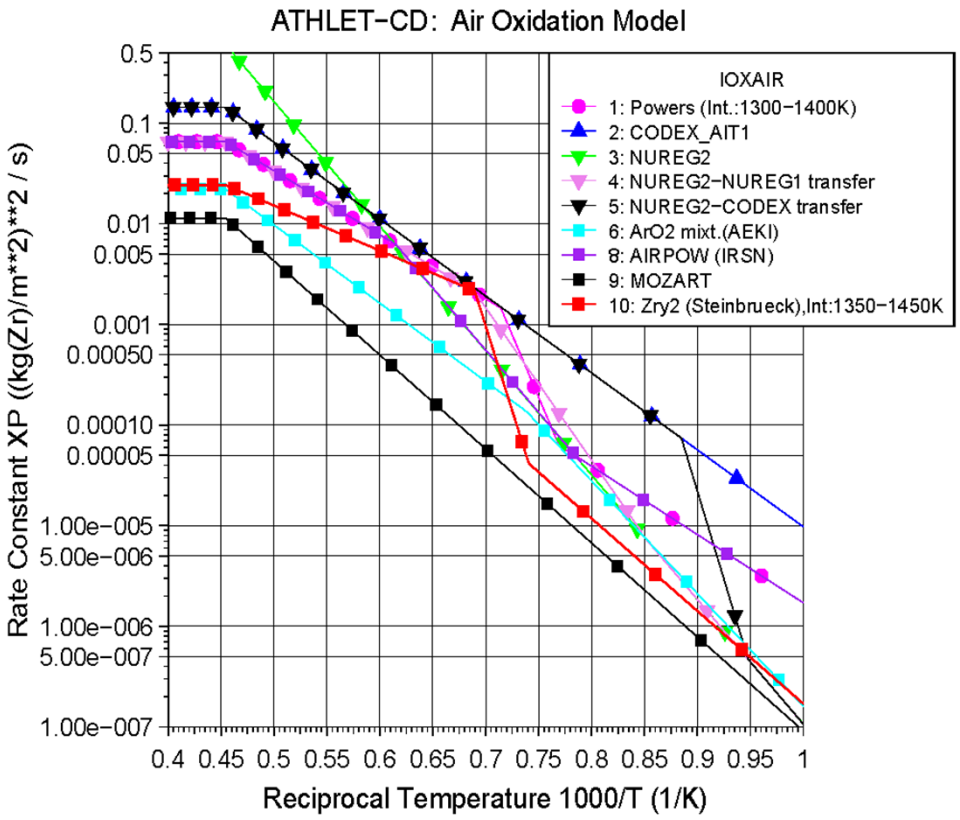


Options used in the ATHLET-CD Air Ingress Model: Oxidation

Options as used in QUENCH-16 simulation

- **IOXAIR=10**
  with interpolation at 1350-1450 K
  (Steinbrück, red curve)
- **ROXLMA= 1**
  oxide layer thickness for transition from parabolic rate to linear rate;
  (ROXLMA=1: no transition to linear rate);
- **OXO2LIM=0.05**
  the calculated oxidation rate will be reduced (→ 0) if relative availability of O_2
  $g(p_{O2})$ < OXO2LIM
  → start of starvation condition
- **FLIMOX=1.0** for rods
  (multiplier for oxidation rate),

  reduced reaction for HECU-components:
  FLIMOX=0.1 for shroud,
  FLIMOX=0.0 for grid spacers
  (not considered for air oxidation)

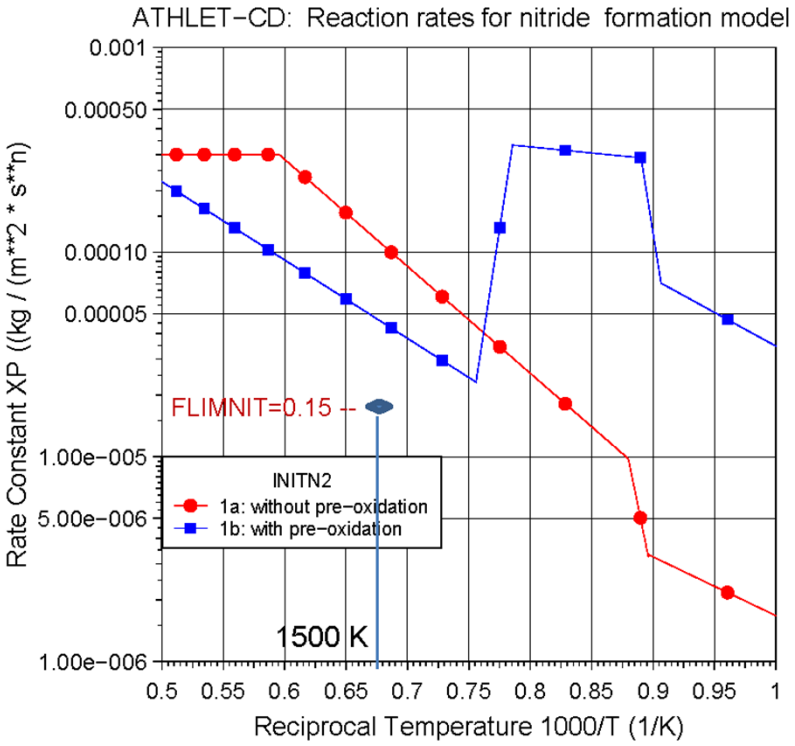


Options used in the ATHLET-CD Air Ingress Model: Nitride Formation

Options as used in QUENCH-16 simulation:

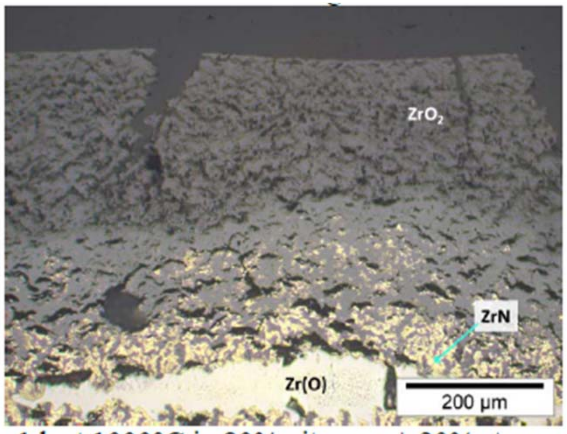
- **INITN2=1a** → red curve
  Hollands correlation, without pre-oxidation,
- **FLIMNIT=0.15** for rods,
  multiplier to reaction rate for nitride formation,
  reduction factor derived from QU-16

  reduced reaction for HECU-components:
  FLIMNIT=0.1 for shroud,
  FLIMNIT=0.0 for grid spacers
  (not considered for nitride formation)
- **NITN2LIM=0.1**
  the calculated nitride formation
  will be reduced (→ 0) if relative availability of N_2
  $g(p_{N2})$ < NITN2LIM,
  → start of starvation condition

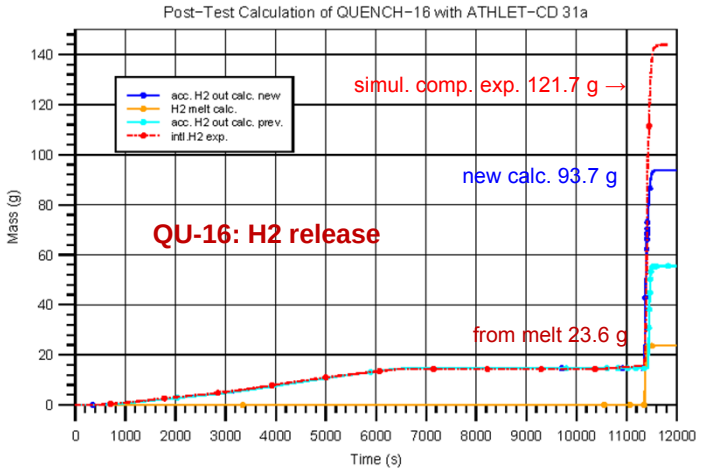


Code version for pre-test calculation of AIT3 with ATHLET-CD

- Use of code version 3.1a with the following changes for air ingress model:
  - ✓ linear kinetics for steam oxidation after nitride formation for rods (ECORE) and shroud (HECU)
  - ✓ change in the modelling of nitride formation: reaction to ZrN is possible after O_2 starvation also in presence of steam based on literature (1)
  - ✓ as previously: no reaction of steam is possible as long as O_2 is available!
- Basis is improved post-test simulation of **QUENCH-16** (2)



1 h at 1000°C in 80% nitrogen + 20% steam



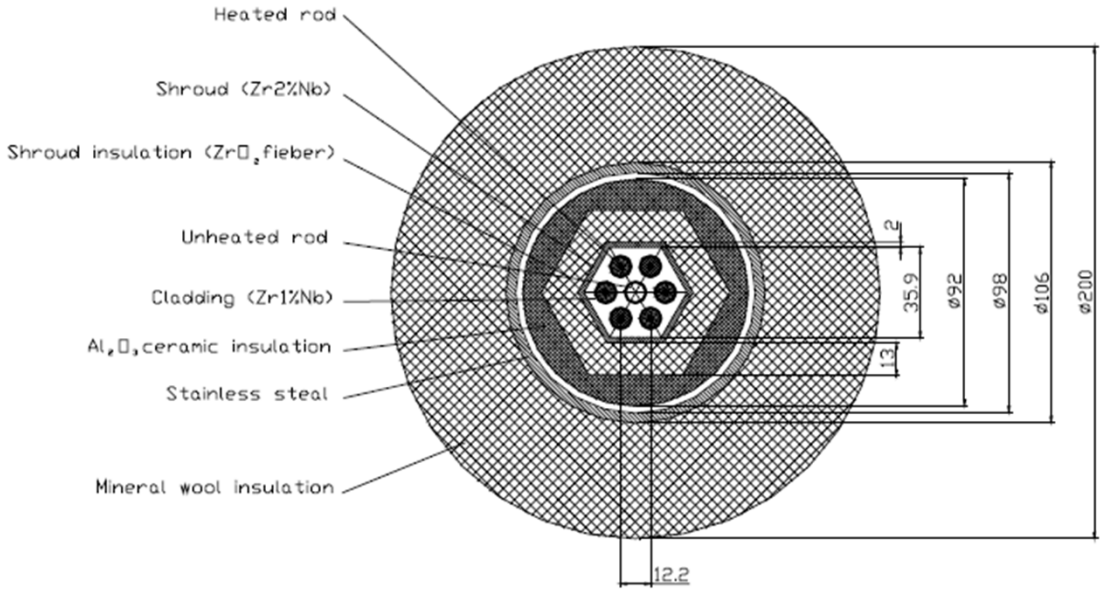
(1) M. Steinbrück, M. Grosse, F. Oliveira da Silva, Oxidation of Zircaloy-4 in Steam-Nitrogen Mixtures at 600-1200°C, ICAPP 2016, San Francisco, April 17-20, 2016

(2) C. Bals, T. Hollands, New Results of Post-test Calculation of QUENCH-16 with ATHLET-CD; Seminar on Cladding Oxidation by Air under Severe Accident Conditions in a Nuclear Reactor, June 14, 2016, Budapest

Verification with CODEX post-test simulations: (1)

CODEX-2 experiment

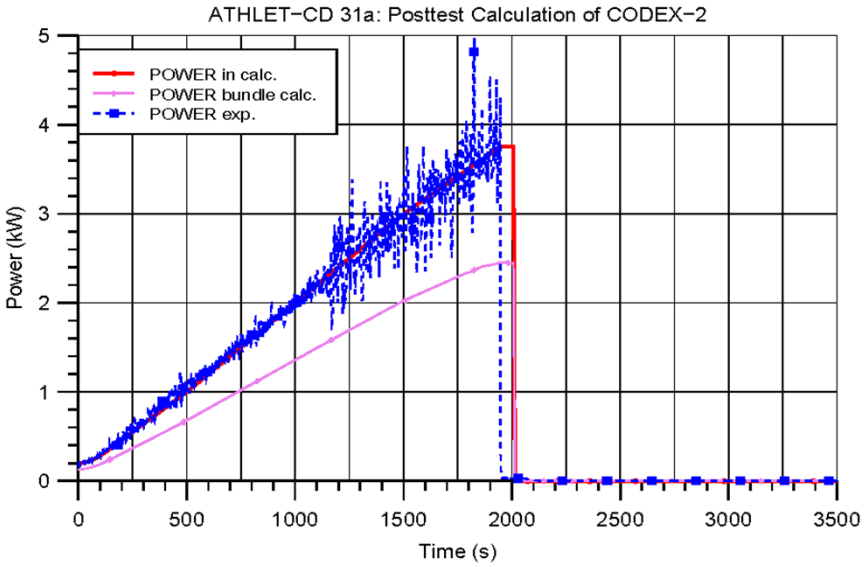
- VVER-440 - 7-rod bundle design, heater 3 mm diameter, UO_2 pellets, unheated central rod, heated length 600 mm
- Pre-heating in argon to ~500°C
  G(Ar) ≈ 1.4 g/s,
  T(Ar) ~ 600°C
- Heat up with linear increasing power in steam and argon
  G(Ar) ≈ 1.4 g/s
  G(steam) ≈ 0.8 – 1.0 g/s,
  T(steam) ~ 600°C
- After temperature escalation power was switched off, steam flow was stopped;
- Slow cool-down with Ar: G(Ar) = 1.4 g/s
- Main results of experiment (1995):
  max. temperature ~ 2000°C,
  H_2 generation: 26.6 g,
  final bundle state: break-up of the bundle in the upper test section



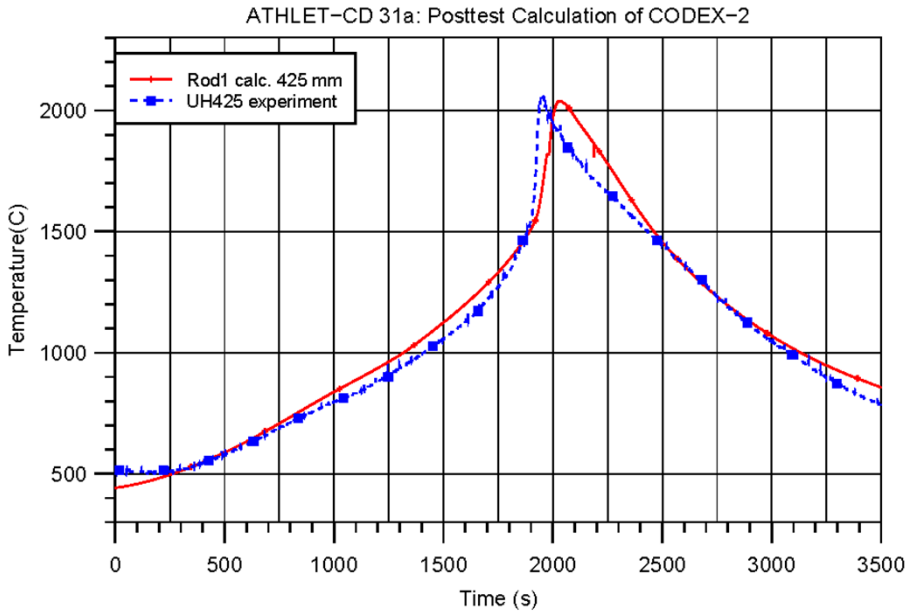
Verification with CODEX post-test simulations: (2)

CODEX-2 simulation

- Post-test simulation with the following options:
  - → WHRES0=0.006 Ω/rod
    external electrical resistance
  - → IOXM=2
    steam oxidation with correlation of Cathcart – Urbanik-Heidrik
  - → ROXLIM=1
    no transfer from parabolic to linear oxidation kinetics (steam);
- ➢ the same values and options were used for the later AIT3 pre-test calculation!
- Result of simulation:
  - → good agreement of temperatures
  - → underestimation of hydrogen generation:
    exp.: 26.6 g
    calc. 10.3 g ?



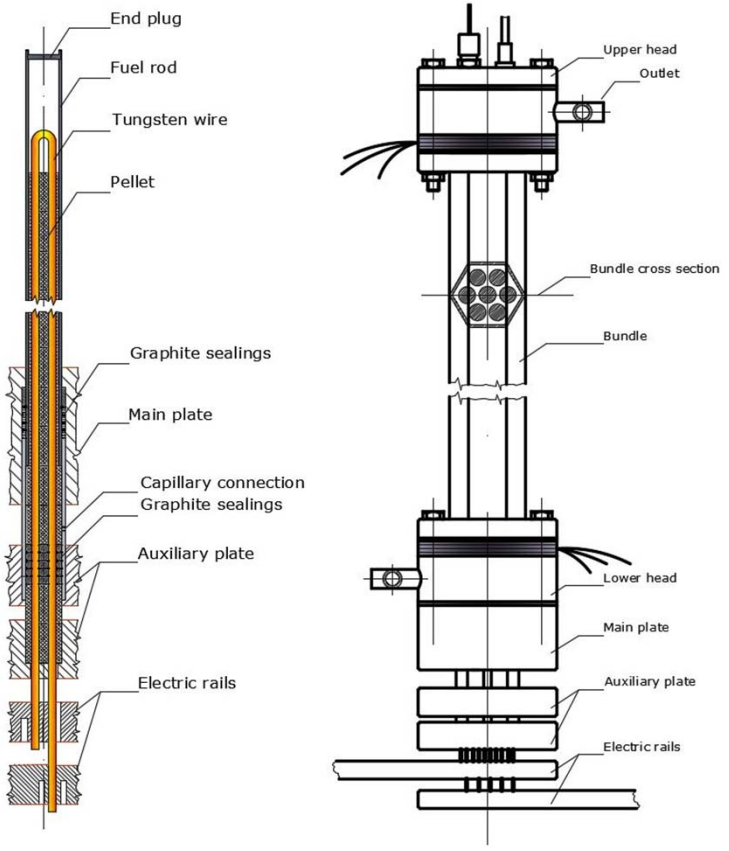
Power of Rods



Rod Temperature at 425 mm

Verification with CODEX post-test simulations: (3)

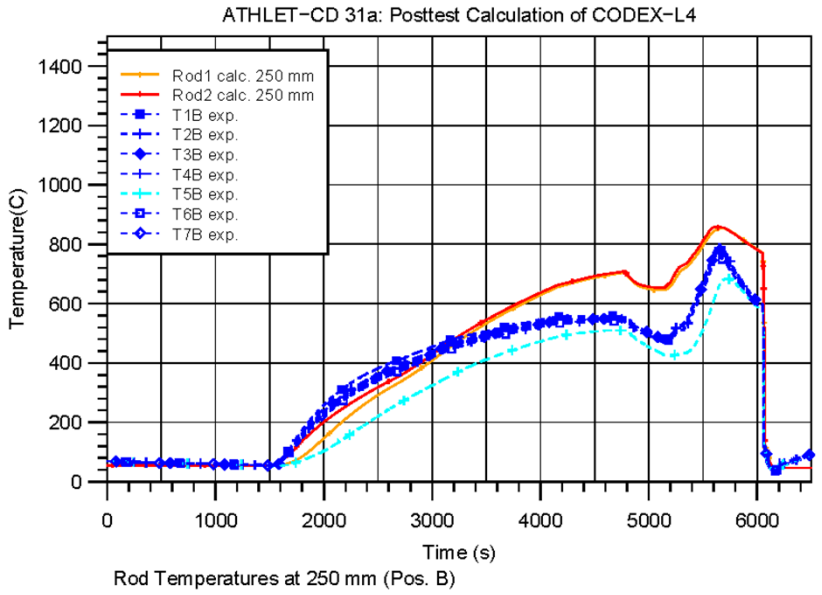
- **LOCA E4 experiment**
  - 7-rod bundle design,
    2 heaters (up and down) with 1.86 mm diameter
    (as planned for AIT3), Al_2O_3 pellets,
    unheated central rod,
    heated length 580 mm
  - Pre-heating in argon
    G(Ar) = 0.02 g/s,
    T(Ar) ~ 600°C
  - Heat up with power of 1.2 kW
    in steam and argon
    G(Ar) ≈ 0.02 g/s
    G(steam) ≈ 0.78 g/s
  - After ~360 s at T_{max} ~1000°C
    power was switched off,
    steam and argon flow were stopped;
  - Fast cool-down with water: G(cool) = 28.75 g/s



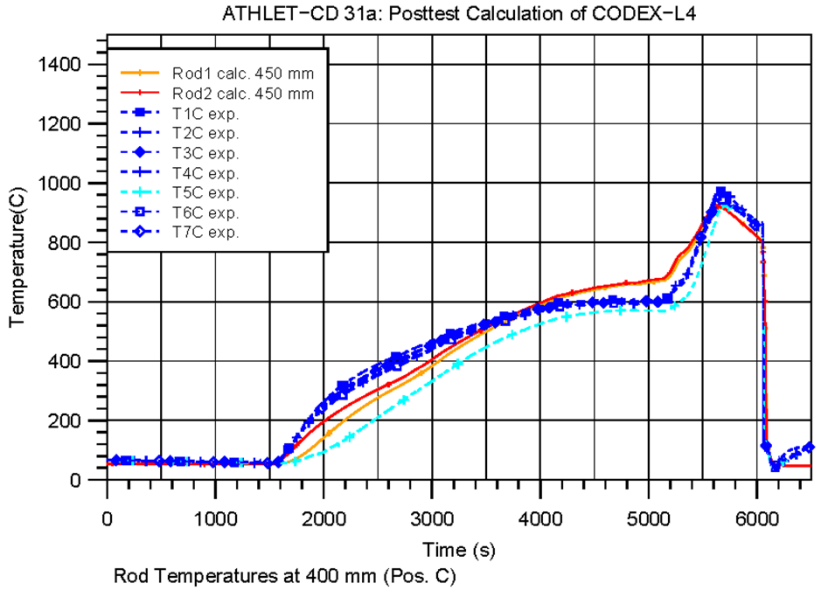
Verification with CODEX post-test simulations: (4)

LOCA E4 simulation

- Post-test simulation with the following options:
  - → WHRES0=0.006 Ω/rod
    external resistance
  - → IOXM=2
    steam oxidation with correlation of
    Cathcart – Urbanik-Heidrik
  - → ROXLIM=1
    no transfer from parabolic to linear
    oxidation kinetics (steam);
  - ➢ the same values and options were used for the later AIT3 pre-test calculation!
- Results of simulation:
  - → temperatures are overestimated in the lower heated region
  - → temperatures are in satisfactory agreement in the upper heated region



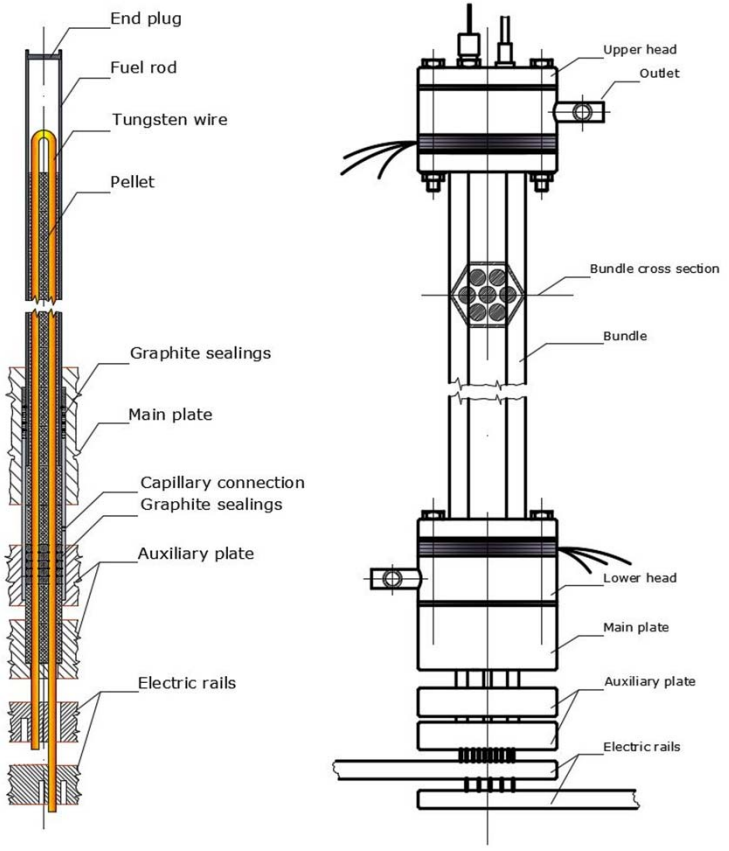
Rod Temperatures at 250 mm (Pos. B)



Rod Temperatures at 400 mm (Pos. C)

Verification with CODEX post-test simulations: (5)

- **Commissioning test (prior to AIT3)**
  - 7-rod bundle design,
    2 heaters (up and down) with 1,86 mm diameter,
    ZrO_2 pellets (as planned for AIT-3),
    heated central rod (different to AIT-3)
    heated length 580 mm
  - Pre-heating in argon
    G(Ar) = 5 l/min = 0.15 g/s,
    T(Ar) = 20°C
  - Heat-up with 2 steps of power: 150 W – 300 W
  - After ~12000 s a max. temperature of ~ 550°C is reached, then the measurement of temperatures was stopped;
  - Different to the planned AIT-3 test:
    - heated central rod
    - external cooling of the shroud with G=0.12 g/s (not simulated);
    - no external calcium silicate insulation (considered in the simulation)



Verification with CODEX post-test simulations: (6)

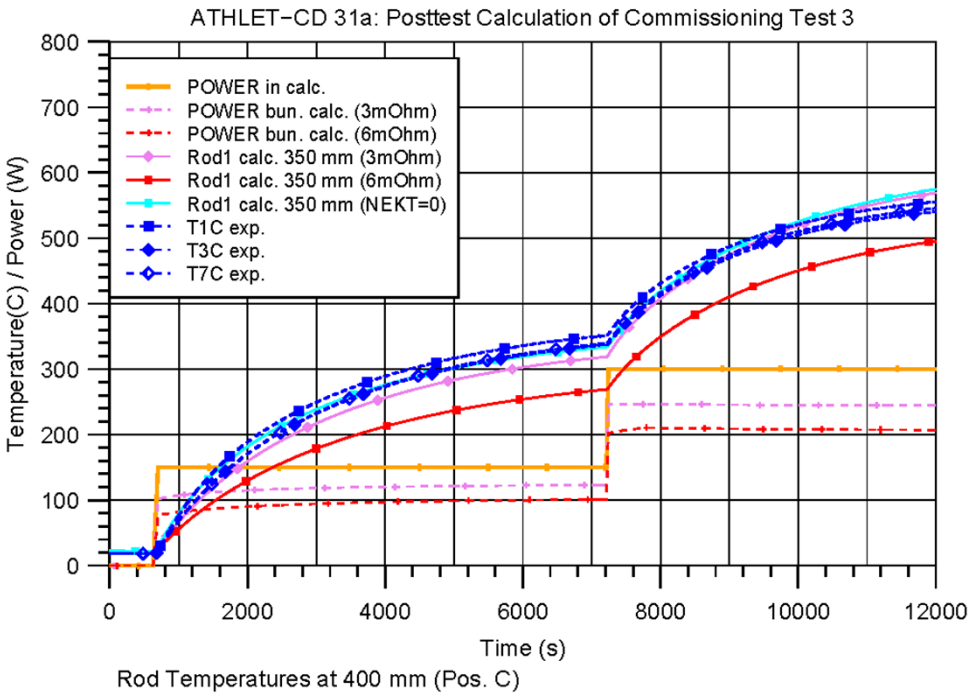
Commissioning test (simulation)

- Post-test simulation with the following options:
  - WHRES0=0.006 Ω/rod
    standard electrical heater rod model used
  - WHRES0=0.003 Ω/rod
    standard electrical heater rod model used
  - direct input of power table with reduced power (-20 % to consider the loss due to external resistance)
- no influence of oxidation and radiation due to low temperatures

- Results of simulation:

→ best agreement of heat-up if standard electrical heater model not used; but: influence of electrical resistance at higher temperatures will not be considered correctly!

→ temperature increase is underestimated with standard electrical heater rod model and the same value for external resistance (WHRES0=0.006 Ω/rod) which lead to good agreement for the calculation of temperatures in the CODEX-2 and LOCA-E4 simulation.



Results of pre-test calculations for test CODEX-AIT3 (1)

Test calculations with changed boundary conditions

Phase	Parameter	Result	Conclusion for reference calculation
Pre-oxidation	G(argon) as given: 0.02 g/s	T_{max} > 1100 °C $\partial_{ox,max}$ > 150 µm	Higher argon flow rate G(argon) = 1.0 g/s
Steam-air-phase	Power as given 2000 W-3000 W	quick heat up to T=1600°C, Δt(air-phase) too short	Lower power P=800 W
	without intermediate cooling	quick heat up to T=1600°C, Δt(air-phase) too short, << 1 h	Intermediate cooling is necessary to get a longer time period of air phase
	G(steam) < 0.02 g/s G(air) < 0.02 g/s	too low mass flow rates should be avoided (measurement!)	G(steam,air) ≥ 0.02 g/s
	G(steam) > 0.02 g/s	no steam starvation	G(steam) = 0.02 g/s
	G(air) >> G(steam)	Steam consumption starts late → no steam starvation	G(air) = 0.05 g/s
	G(argon) as given: 0.02 g/s	quick heat up to T=1600°C, Δt(air-phase) too short	Higher argon flow rate G(argon) = 0.1 g/s

Results of pre-test calculations for test CODEX-AIT3 (2)

- **Boundary conditions for reference calculation:**

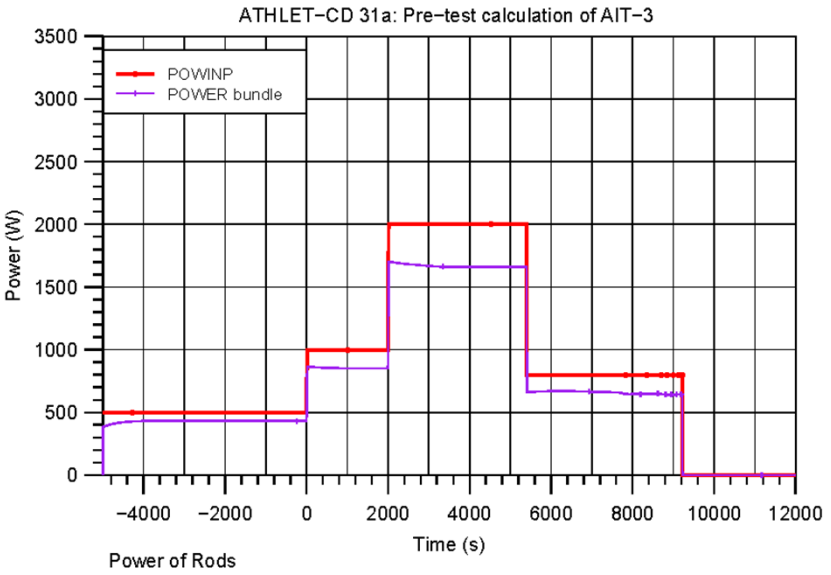
  - Pre-heating (t= - 5000 - 0 s):
    Power 500W
    → T_{max}≈500 – 600 °C

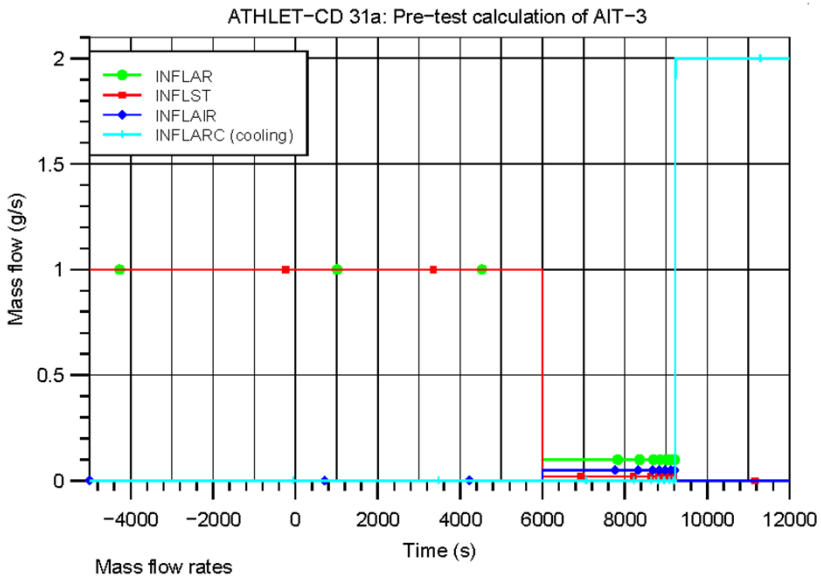
  - Pre-oxidation period (t=0 - 5400 s):
    Power as specified
    t=0 - 2000s: 1000W
    t=2000 - 5400s: 2000W
    G(steam) = 1 g/s (as specified)
    G(argon) = 1 g/s (higher than spec.)
    → T_{max}≈1100°C, $\partial_{ox,max}$ ≈100 µm)

  - Intermediate cooling (t=5400 – 6000 s):
    Power reduction to 800 W
    with steam and argon flow as previous
    → T_{max}≈ 900°C, to avoid too quick increase of temperatures after start of air injection

  - Air-ingress period: constant power of 800 W
    (lower than specified)
    G(steam) = 0.02 g/s
    G(air) = 0.05 g/s
    G(argon) = 0.1 g/s (higher than spec.)
    → to get slower increase of temp. after steam and air starvation (aim: Δt=3600s until T_{max}= 1600°C);

  - Start of cool down phase:
    G(argon) = 2 g/s (as specified)

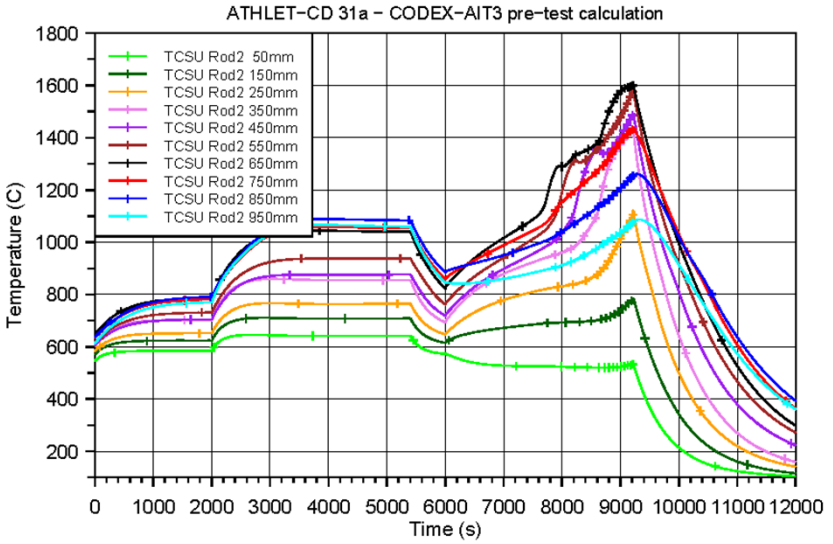




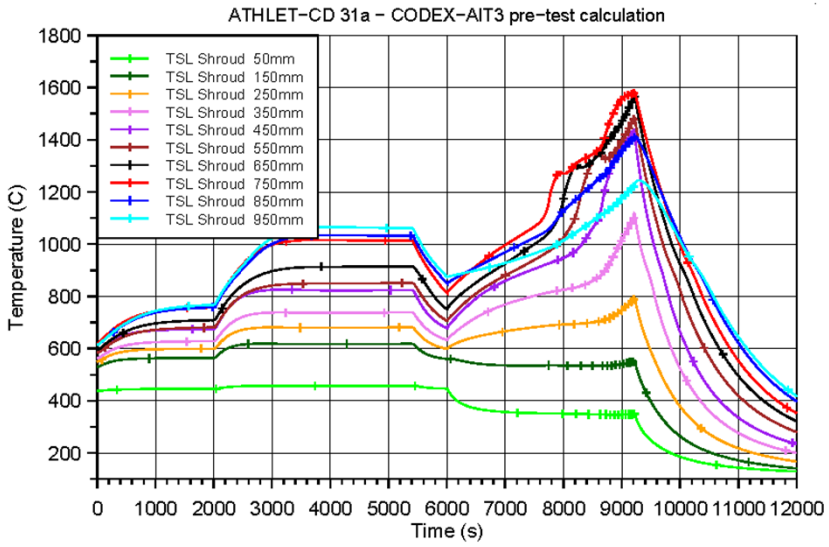
Results of pre-test calculations for test CODEX-AIT3 (3)

- **Boundary conditions for reference calc.:**
  - Start of cool down phase:
    at T_{max}= 1600°C – reached at t= 9224 s
    with power shut off,
    stop of steam and air flow;
    ➢ duration of air ingress phase 3224 s

- **Parameters for reference calculation**
  - standard electrical heater rod model with external resistance WHRES0 = 0.006 Ω/rod,
  - RPELI=1.315 mm volume-equal heater radius for 2 wires with 1.86 mm diameter
  - updated material properties for Tungsten-ZrO_2 mixture with pellet radius 3.825 mm and wire thickness 1.315 mm



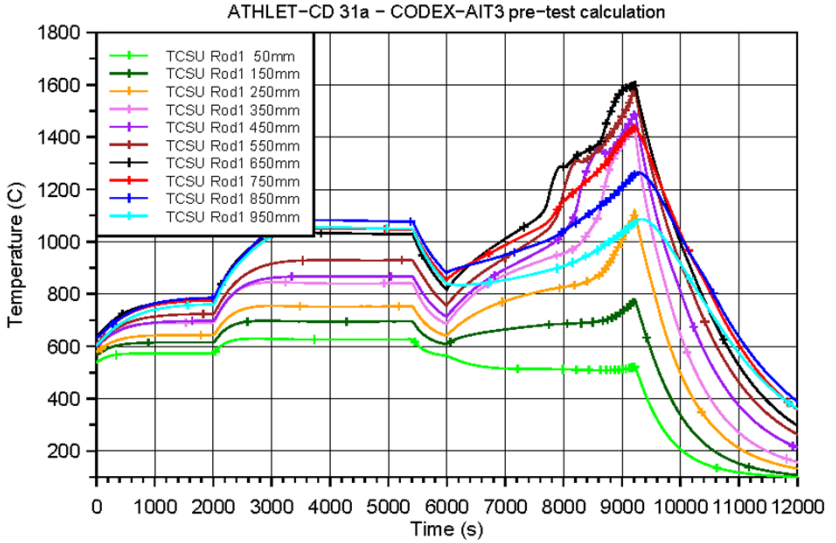
Cladding temperature outer rods



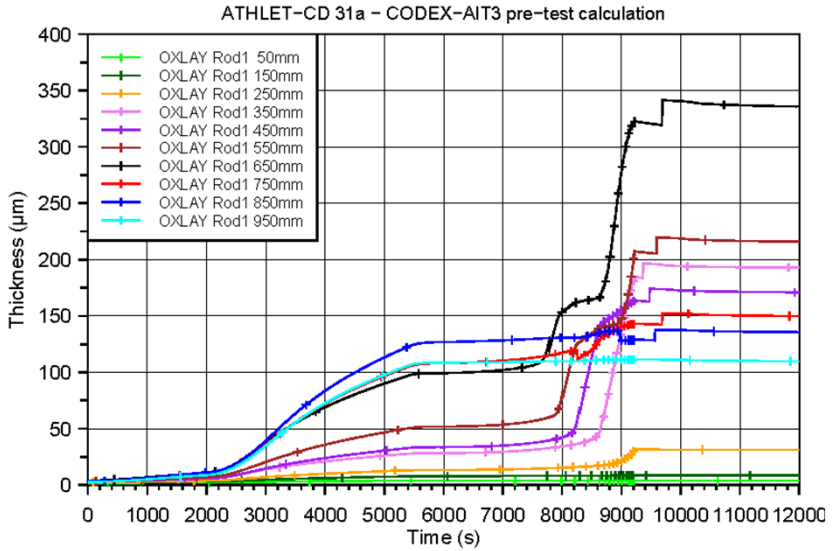
Surface temperature shroud

Results of pre-test calculations for test CODEX-AIT3 (4)

- **Results for temperatures and oxide layer thickness (reference calc.):**
  - Temperatures reach ~600°C at end of pre-heating phase (t=0s) in spite of higher flow rates than specified;
  - End of pre-oxidation:
    T_{max}= 1087°C (850 mm height)
    $\partial_{ox,max}$ = 132 µm at t= 5400s
  - End of intermediate cooling phase:
    (power 800 W)
    T_{max}= 884°C
  - Duration of air ingress phase 3224 s:
    t=6000 s - 9224 s
    when T_{max}= 1600°C is reached;
    $\partial_{ox,max}$ = 336 µm (650 mm height)
  - End of calculation at t=12000 s
    when T_{max} < 400°C
  - Only small difference of temperatures and oxide layer thicknesses between outer heated rods and central unheated rod (ΔT= 2°C);



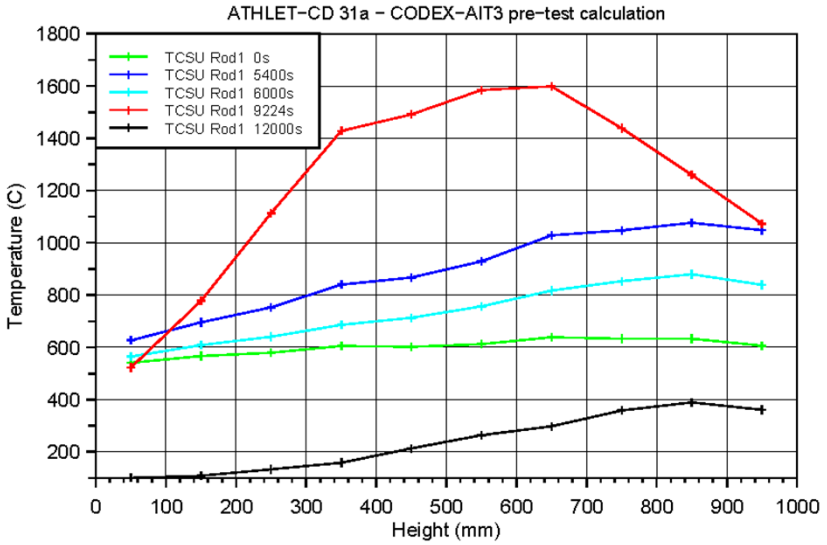
Cladding temperature of central unheated rod



oxide layer thickness of central unheated rod

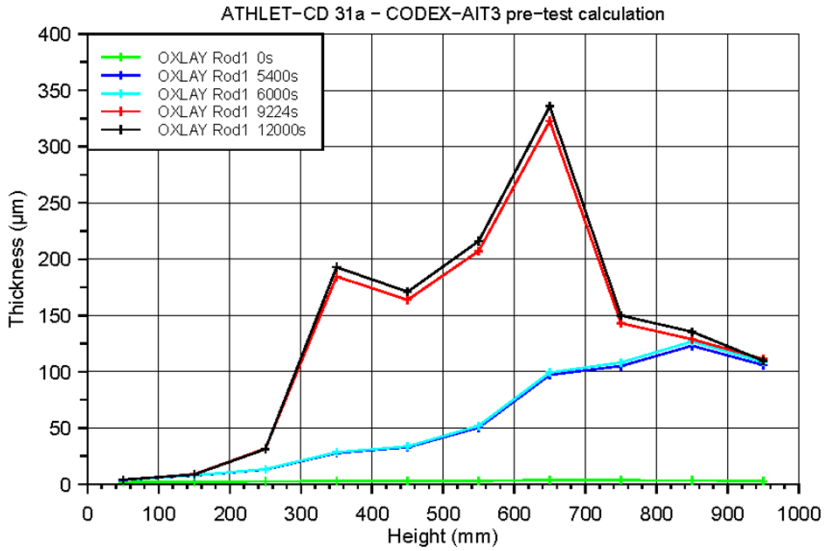
Results of pre-test calculations for test CODEX-AIT3 (5)

- **Temperature distribution over bundle length (reference calc.):**
  - After pre-oxidation period: highest temperature at 850 mm elevation
  - After air ingress period: highest temperature at 650 mm elevation



Surface temperature of central rod

- **Oxide layer** over bundle length:
  - Corresponding to the distribution of temperatures the max. oxide layer shifts from 850 mm after pre-oxidation to 650 mm after air ingress period;



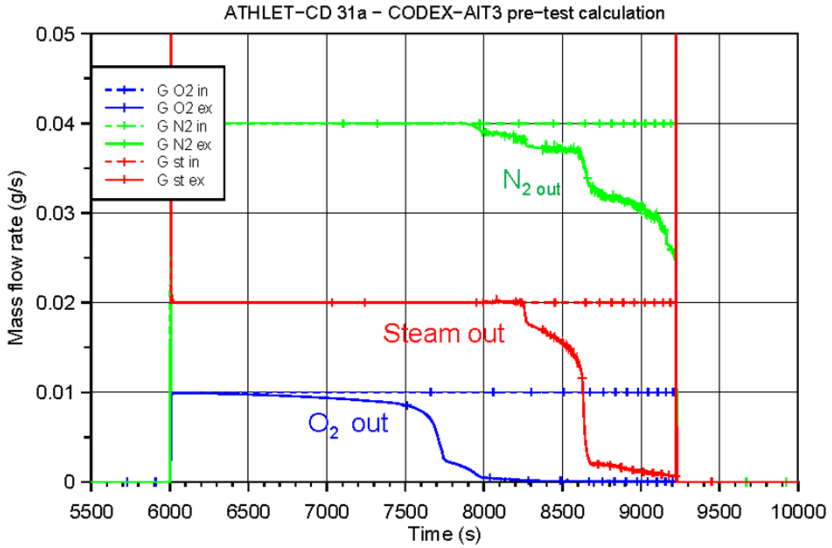
Oxide laxer thickness of central rod

Results of pre-test calculations for test CODEX-AIT3 (6)

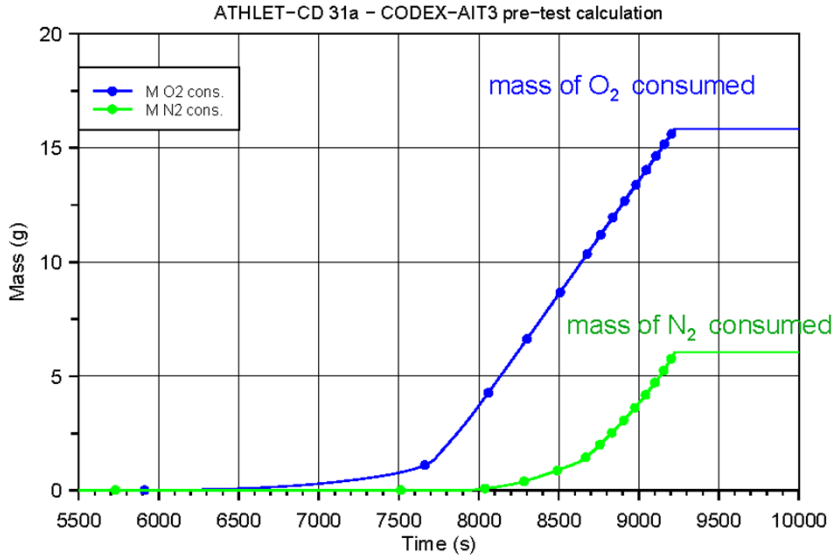
- **Results for consumed mass of O_2-N_2-steam (air ingress phase):**
  - no consumption of N_2 and steam as long as O_2 is available;
  - Consumption of N_2 starts when f_P = partial pressure of O_2 / p_{total} < 0.01, (derived from QU-16 results);
  - Consumption of steam starts when f_P = partial pressure of O_2 / p_{total} < 0.001;
  - Total O_2 consumption at end of simulation:

    m_{O2} = 15.8 g
  - Total N_2 consumption at end of simulation:

    m_{N2} = 6.0 g



O2 – N2 – steam mass flow rate



Consumed mass of O2 – N2

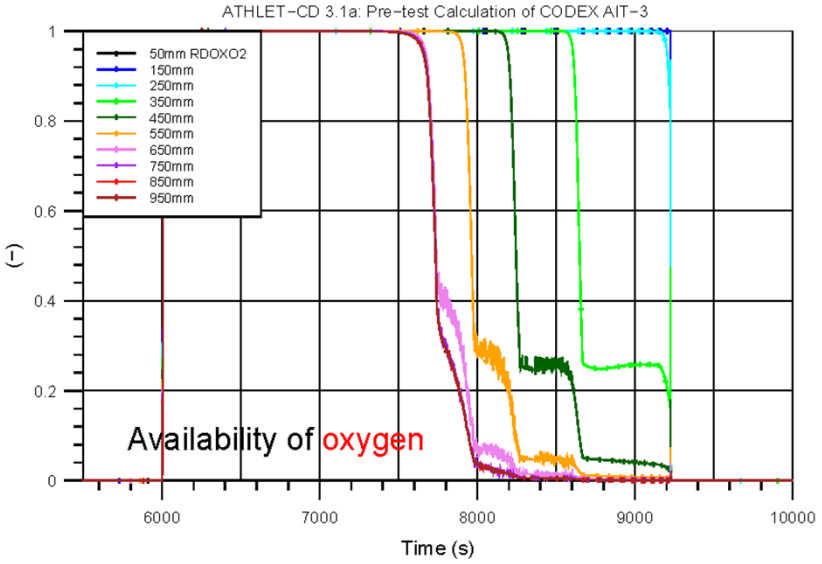
Results of pre-test calculations for test CODEX-AIT3 (7)

- **Results for starvation conditions:**

  - Starvation of oxygen:
    → start of O_2 starvation at t= 7950 s
    (RDOXO2 < 0.05)

    → period of O_2 starvation 1274 s ≈ 20 min.

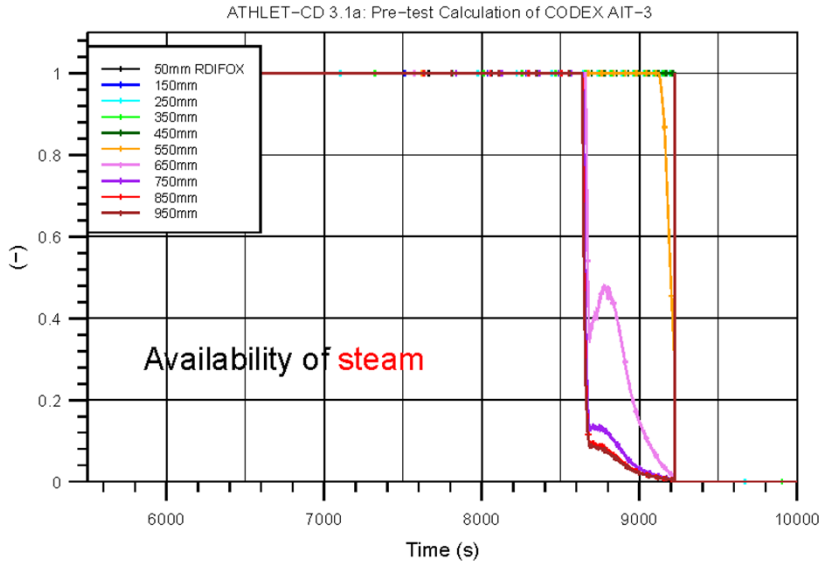
    → lowest elevation where O_2 was completely consumed:
    450 mm at t= 8678 s



  - Starvation of steam:
    → start of steam starvation at t ≈ 8800 s
    (RDIFOX < 0.05)

    → period of steam starvation 424 s ≈ 7 min

    → lowest elevation where steam was completely consumed:
    650 mm at t= 9127 s



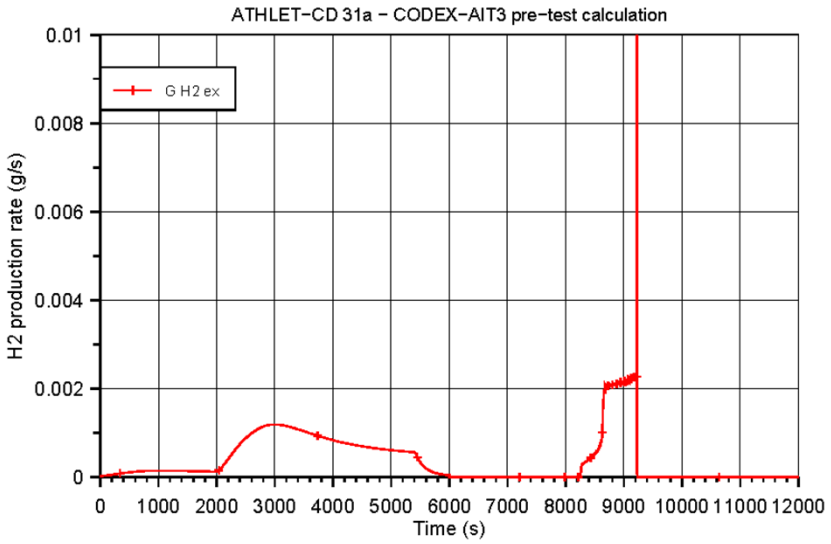
  - no starvation of nitrogen

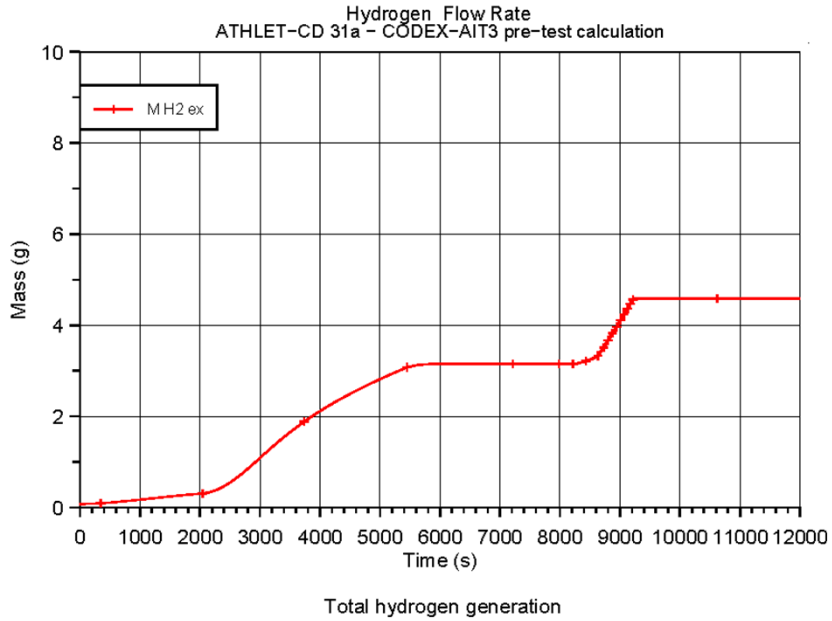
Results of pre-test calculations for test CODEX-AIT3 (8)

- **Results for produced hydrogen:**
  - Total H_2 generation at end of pre-oxidation:

    m_{H2} = 3.2 g
  - no hydrogen production during air ingress as long as O_2 is available;
  - Increase of hydrogen mass after O_2 starvation
  - Total H_2 generation at end of simulation:

    m_{H2} = 4.6 g





Summary (1)

- **Modelling:**
  - Pre-test simulation for CODEX-AIT3 was made
    - with given geometry of bundle and rod configuration (2 wires of heater were summarized to 1 heater with equivalent volume; re-calculation of material properties for tungsten-ZrO_2 mixture dep. on thickness of single heater element)
    - with given material properties for insulation,
    - with options for air ingress as adopted for QUENCH-16
  - using a test version of ATHLET-CD 31a which was adopted previously for an improved post-test calculation of QUENCH-16 (with resulting 94 g H_2 instead of 122 g in experiment from simulated components)
  - after verification with post-test calculation of some previous CODEX experiments: CODEX-2, LOCA E4, Commissioning test previous to AIT3

- **Parameter studies:**
  - Pre-test simulations were made with
    - variation of power
    - variation of argon flow rate
    - variation of steam and air flow rate
  - ➢ with the aim of T_{max}~ 1100°C and max. oxide layer thickness ~ 100µm at the end of pre-oxidation phase (t=5400 s);
  - ➢ with the aim of slow temperature increase up to a max. of 1600°C over a time period of ~ 3600 s during steam-air phase with oxygen and steam starvation;

Summary (2)

- **Pre-oxidation phase:**
  - With the specified argon flow rate of G=0.02 g/s the calculated temperature is higher than the aimed value of **T_{max} ~ 1100 °C** and a max. oxide layer of **~100 µm;**
  - With an argon flow rate of G=1.0 g/s (and a steam flow rate of 1.0 g/s) the aimed value of **T_{max} ~ 1100 °C** and a max. oxide layer of **~100 µm** can be kept;

- **Air oxidation phase with air and steam:**
  - The specified power (2kW-3kW) leads to a quick increase to temperatures T>1600°C and a very short air ingress phase – the aimed conditions were reached with reduced power;
  - Intermediate cooling down to ~900°C after pre-oxidation (800W with steam and argon flow 1.0 g/s) helps to get a slower temperature increase (duration of air-steam phase ~ 1 hour);
  - Very low mass flow rates for steam and air with G < 0.02 g/s were avoided (measurement!);
  - A duration of the air ingress phase of Δt=3224 s until T_{max} > 1600°C with air and steam starvation was calculated with G(argon)=0.1 g/s, G(air)=0.05 g/s, G(steam)=0.02 g/s at a power of 800 W;
  - Influence of external electrical resistance → uncertainty of results; (WHRES0=0.006 was taken due to best estimate for CODEX-2, LOCA E4; WHRES0=0.003 was best estimate value for Commissioning test)
    - Higher temperatures (quicker heat up) would be calculated for simulation with lower value of electrical resistance!

- **Cool down:**
  - ATHLET-CD calculates cool down to temperatures T < 400°C within ~ 2800 s with the specified argon flow rate of G=2.0 g/s.

Acknowledgement

- This work was carried out on behalf of the German Federal Ministry for Economic Affairs and Energy (project no. RS1519 and RS1548) on the basis of a decision by the German Bundestag.

Thank you for your attention!

☺

Integrated uncertainty analysis of CORA BWR experiments using RELAP/SCDAPSIM

Rawan Mustafa – Jordan Atomic Energy Commission
Chris Allison and Marina Perez – Innovative Systems Software
Hiroshi Madokoro – University of Tokyo (KIT)
Heriberto Sanchez - University of Mexico

The reassessment of the CORA BWR experiments using RELAP/SCDAPSIM is an important component of our (Innovative Systems Software's) ongoing Fukushima Daiichi assessment activities. These experiments (a) accurately reflect the design of a BWR fuel assembly with associated channel box/ control blade and (b) cover a range of thermal hydraulic conditions most like those expected among the three units. Several, although not all, of these experiments have been previously analyzed using both SCDAP/RELAP5 (in the 1980s) and by RELAP/SCDAPSIM over the past 10-15 years. The calculated results, were in general, in reasonably good agreement with the experimental results with notable sources of uncertainty associated with the modeling of the electrically heat fuel rod simulators, shroud thermal properties and associated radial heat losses, and unique thermal hydraulic design of the CORA facility. However, no systematic uncertainty studies have been performed previously.

This presentation provides one of two first examples at this meeting of an integrated uncertainty analysis of German CORA and QUENCH electrically heated experiments, looking at both the influence of uncertainties in experimental conditions and important models/correlations. The CORA calculations demonstrated the use of the new IUA, "Integrated Uncertainty Analysis", option introduced into RELAP/SCDAPSIM/MOD3.4 in the summer of 2017. RELAP/SCDAPSIM/MOD3.X(IUA) allows users to easily compute ranking statistics and uncertainty distributions using base assessment input model together with a list of modeling parameters and associated uncertainty distributions. These parameters can be "source code" parameters that cannot be modified through user input. Examples include the uncertainties in (a) parabolic equations for Zircaloy oxidation, (b) tungsten heater element resistances, and (c) convective heat transfer coefficients. These parameters can also be "input" quantities defined in the base assessment input model. Examples are the contact resistance of the heater elements and thermal conductivity of porous zirconia used in the shroud.

The presentation included comparisons between calculated and measured bundle temperatures for the CORA-17 and CORA-18 and the results of the integrated uncertainty analysis for the CORA-18 experiment.



SDTP

Development of best estimate methods for reactor and advanced fluid systems safety analysis and training

Integrated uncertainty analysis of CORA BWR experiments using RELAP/SCDAPSIM

Analysis and contributions by

Rawan Mustafa – Jordan Atomic Energy Agency

Heriberto Sanchez - University of Mexico

Hiroshi Madokoro – University of Tokyo (KIT)

Dr. Marina Perez – ISS

Dr. S. Hagen - KIT

Presented

Dr. Chris Allison

Innovative Systems Software (ISS)

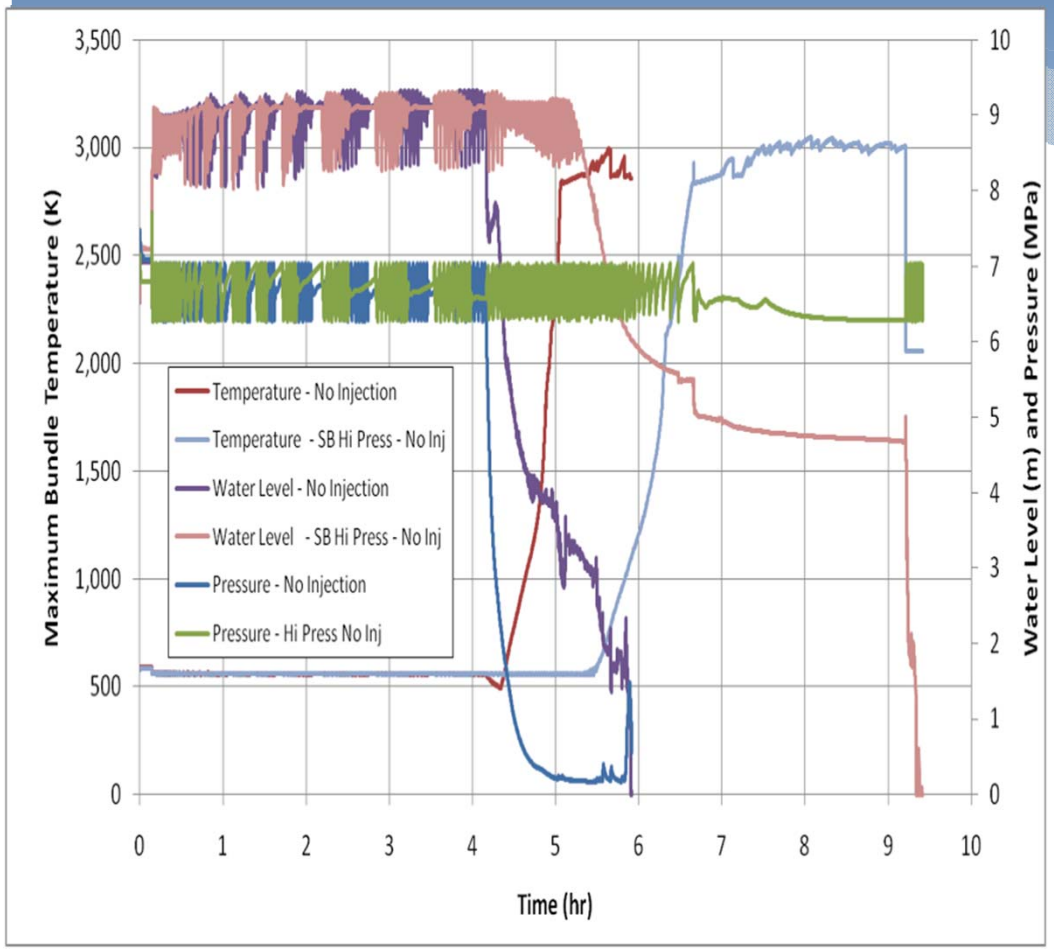
Outline

* **Motivation** – why is the reassessment of the code using the CORA BWR experiments important?
* **Examples of recent calculations** using RELAP/SCDAPSIM/MOD3.4 (most widely used production version)
* **Application of MOD3.4 with new integrated uncertainty analysis** (IUA) option to CORA-18
* **Future plans** to expand capabilities of MOD3.x with IAU

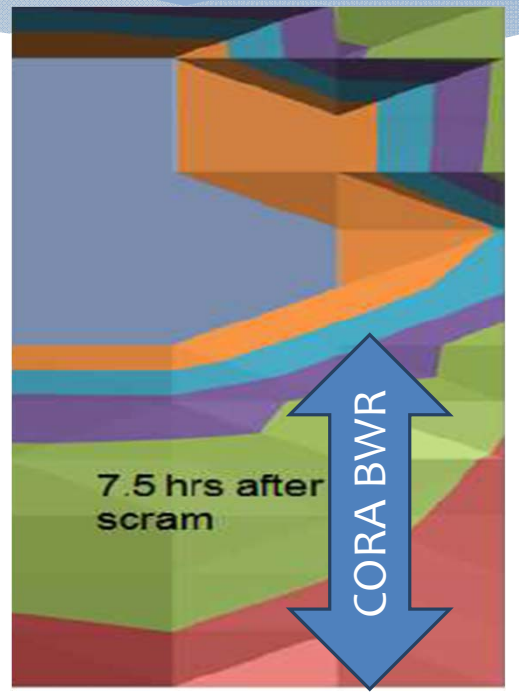
Additional slides

* Other historic and other recent "published" calculations for CORA BWR experiments

CORA BWR experiments help define likely state of lower regions of Fukushima Daiichi core prior to relocation of molten [U-Zr]-O_2

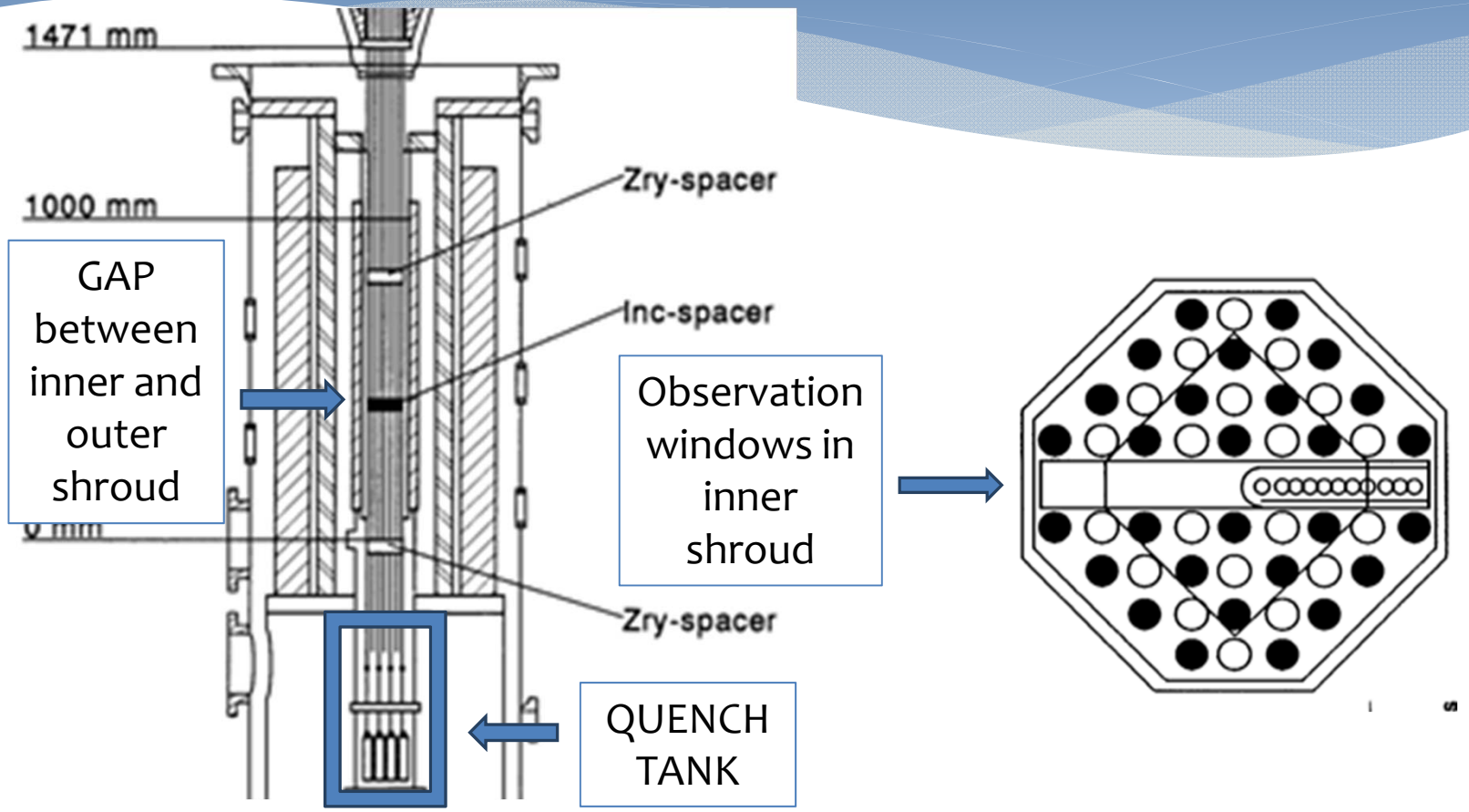


No depressurization prior to fuel melting

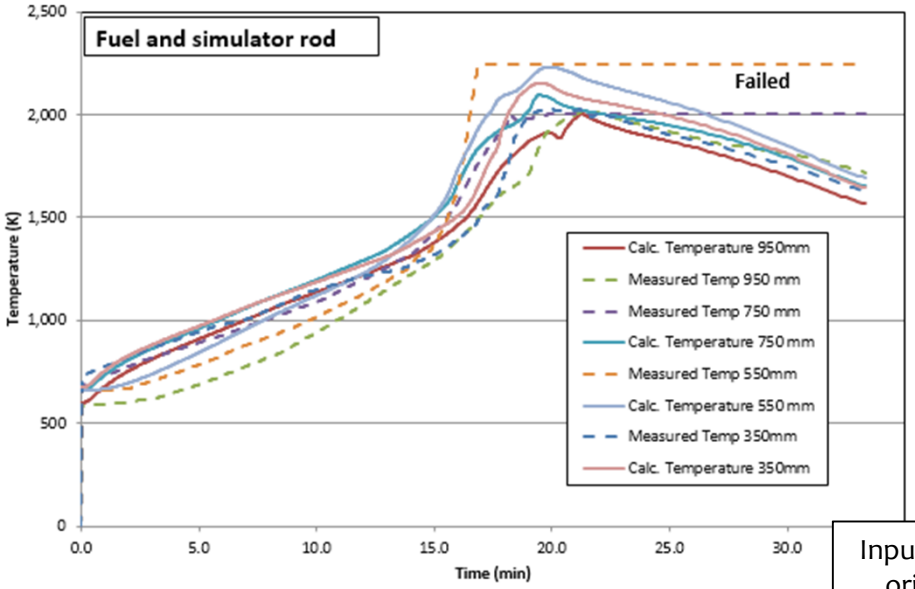




CORA facility had more complicated thermal boundary conditions than that of Quench facility



Examples

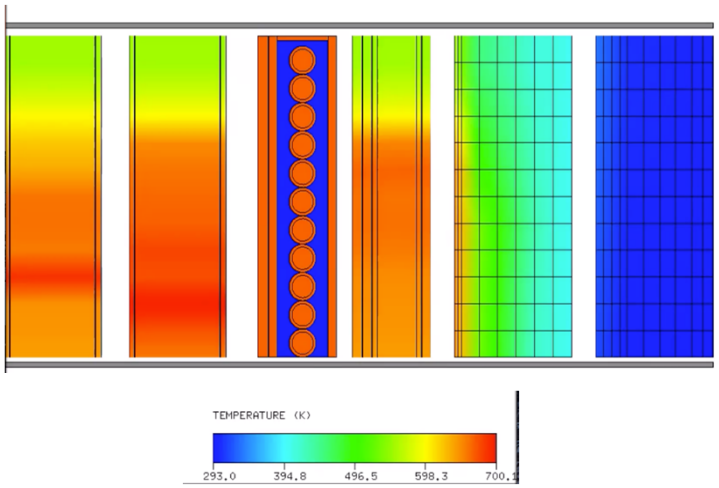


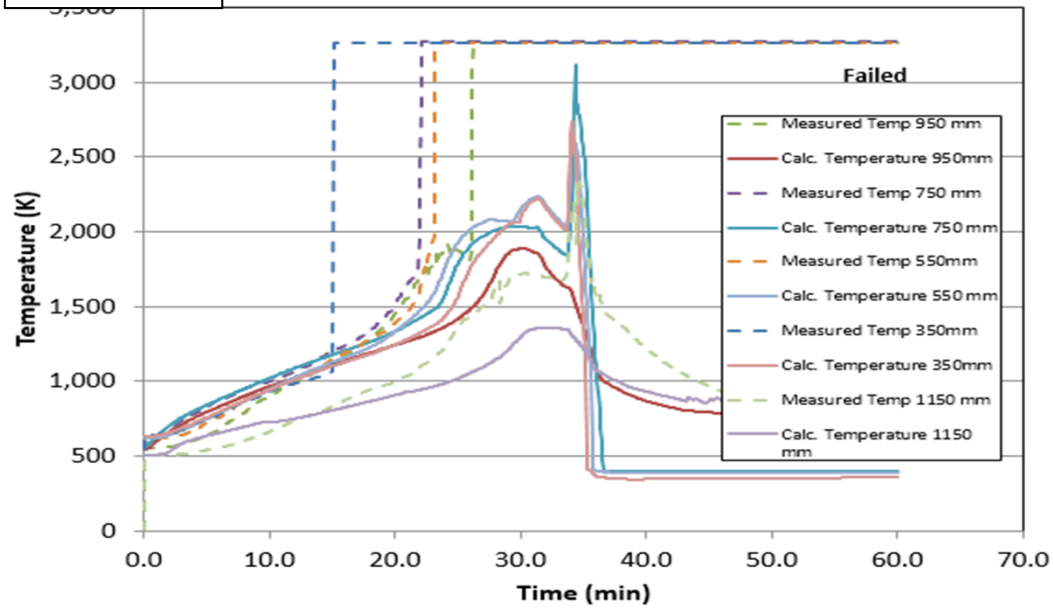
Analysis and contributions by
Rawan Mustafa – Jordan Atomic Energy Agency
Heriberto Sanchez - University of Mexico

CORA-17
MOD3.4

Input models originally developed by ORNL

CORA-18
MOD3.4





CORA-18 uncertainty analysis using RELAP/SCDAPSIM/MOD3.4 with IUA (integrated uncertainty analysis) underway

* RELAP/SCDAPSIM/MOD3.X(IUA) allows user to easily compute ranking statistics and uncertainty distributions using base input model
  * User defines parameters to include plus uncertainty distributions for defined parameters
    * Input parameters
    * Source code parameters
  * New "setup" option builds input models and uncertainty files for uncertainty analysis
  * New "post processing" option computes uncertainty distributions and ranking statistics for user selected quantities

CORA-18 uncertainty analysis using RELAP/SCDAPSIM/MOD3.4 with IUA (integrated uncertainty analysis) underway

* RELAP/SCDAPSIM/MOD3.X(IUA) allows user to easily compute ranking statistics and uncertainty

User defines parameters to include plus uncertainty distributions for defined parameters

- Input parameters
- Source code parameters

computes uncertainty distributions and ranking statistics for user selected quantities

CORA-18 Example

* Wall to fluid heat transfer
* Tungsten resistivity
* Contact resistance
* Zr oxidation parabolic rate
* Shroud Zirconia therm. cond.

CORA-18 uncertainty analysis … RELAP5 SETUP input(1)

100 uncsetup

29000000 0 0 0.95 0.95 1 0 1 200 298341

* Source correlations parameters

*Heat transfer coef. (Wall to fluid heat transfer - vapor)

29000002 HTC.09.110 UD 0.70 1.30 … *Trapezoidal distribution

*Tungsten resistivity

29000003 RES.COR.TUNG ND 1.00 0.025 … *Normal distribution

*Oxidation rate

29000004 OXI.MATPRO ND 1.00 0.075 … *Normal distribution

CORA-18 uncertainty analysis … RELAP5 SETUP input(2)

*Shroud thermal conductivity

29101000 UD 0.8 1.2 0.0 0.0

29101001 -1.0 20 20100401 2 2 1

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.

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*external heater rod resistance

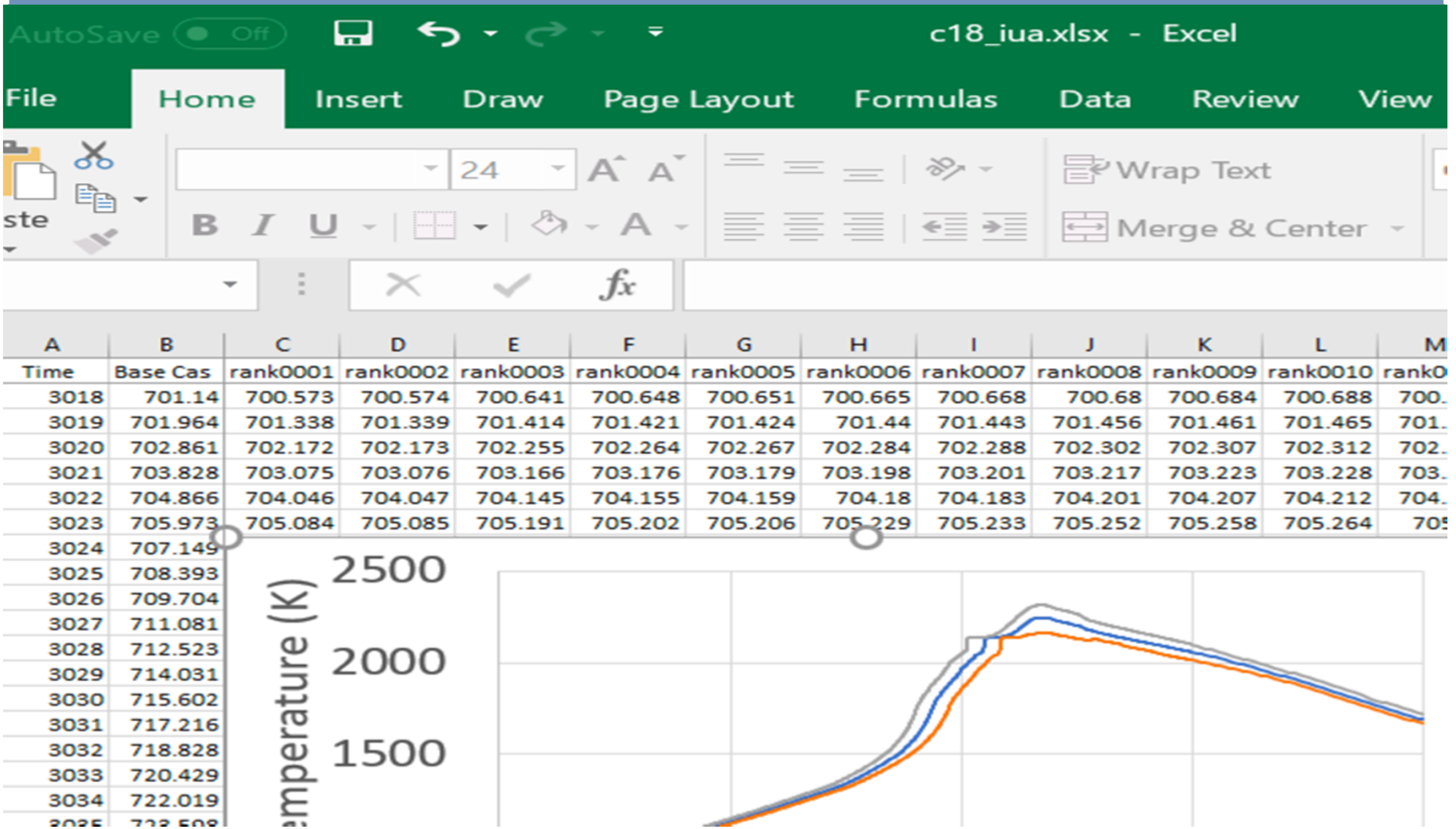
29102000 UD 0.814814815 1.185185185 0.0 0.0

29102001 -1.0 20 40020100 4 4 1

***Base input model**

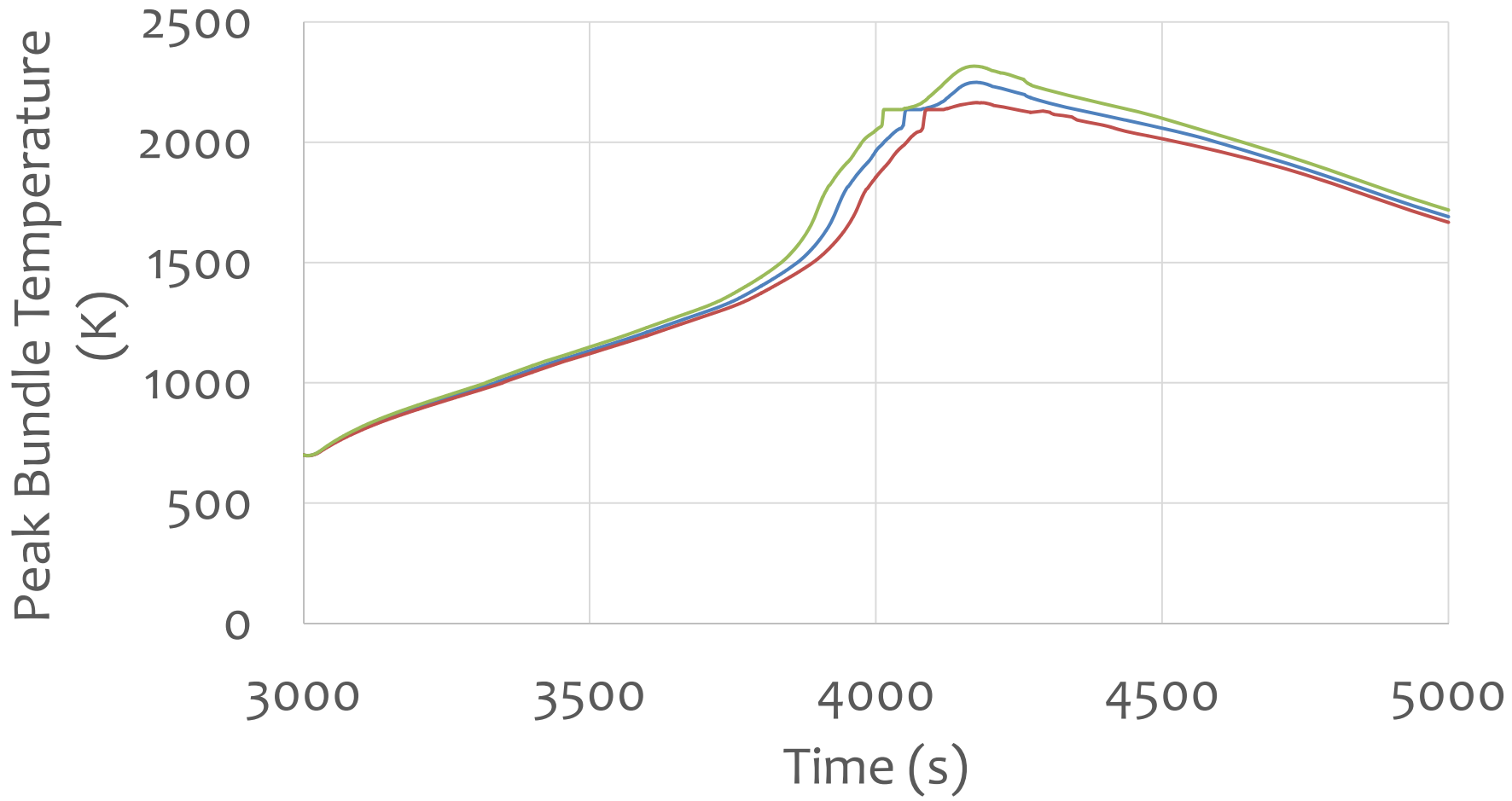
…

CORA-18 uncertainty analysis … generating uncertainty distributions and ranking

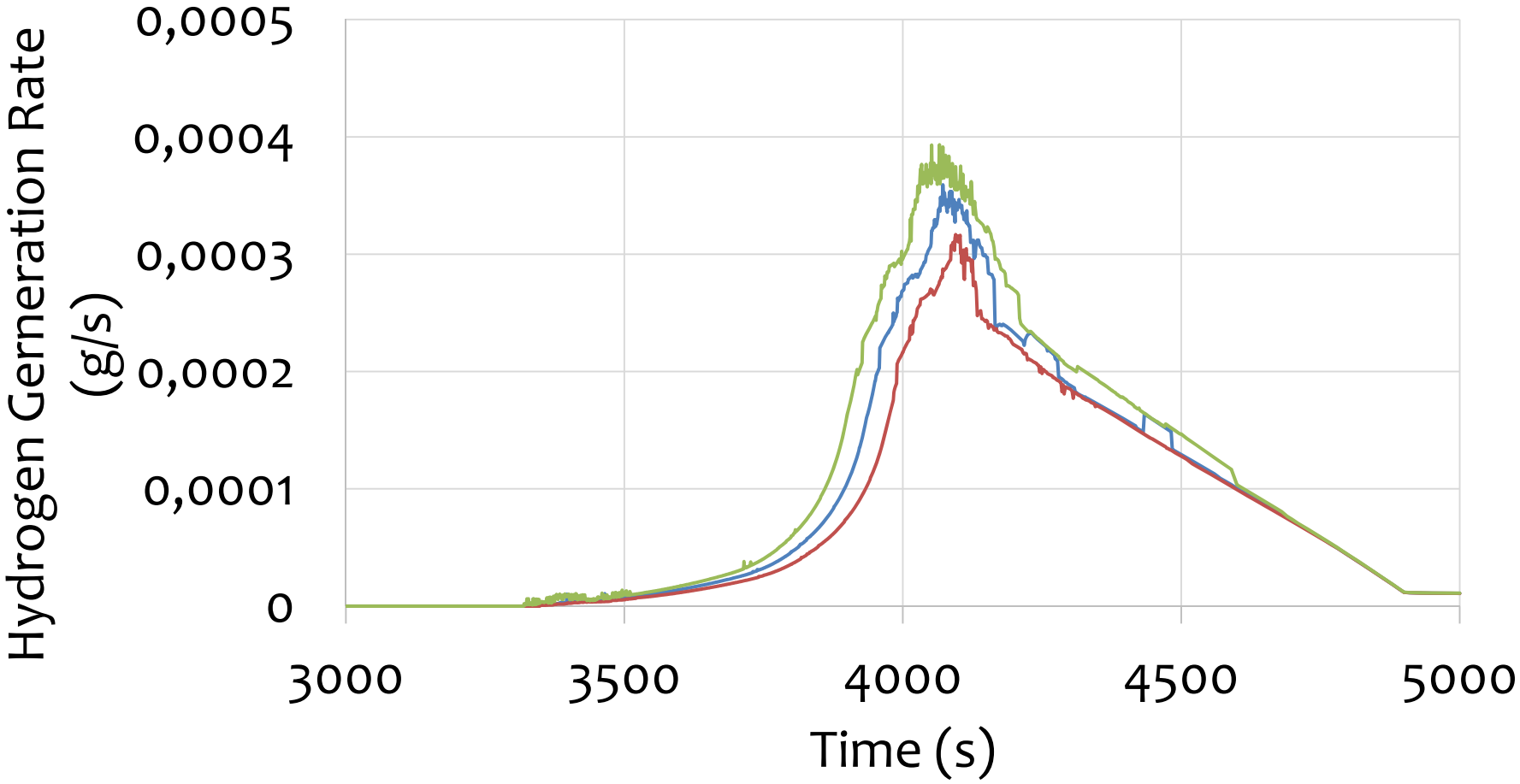


A	B	C	D	E	F	G	H	I	J	K	L	M
Time	Base Cas	rank0001	rank0002	rank0003	rank0004	rank0005	rank0006	rank0007	rank0008	rank0009	rank0010	rank0
3018	701.14	700.573	700.574	700.641	700.648	700.651	700.665	700.668	700.68	700.684	700.688	700.
3019	701.964	701.338	701.339	701.414	701.421	701.424	701.44	701.443	701.456	701.461	701.465	701.
3020	702.861	702.172	702.173	702.255	702.264	702.267	702.284	702.288	702.302	702.307	702.312	702.
3021	703.828	703.075	703.076	703.166	703.176	703.179	703.198	703.201	703.217	703.223	703.228	703.
3022	704.866	704.046	704.047	704.145	704.155	704.159	704.18	704.183	704.201	704.207	704.212	704.
3023	705.973	705.084	705.085	705.191	705.202	705.206	705.229	705.233	705.252	705.258	705.264	705
3024	707.149											
3025	708.393											
3026	709.704											
3027	711.081											
3028	712.523											
3029	714.031											
3030	715.602											
3031	717.216											
3032	718.828											
3033	720.429											
3034	722.019											

Peak Bundle Temperatures



Hydrogen Generation Rate



Future development plans for IUA options

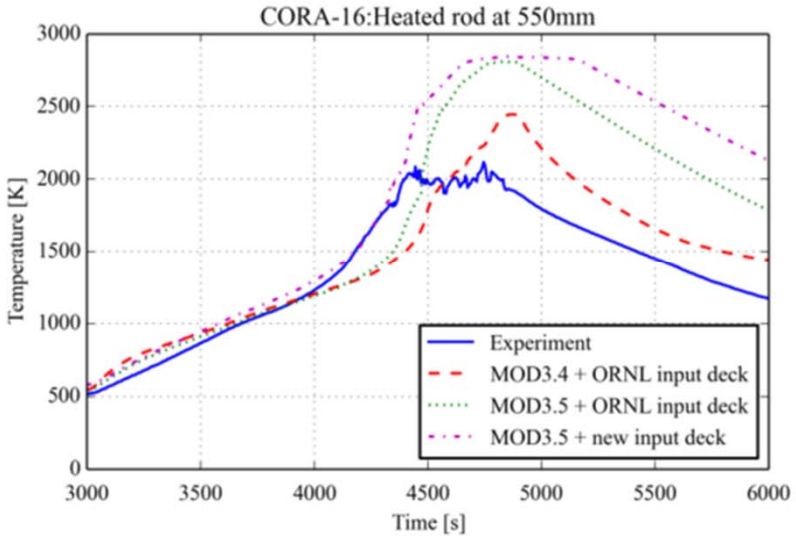
* **Develop key model parameter assessment distributions data base based on reassessment activities** (reduce user effects)
  * Protect current modeling options used for model development and assessment in experimental versions
  * Ensure best estimate model parameters are automatically used by code for base calculations
* **Integrate uncertainty assessment distributions data base** for correlations (MATPRO and other important correlations) and key models (further simplify ease of use)
  * Users can then just identify correlations and models by defined name to include in uncertainty assessment

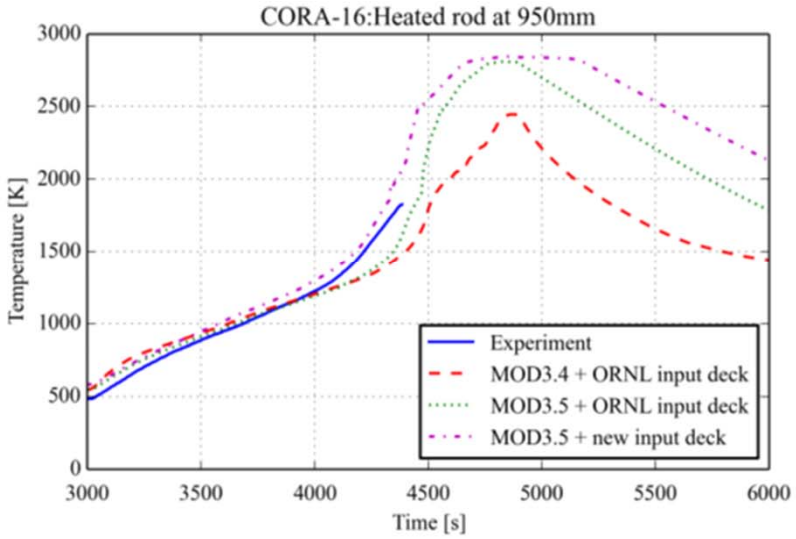
Additional slides

Base calculations for CORA 16, 17, and/or 18 performed using several versions of RELAP/SCDAPSIM by participants from Technical University of Sophia, University of Tokyo, as well as ISS

* Historic and other recent "published" sensitivity studies have focused on influence of uncertainty in thermal hydraulic boundary conditions experimental uncertainties in code-to-data comparisons
  * Input models follow recommended guidelines for nodalization and modeling parameters for CORA facility so have little impact on results
  * **Dominant contributors**
    * **Bundle temperatures** - Shroud thermal properties (radial heat losses), axial power distribution (due to temperature dependency of resistance), flow bypass between inner and outer shroud, observation windows in inner shroud
    * **Hydrogen generation rates** - Time delays and variations in hydrogen measurements, steam generation from lower quench tank in CORA facility

Examples

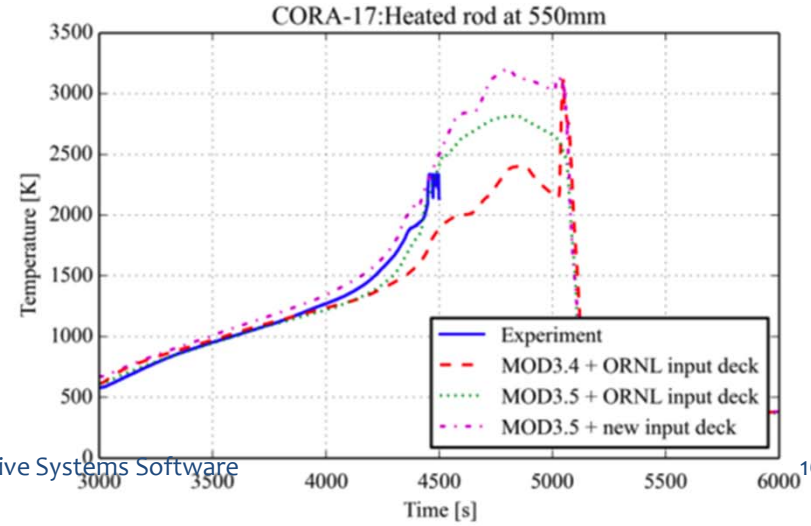


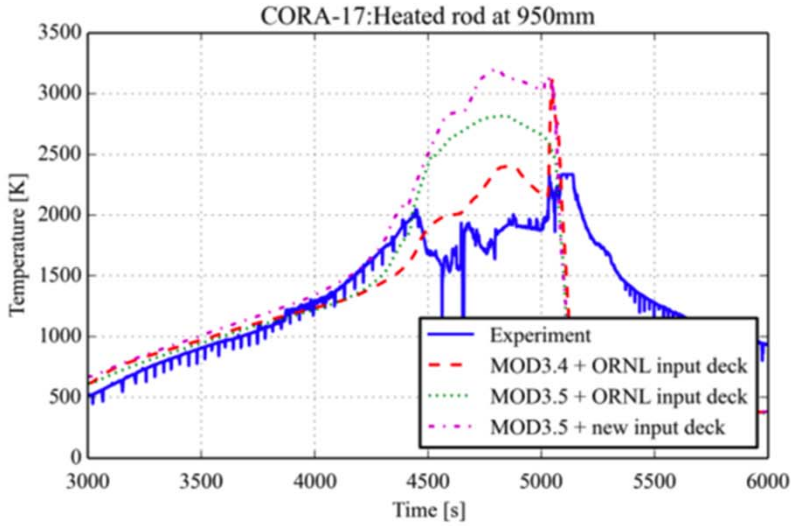


H. Madokoro, "Assessment of RELAP/SCDAPSIM with QUENCH and CORA Analysis", Master Thesis, Department of Nuclear Engineering and Management, School of Engineering, University of Tokyo, 37-126808, September 2014.

MOD3.5dp

* Improvements in fuel rod simulator, B4C control rod, and shroud components
* Improved fuel rod gap conductance



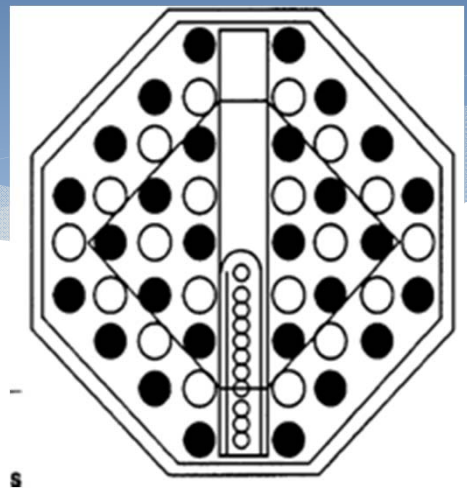


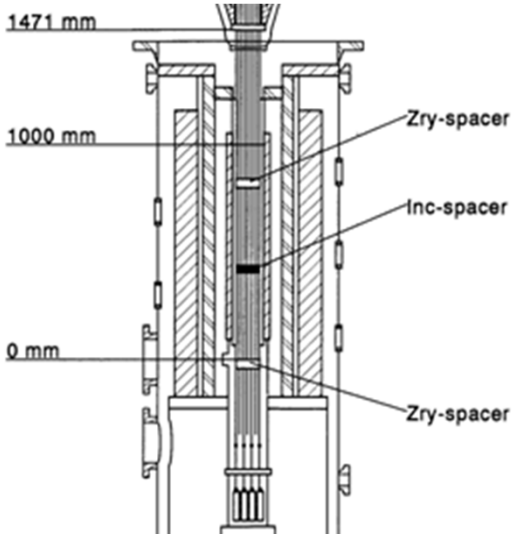
Reassessment and uncertainty analysis started through SDTP university support activities – including BWRs

- **Introduction of MOD3.x with (IUA) integrated uncertainty analysis option in summer of 2017**
- **Re-assessment of RELAP/SCDAPSIM for BWR applications using historical SA experiments (2014-current)**
  - **Representative BWR assembly experiments in CORA facility – CORA 16/17/18/28/30/31/33 (CORA 17 published in open literature)**
  - **In-pile experiments with {U-Zr}-O_2 melt formation and early relocation – LOFT-FP-2, PHEBUS-FPT, PBF-SFD (FPT3, SFD ST, 1-4 published)**

Representative BWR assembly experiments in CORA facility – CORA 16/17/18/28/30/31/33

Experiment	Conditions
CORA 16	Reference experiment
CORA 17	Water addition
CORA 18	Large bundle – influence of size
CORA 28	Influence of pre-oxidation
CORA 31	Influence of slow initial heatup (0.3 K/s)
CORA 33	Influence of steam starvation





Study of the axial discretization influence in simulation of QUENCH-06 with RELAP/SCDAPSIM mod3.5dp

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[b] *Innovative Systems Software, Idaho 83406, USA*
issgml97@gmail.com

Abstract

The physical-chemical behavior of the reactor core material in accidents related to loss of coolant and re-flooding is essential for the nuclear safety. QUENCH-06 experiment is used for investigation of the pre-oxidized LWR fuel rod behavior during the cooling down with water. It is included in International Standard Problem ISP-45. The objective of this work is to study the axial discretization influence in simulation of QUENCH-06 with RELAP/SCDAPSIM mod3.5dp. For that purpose five models with different nodalization in the heated part of the test bundle are tested. They are based on the basic ISS input deck. The new SCDAP heater rod model is used. The measured electrical resistance is applied. The results show good agreement between the measured and predicted bundle temperatures for all phases of the experiment, as the maximum deviation is about 100 ˚C. The exception is the shroud temperature at elevation 1150 mm, which is underestimated by all models. The location of cladding and shroud failure is predicted more precisely by the models with axial nodalization in the heated part of the bundle greater than 11 nodes. At the same time the increasing the number of nodes reduces the predicted hydrogen.



Study of the axial discretization influence in simulation of QUENCH-06 with RELAP/SCDAPSIM mod3.5dp

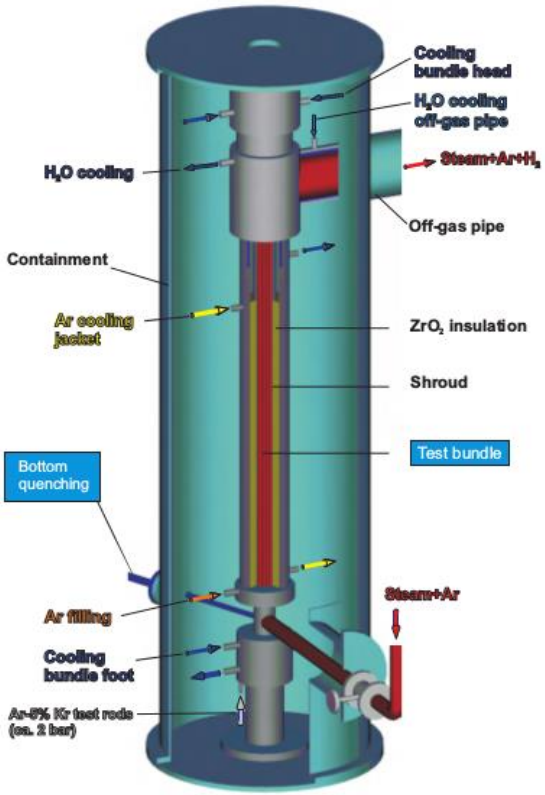
Ivan Spasov (TU-Sofia)
Chris Allison (ISS)

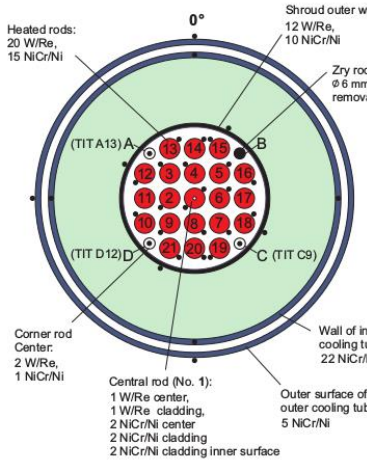
18.10.2017

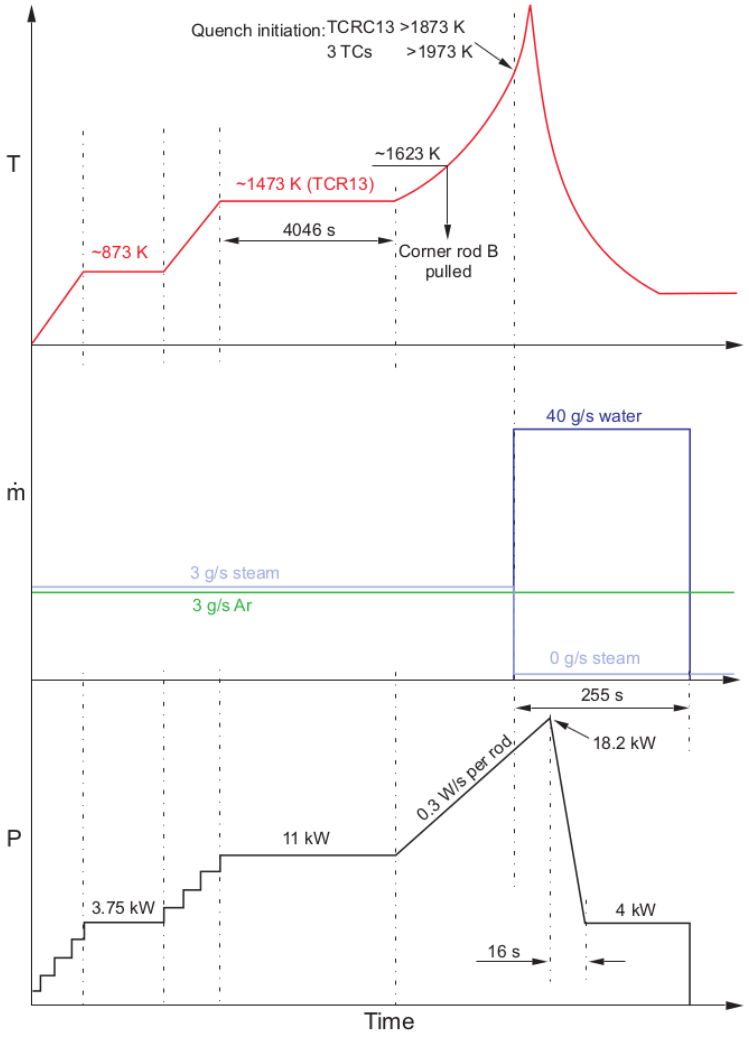
OUTLINE

1. **Objective:** explore the impact of the axial discretization in simulation of QUENCH-06 with RELAP/SCDAPSIM mod3.5dp
2. **Facility and experiment description**
3. **Nodalization scheme**
4. **Modeling assumptions**
5. **Results**
6. **Conclusions**

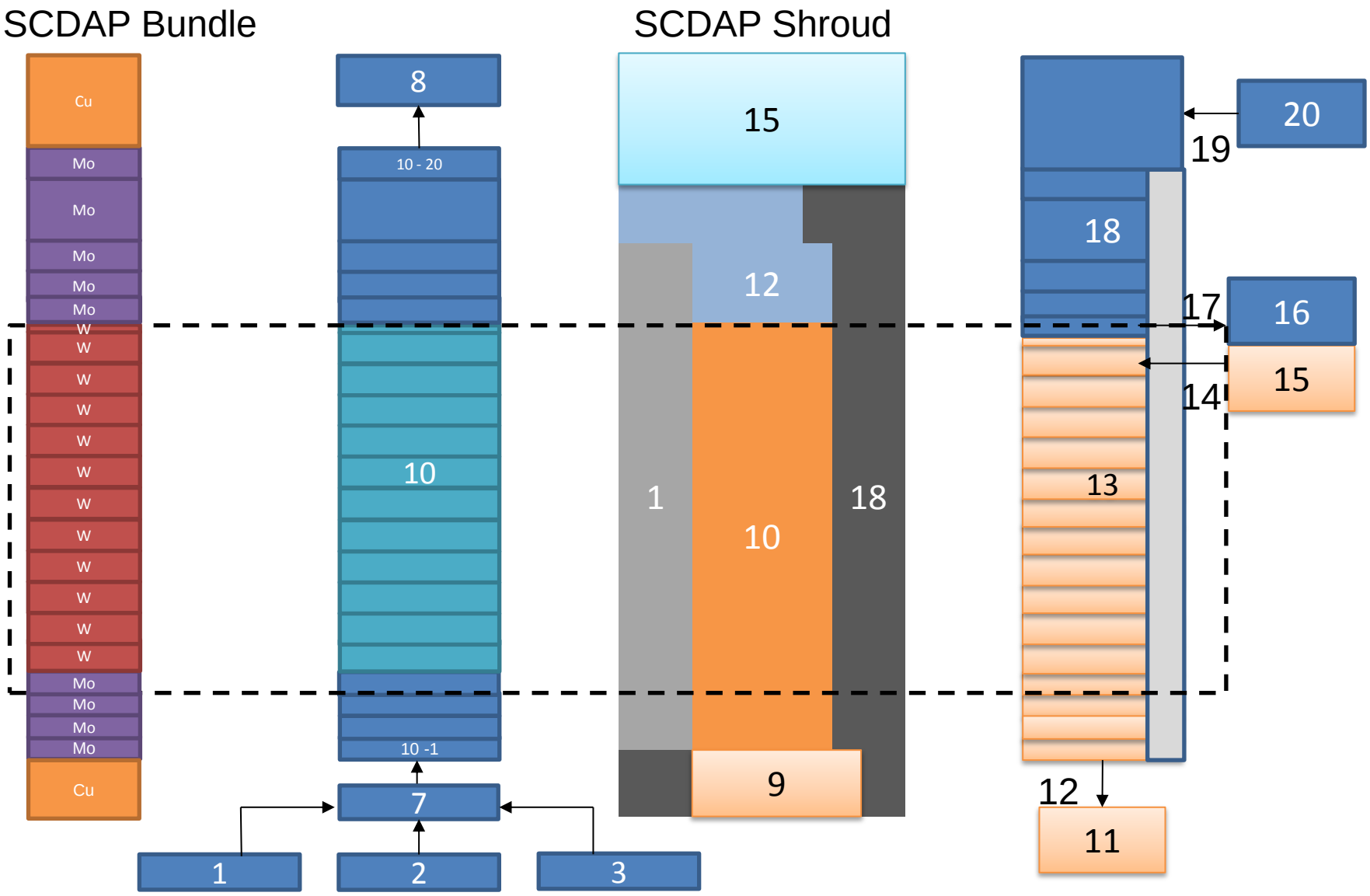
Facility and experiment description







NODALIZATION SCHEME



MODELING ASSUMPTIONS

- 5 models with 17, 22, 27, 32 and 37 axial nodes are tested
- In axial direction the heated part of test bundle is modeled as:
  - 5 nodes x 0.2 m and 1 node x 0.025 m (17 axial nodes);
  - 10 nodes x 0.1 m and 1 node x 0.025 m (22 axial nodes);
  - 15 nodes x 0.06667 m and 1 node x 0.025 m (27 axial nodes);
  - 20 nodes x 0.05 m and 1 nodes x 0.025 m (32 axial nodes);
  - 25 nodes x 0.04 m and 1 nodes x 0.025 m (37 axial nodes).

MODELING ASSUMPTIONS

- Imitators of fuel rods and surrounding shroud are modeled by SCDAP components.
- **The unheated rod** is represented by the SCDAP fuel rod component. The following radial discretization is used:
  - 3 nodes for the ZrO2 pellet;
  - 1 nodes for the gap;
  - 10 nodes for the cladding.

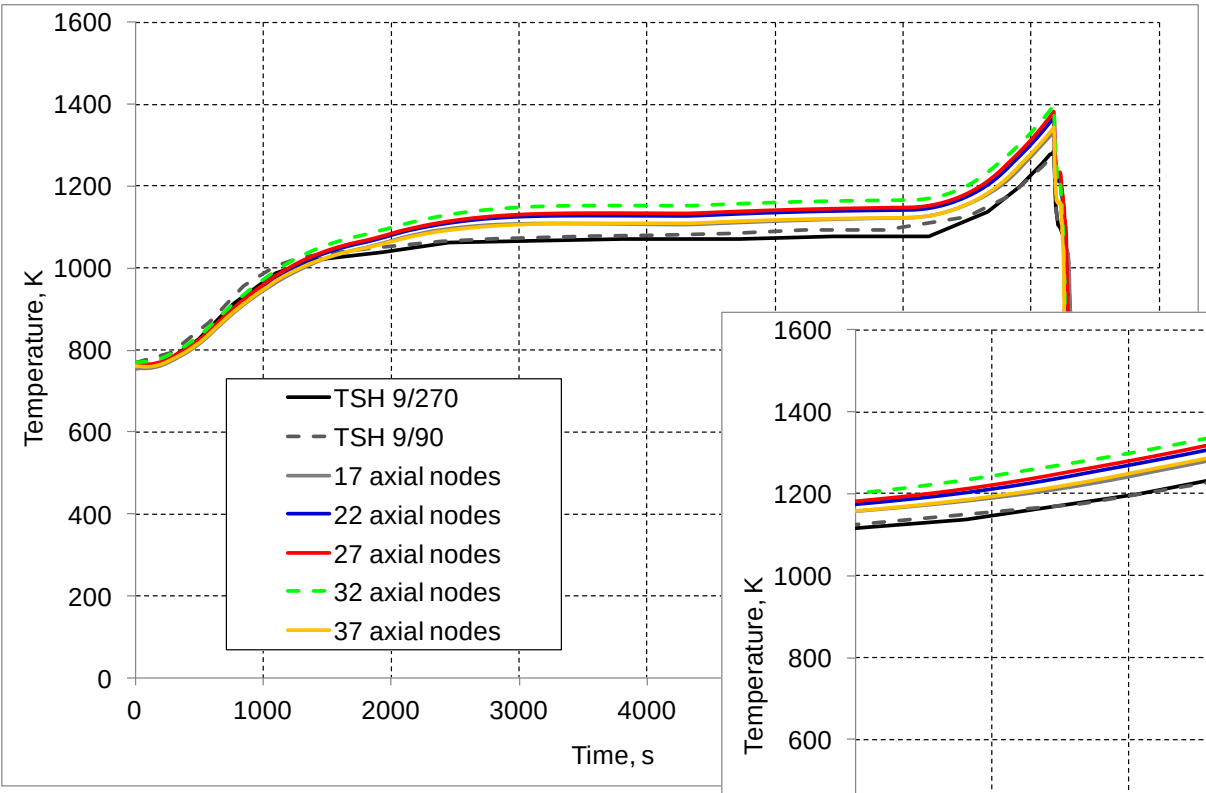
MODELING ASSUMPTIONS

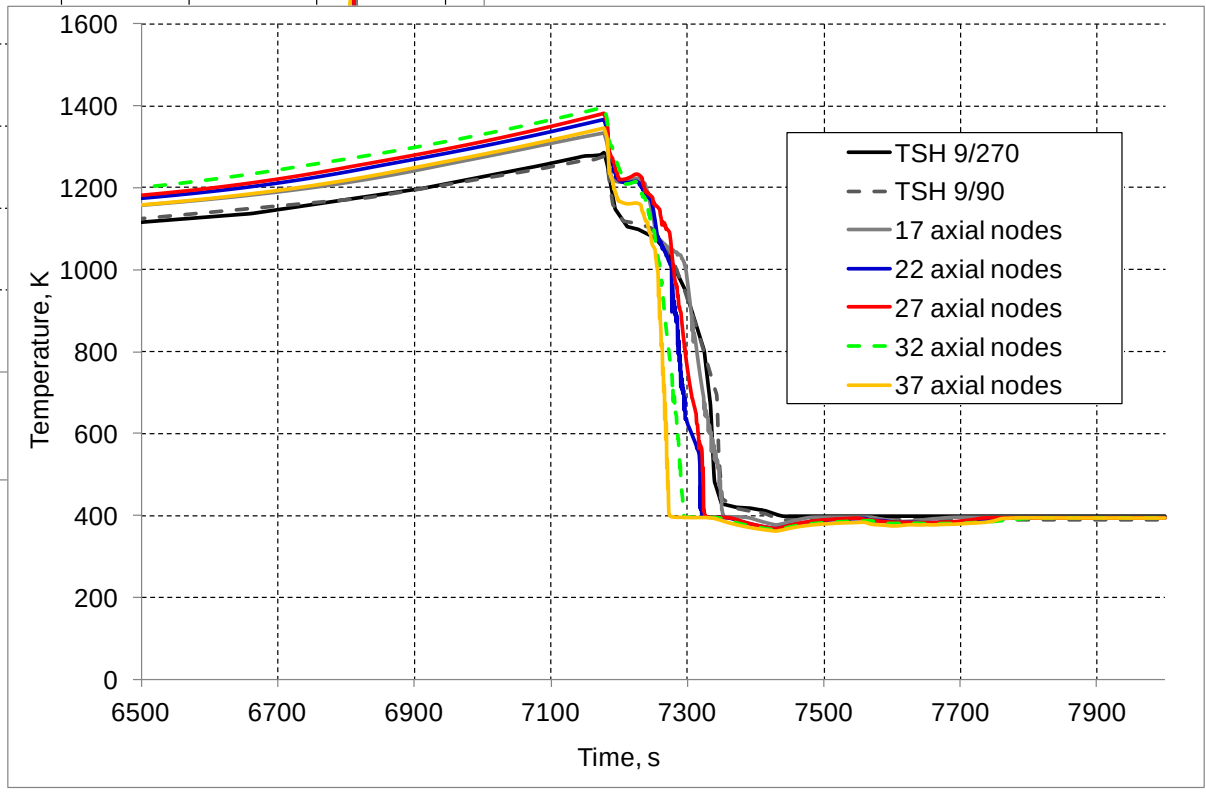
- **Heated rods:**
  - the new simulator component is used;
  - 8 inner and 12 outer rods are modeled using 2 components;
  - External resistance equal to 0.0042 Ω is applied
  - Gap conductance model is used
  - The changes in the radius of heater rod element due to thermal expansion is taken into account
  - Pressure drop caused by ballooning is modeled
- Radial nodalization in the heated part of simulator rods:
  - 3 nodes for the tungsten
  - 1 for the first gap
  - 3 nodes for ZrO2 pellet
  - 1 node for the second gap and 6 nodes for the cladding

MODELING ASSUMPTIONS

- **Corner rods** are modeled with SCDAP fuel rod component;
- Radial nodalization:
  - 1 node for the zircalloy pellet ;
  - 1 nodes for the gap;
  - 9 nodes for the cladding.
- **Shroud:**
  - it is modeled by SCDAP shroud component;
  - 17 radial mesh points are modeled;
  - User specified materials are applied;
  - the thermal properties of shroud provided by the manufacturer are used.

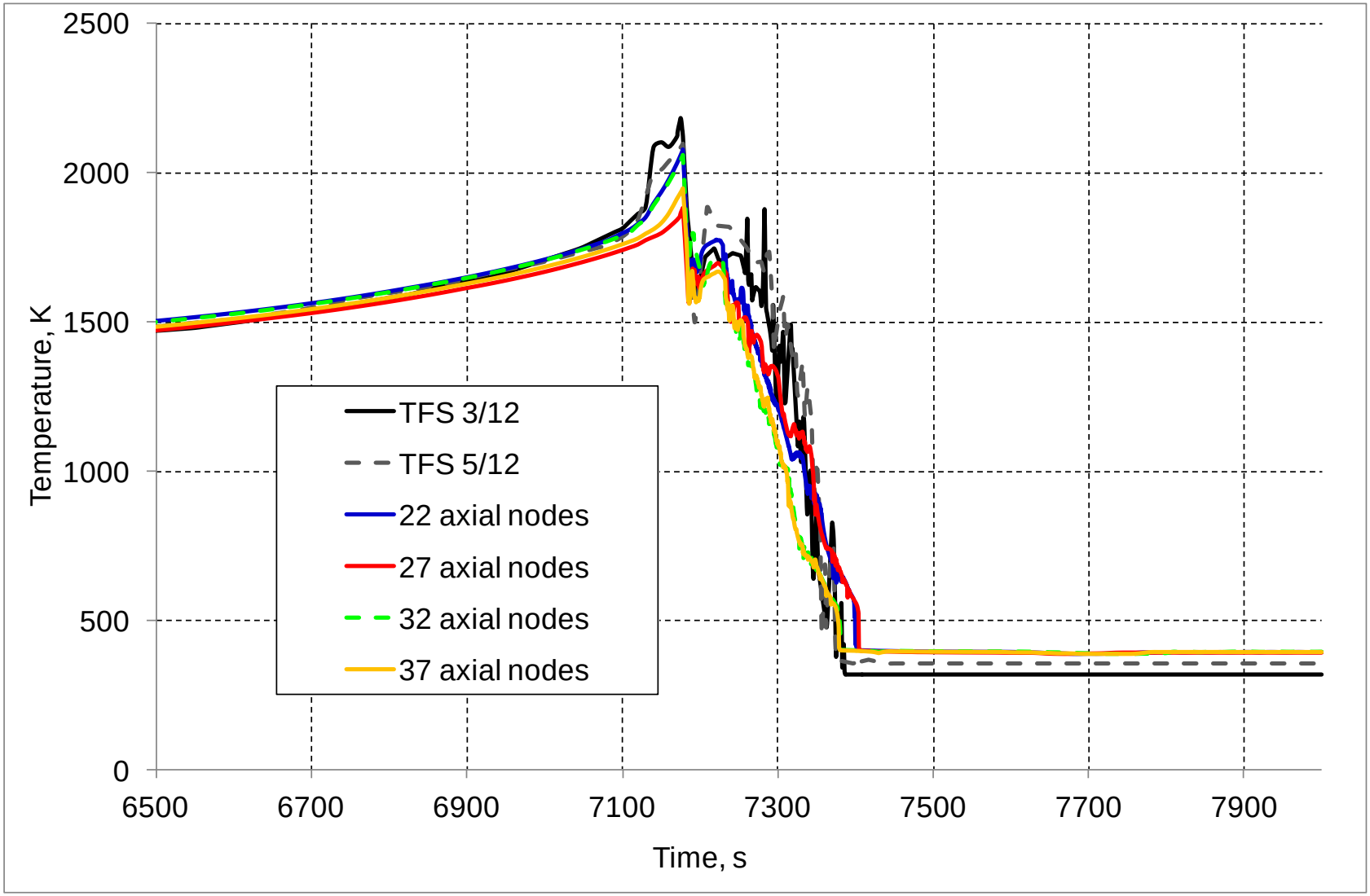
RESULTS





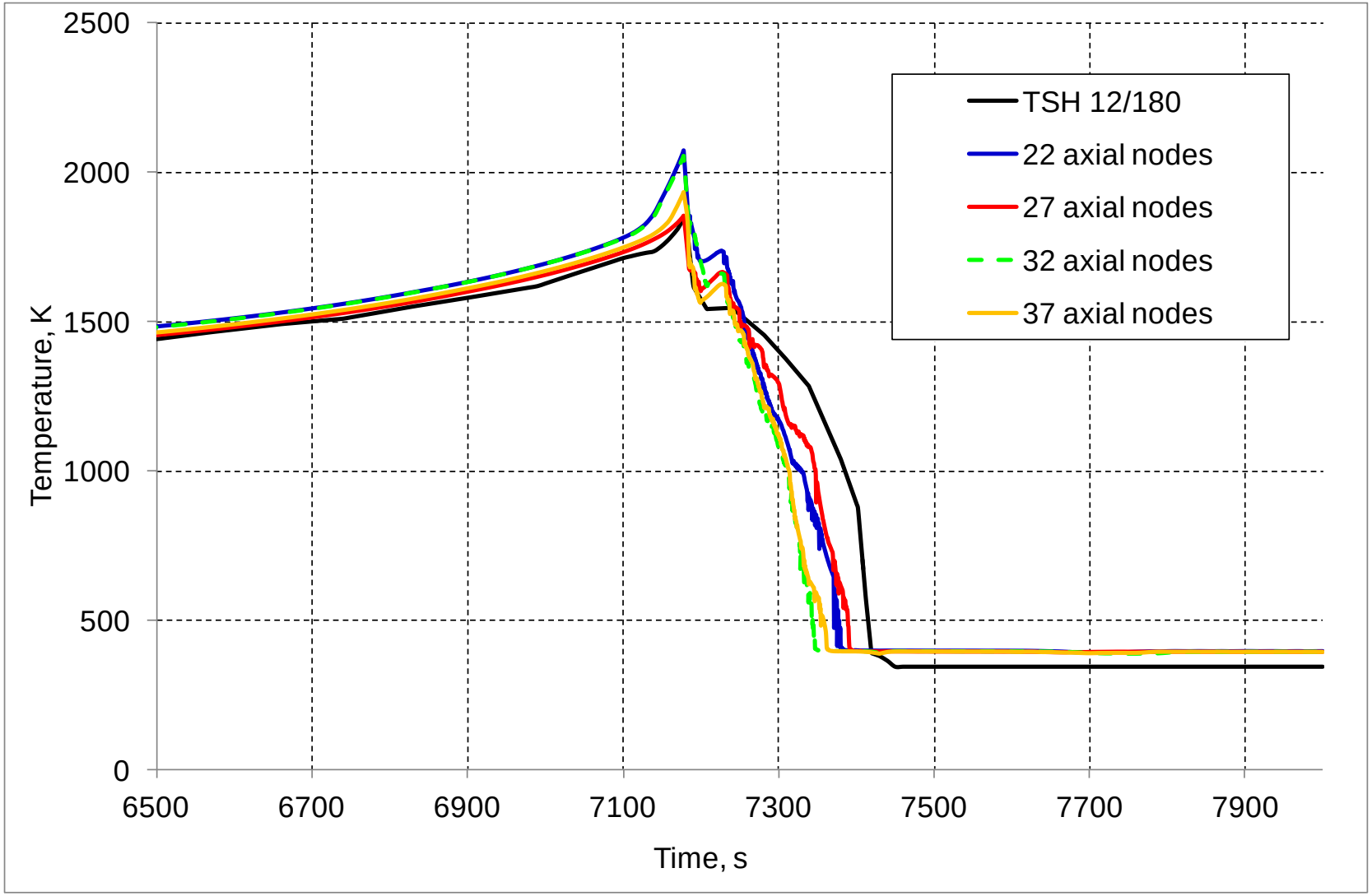
Time history of the shroud temperature at elevation +550

RESULTS



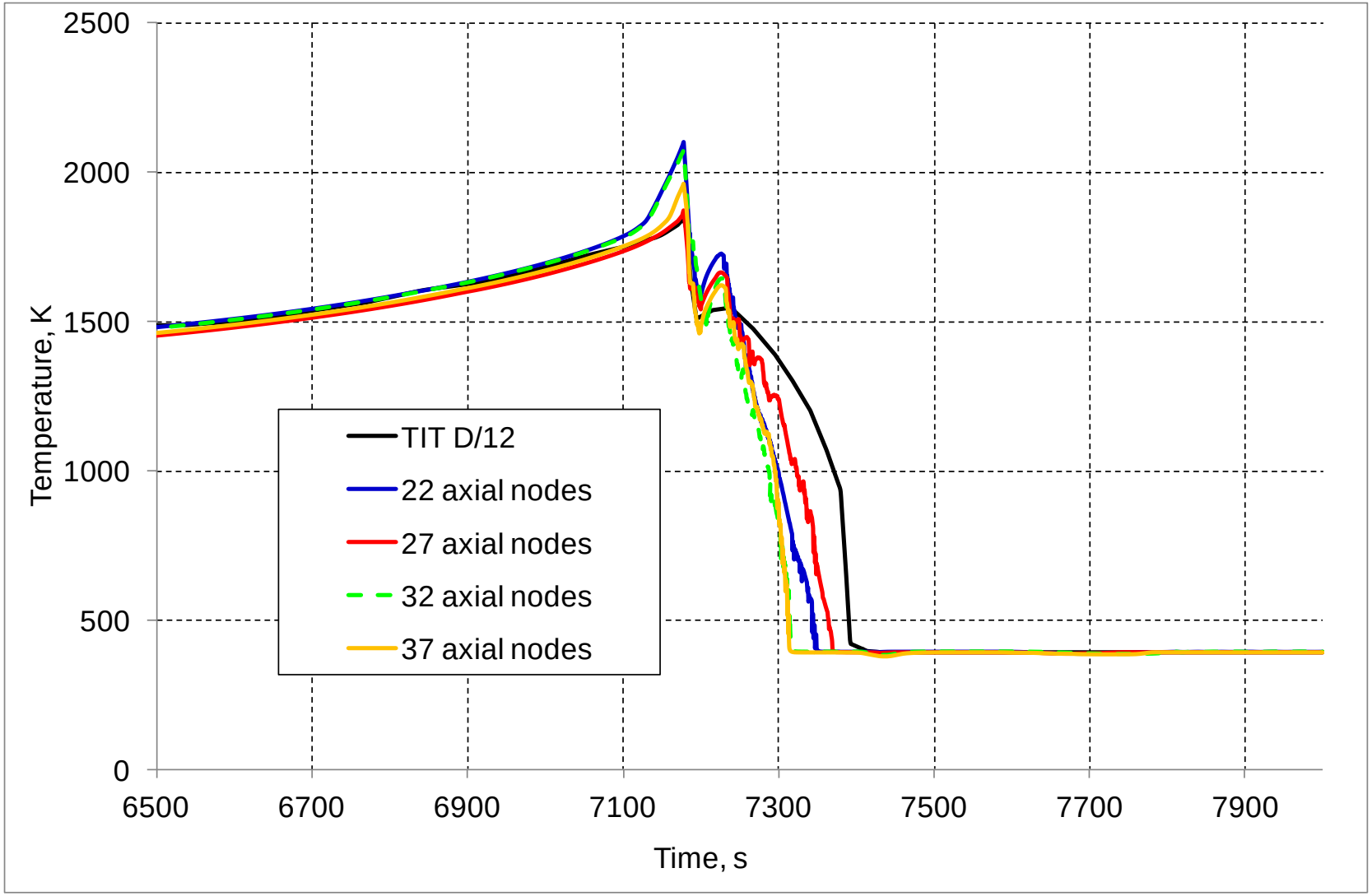
Time history of the central rod surface temperature at elevation +850

RESULTS



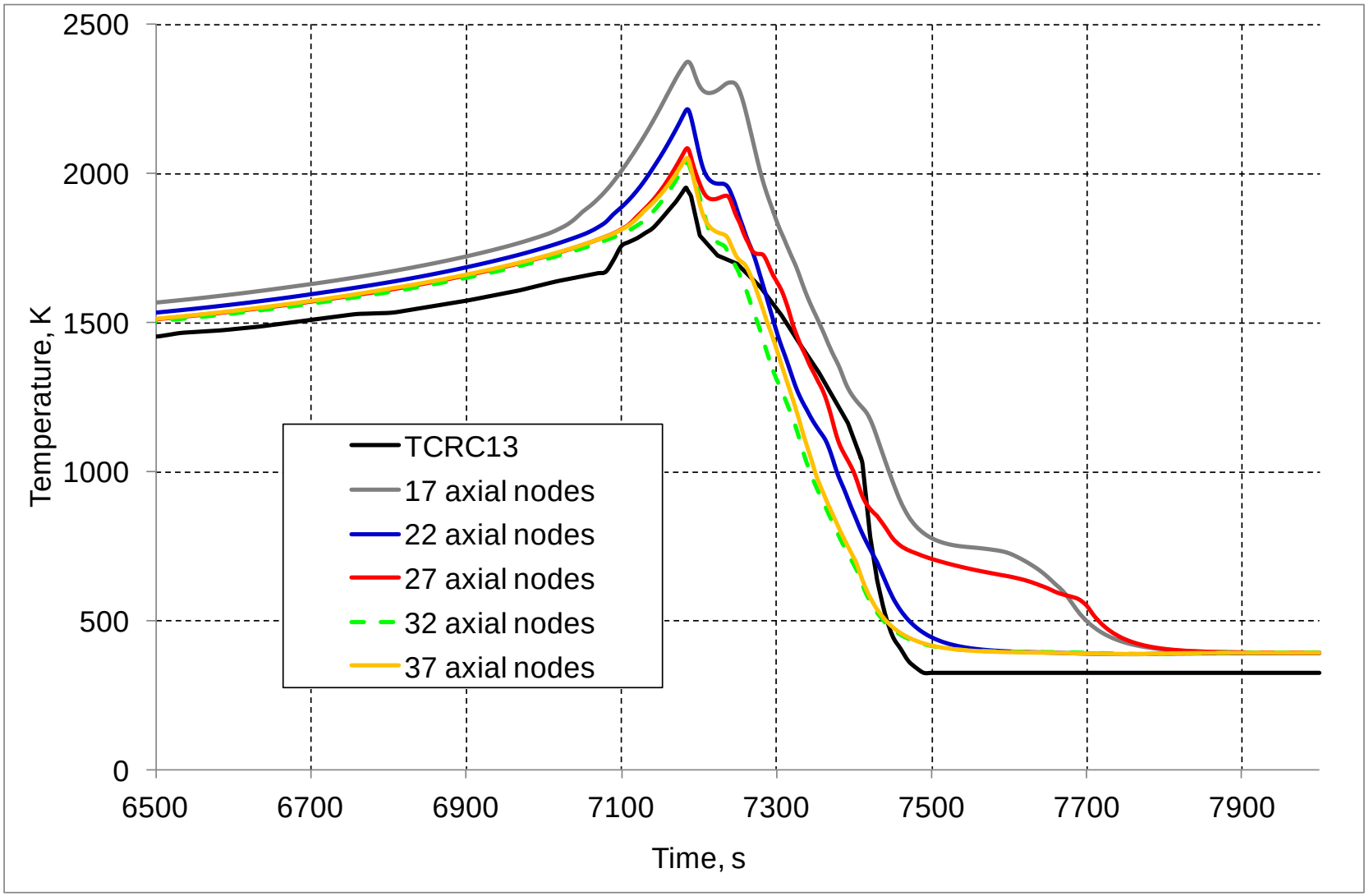
Time history of the shroud temperature at elevation +850

RESULTS



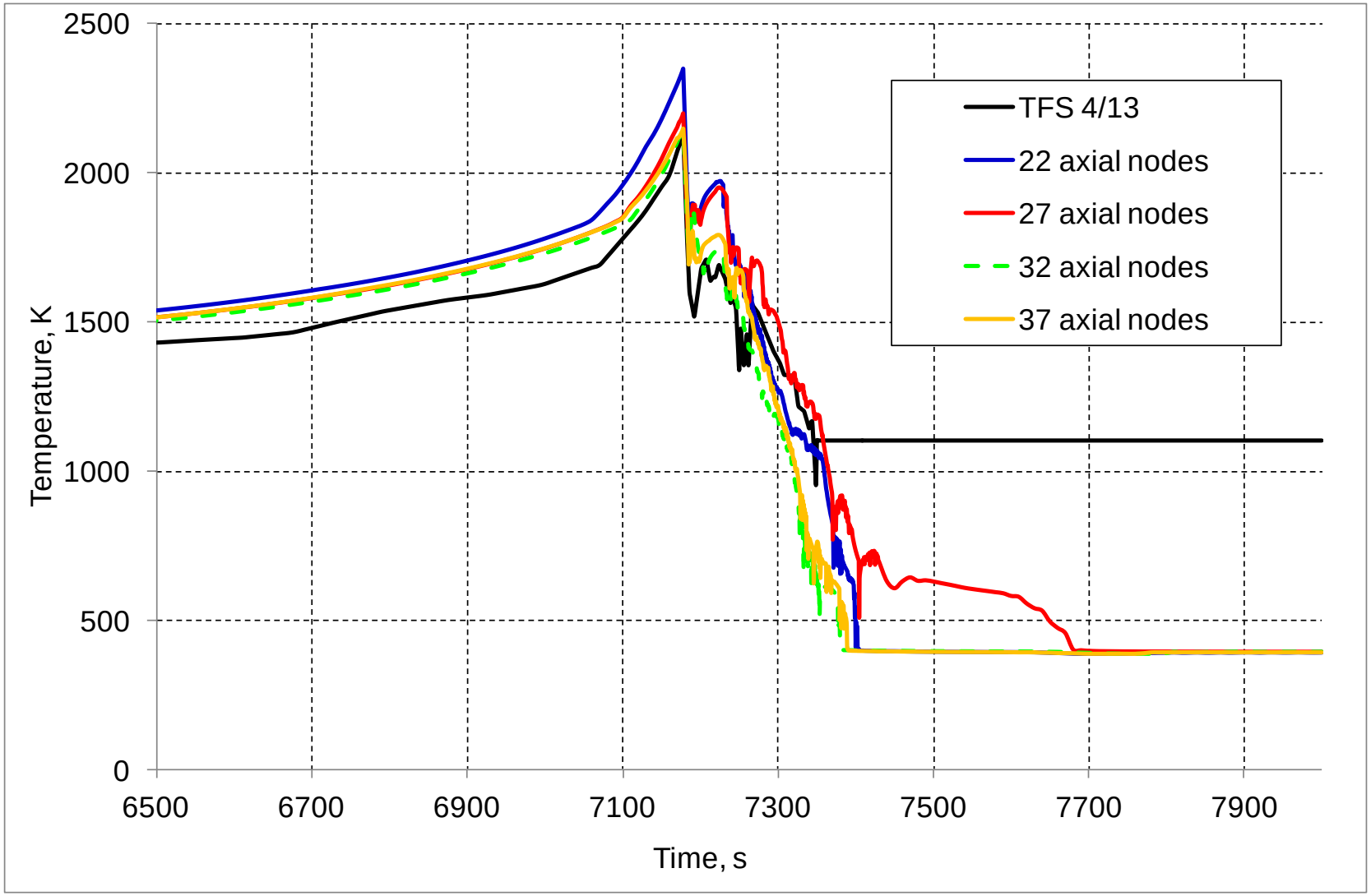
Time history of the corner rod temperature at elevation +850

RESULTS



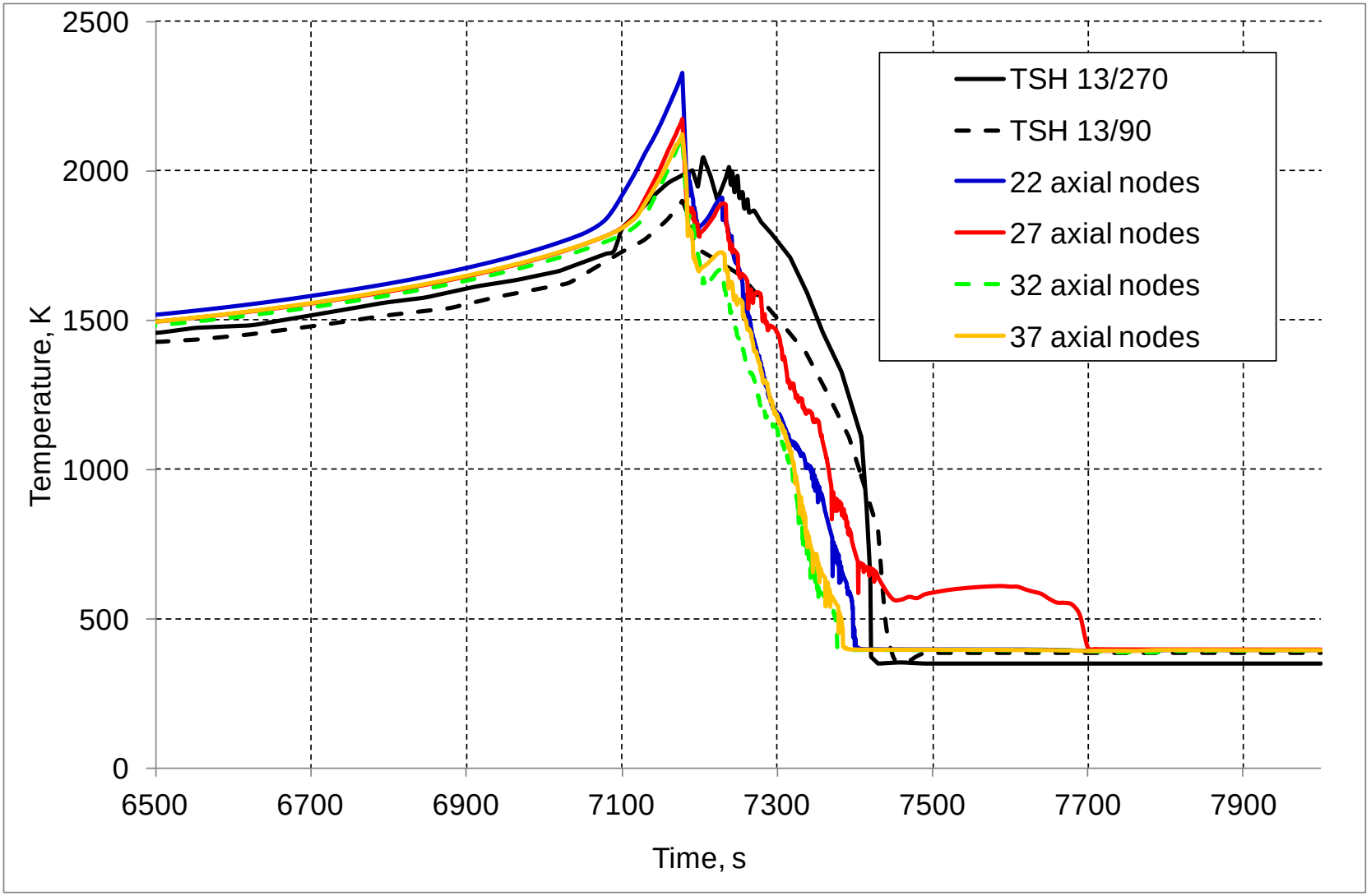
Time history of the central rod temperature at elevation +950

RESULTS



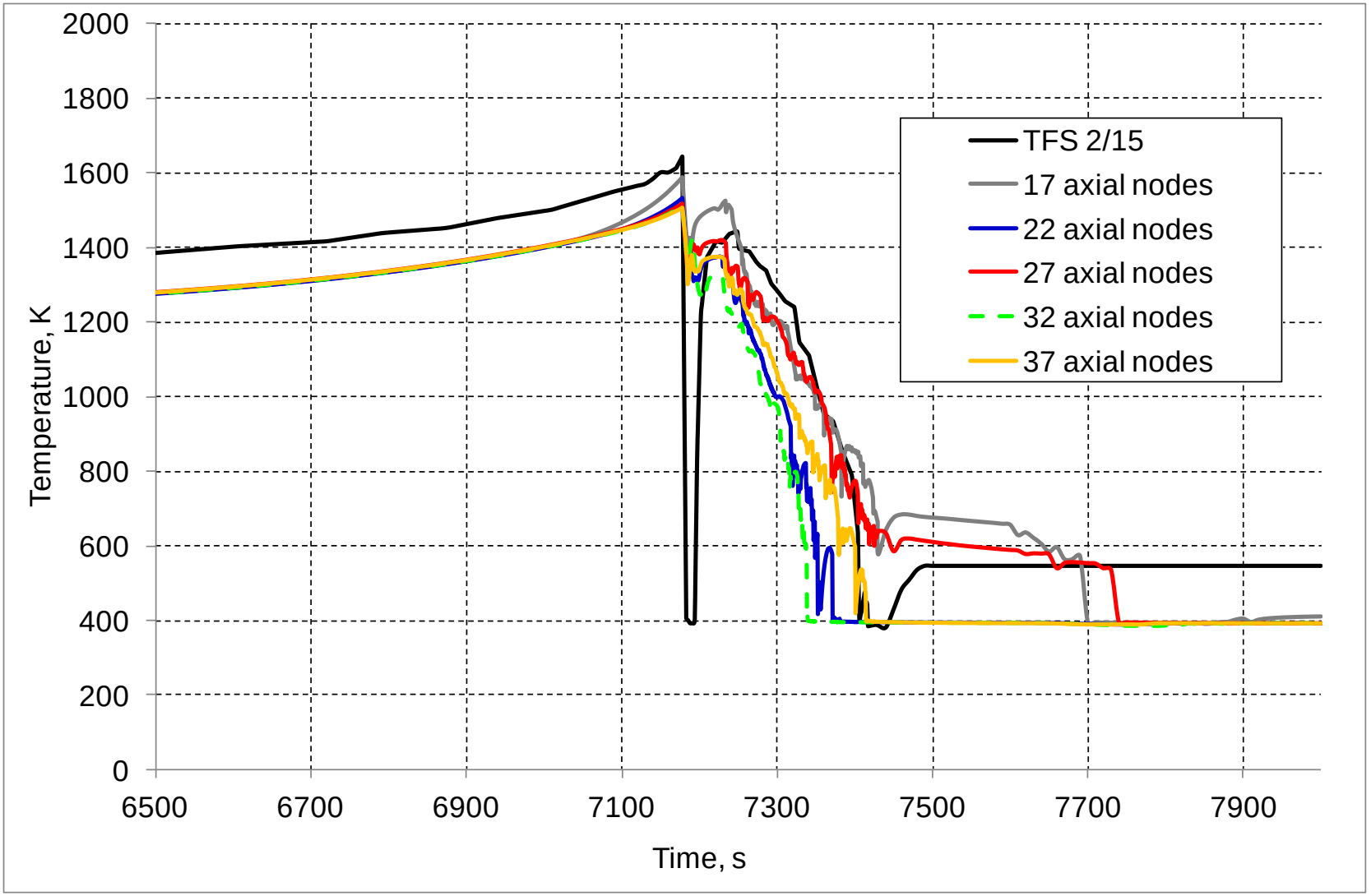
Time history of the outer rod temperature at elevation +950

RESULTS



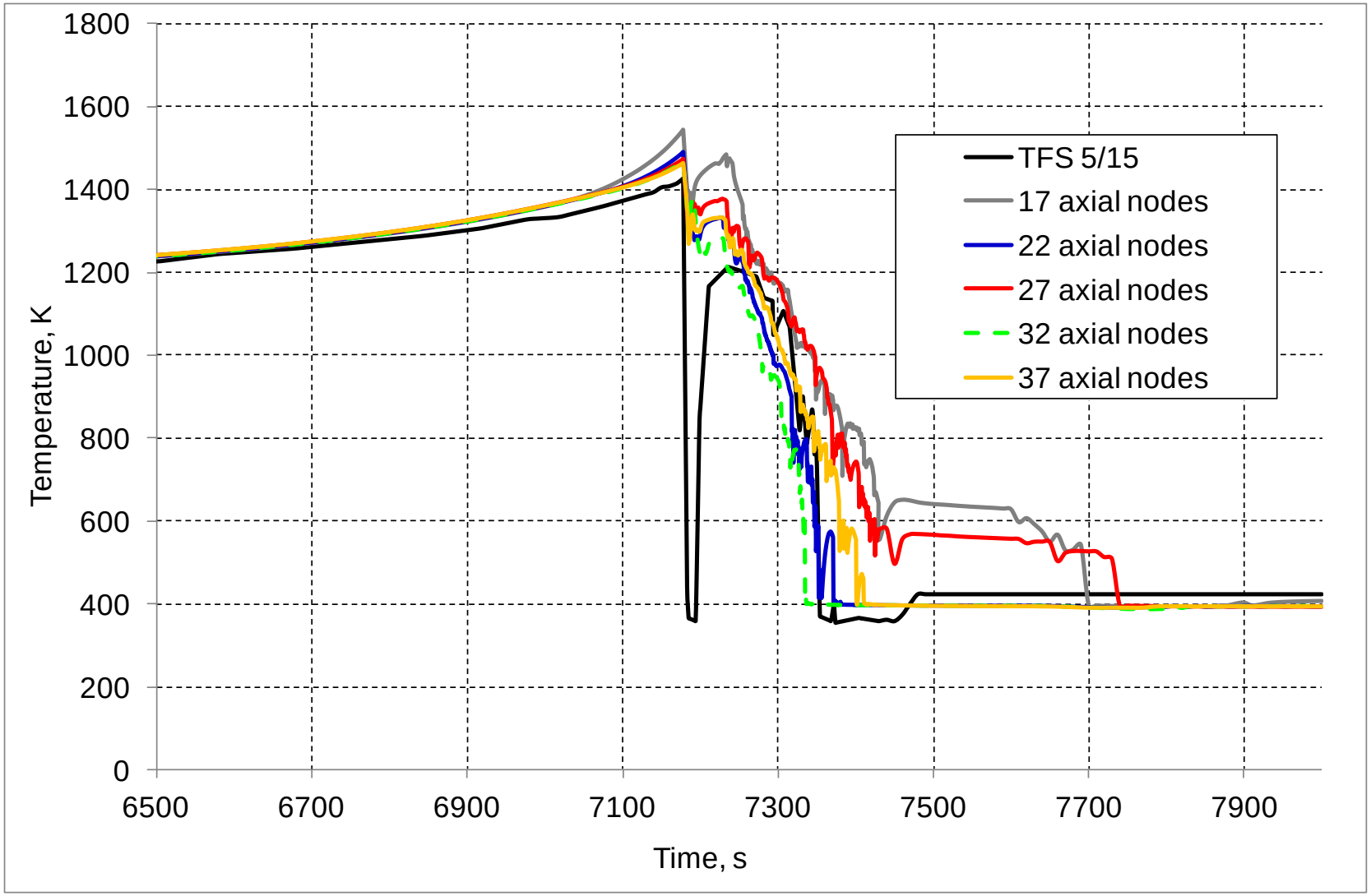
Time history of the shroud rod temperature at elevation +950

RESULTS



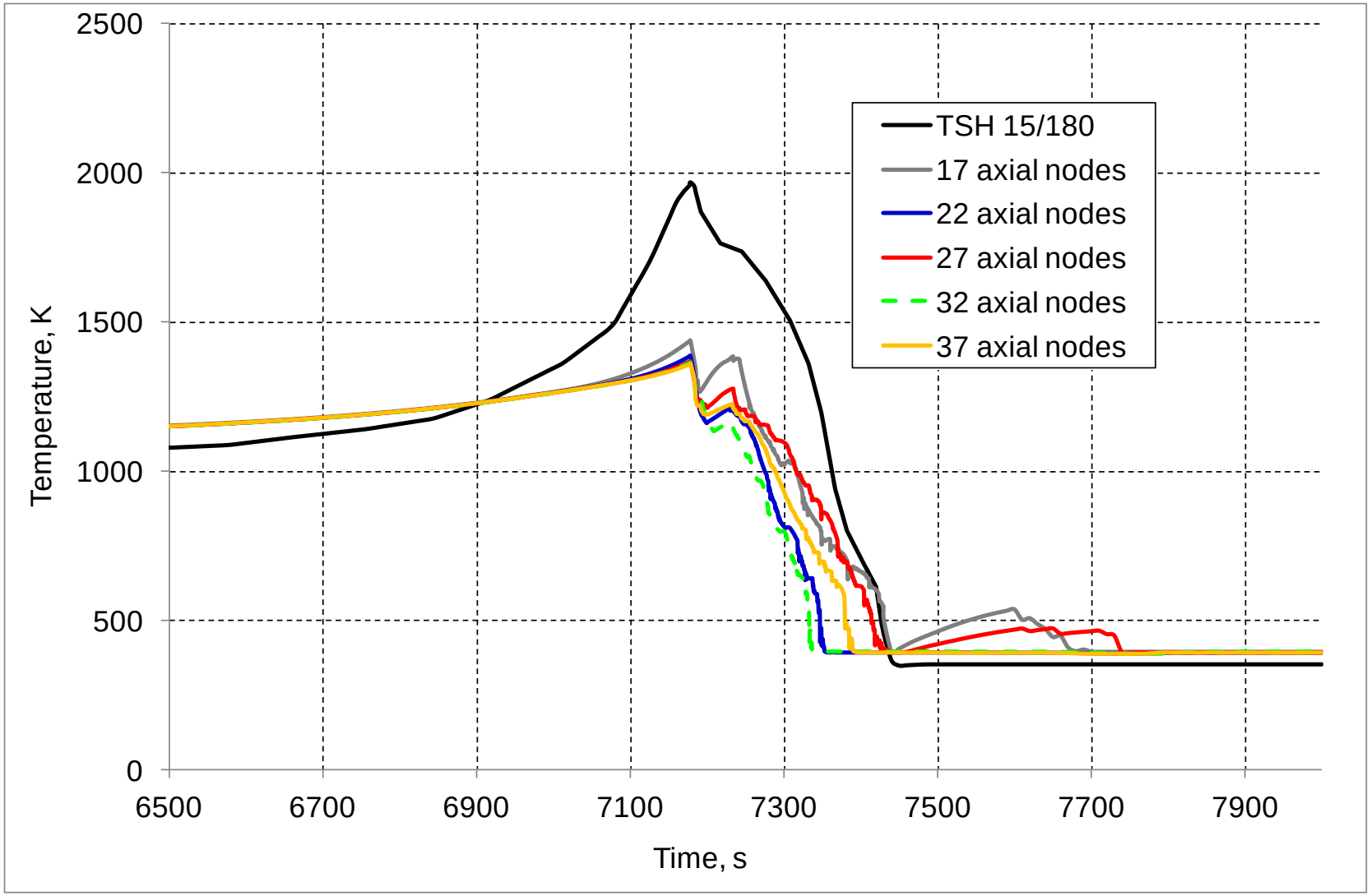
Time history of the inner rod temperature at elevation +1150

RESULTS



Time history of the outer rod temperature at elevation +1150

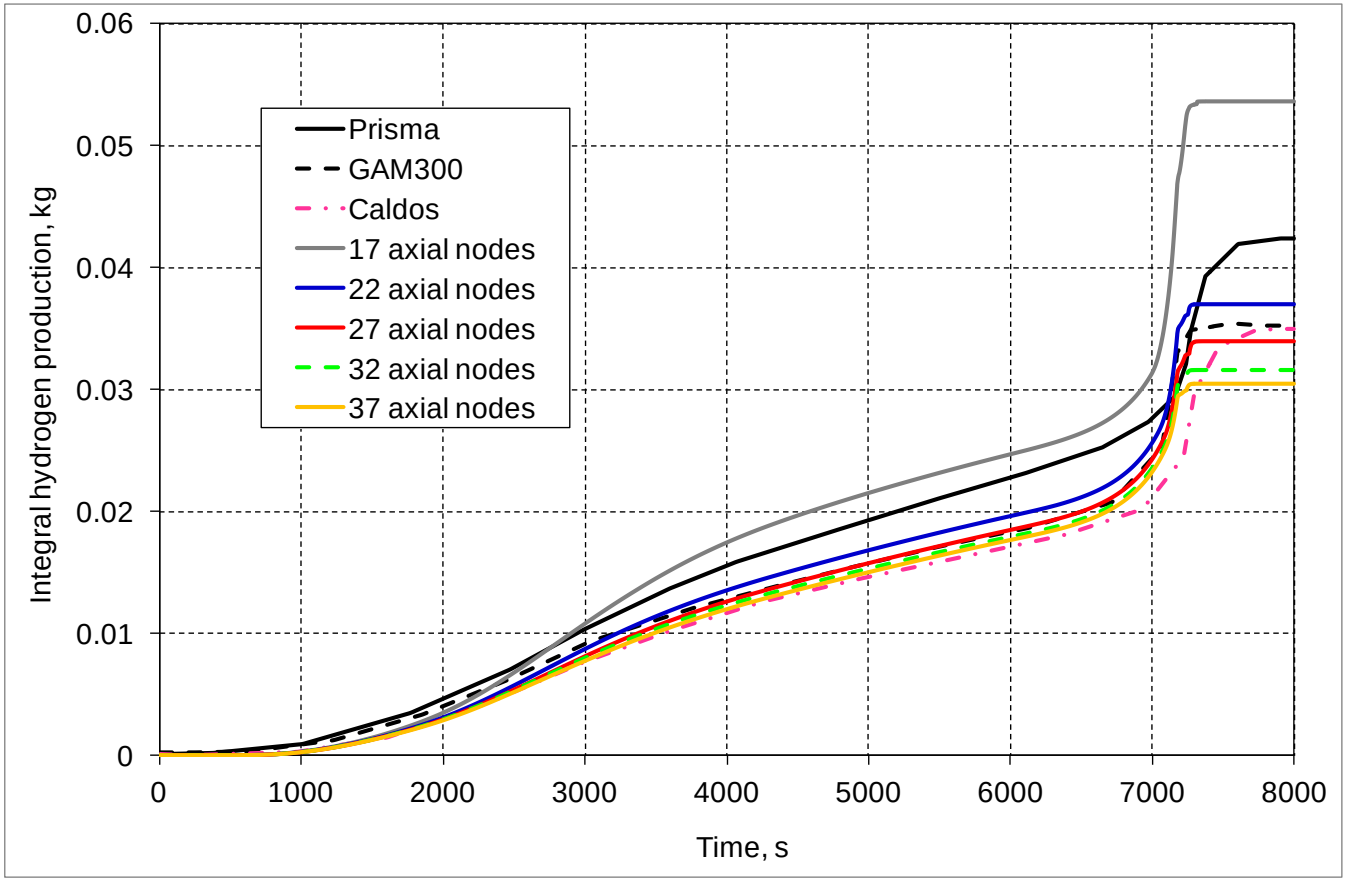
RESULTS



Time history of the shroud rod temperature at elevation +1150

RESULTS

	Measured Prisma	Measured GAM300	Measured Caldos	17 nodes	22 nodes	27 nodes	32 nodes	37 nodes
H, kg	0.0424	0.0352	0.0349	0.0537	0.0370	0.0339	0.0316	0.0305



Time history of the integral hydrogen production

CONCLUSIONS

- 5 QUENCH-06 models with different axial dicretization were tested
- There is a good agreement between the measured and predicted temperatures for all phases of the experiment, as the maximum deviation is about 100 ˚C. The exception is the shroud temperature at elevation 1150 mm, which is underestimated by all models;
- The location of cladding and shroud failure is located more precisely with models with axial discretization of the heated part of the bundle greater than 11 nodes (22 axail nodes).
- At the same time the increasing the number of nodes reduces the predicted hydrogen, as the model with 27 nodes underestimates with 4 gram the measured hydrogen from GAM300 and Caldos
- It can be assumed that sampling between the 20 and 27 nodes is optimal

THANK YOU FOR YOUR ATTENTION

Assessment of the lower head molten pool analysis model in RELAP/SCDAPSIM using LIVE experiments

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Abstract

The COUPLE module, the lower head molten pool analysis model included in a reactor analysis code RELAP/SCDAPSIM, is used to calculate the heat-up of reactor core material that slumped into the lower head of the reactor vessel. Heat transfer in the radial and axial directions to the wall structures and water surround the debris is considered in the module. Due to the recent interest in in-vessel melt retention (IVR) as a key strategy against core-melt severe accidents, the ability and applicability of the module need to be assessed and improved. Despite the recent increase of interest on IVR, few validation works have been conducted comparing with experiments. In the current study, the numerical results are compared with the LIVE (Late In-Vessel Phase Experiments) test series at Karlsruhe Institute of Technology (KIT) that investigate the thermal behavior of melt pools in the reactor pressure vessel (RPV).

The heat flux, wall temperature, crust thickness and melt temperature are compared with the experimental data. The code was modified to allow modelling external cooling vessel and an input deck for the LIVE facility is created. The results using other empirical correlations are also compared and, as a result, the better agreement of heat flux and wall temperature is obtained.


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Assessment of the lower head molten pool analysis model in RELAP/SCDAPSIM using LIVE experiments

Hiroshi Madokoro, Alexei Miassoedov

Karlsruhe Institute of Technology (KIT)

IKET -Institute for Nuclear and Energy Technologies

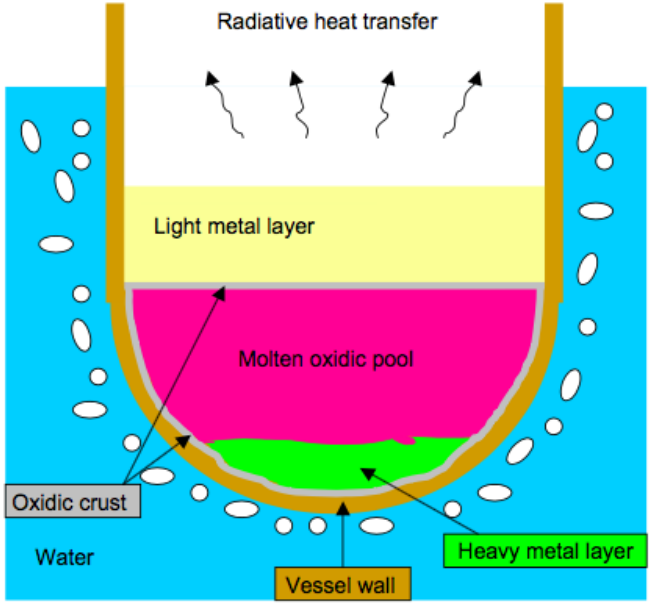


Contents

- Background
- Description of COUPLE
- Description of LIVE experiment
- Results
  - LIVE-L7V analysis
    - Change 1: multiple volumes for the external cooling
    - Change 2: other correlations from recent studies
  - LIVE-L1 analysis
    - Change 3: consideration of convective + radiation heat transfer on inner vessel wall above melt surface
- Conclusion

Background

- Relocation of molten core materials leads to molten pool formation in lower head of RPV
  - Due to thermal and mechanical loads, RPV will fail unless any accident measure is undertaken
- In-vessel Melt Retention (IVR) through external reactor cooling is a key severe accident management strategy for LWRs



* B.R. Sehgal, Nuclear Safety in Light Water Reactors, 2012.

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Background

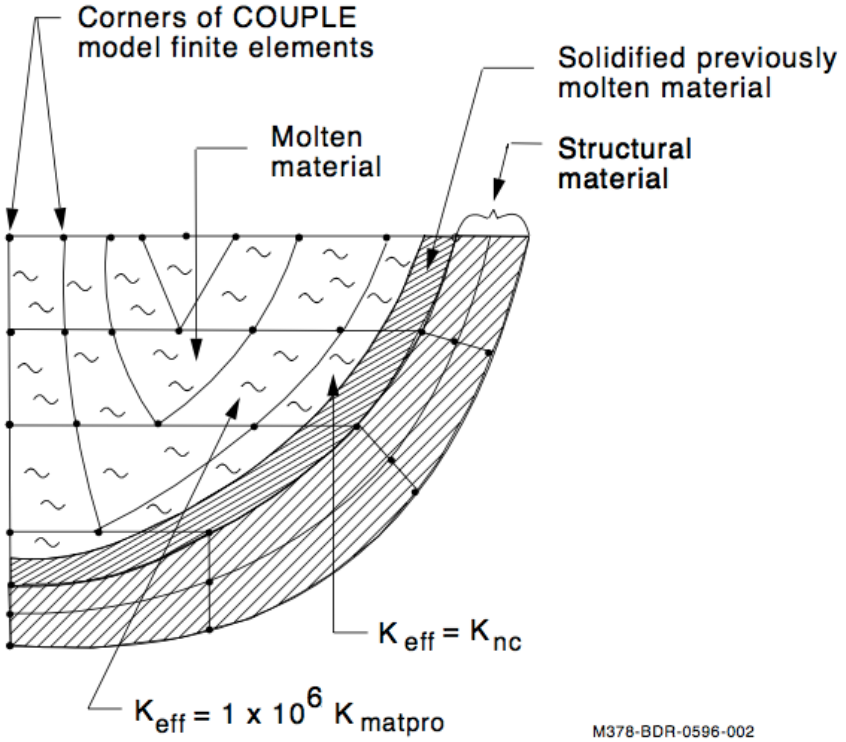
- RELAP/SCDAPSIM is widely used for severe accident analysis
  - RELAP5: thermal hydraulics
  - SCDAP: core damage and degradation
  - COUPLE: lower head heat transfer

- Few assessment studies have been conducted on COUPLE
- Knowing capability of COUPLE and its improvement is of importance

- Assessment work should be conducted using lower head molten pool experiments
  - LIVE-L1 and -L7V tests are calculated

Description of COUPLE

- COUPLE is a two-dimensional finite element heat conduction model
- Effective conductivity
  - Represent the heat transfer in molten pool where heat transfer is dominated by natural convection
  - Uniform molten pool temperature
  - At liquid-solid interface, heat transfer correlations are used
    - $Nu_{up} = 0.36Ra_i^{0.23}$
    - $Nu_{down} = 0.54Ra_i^{0.18}$

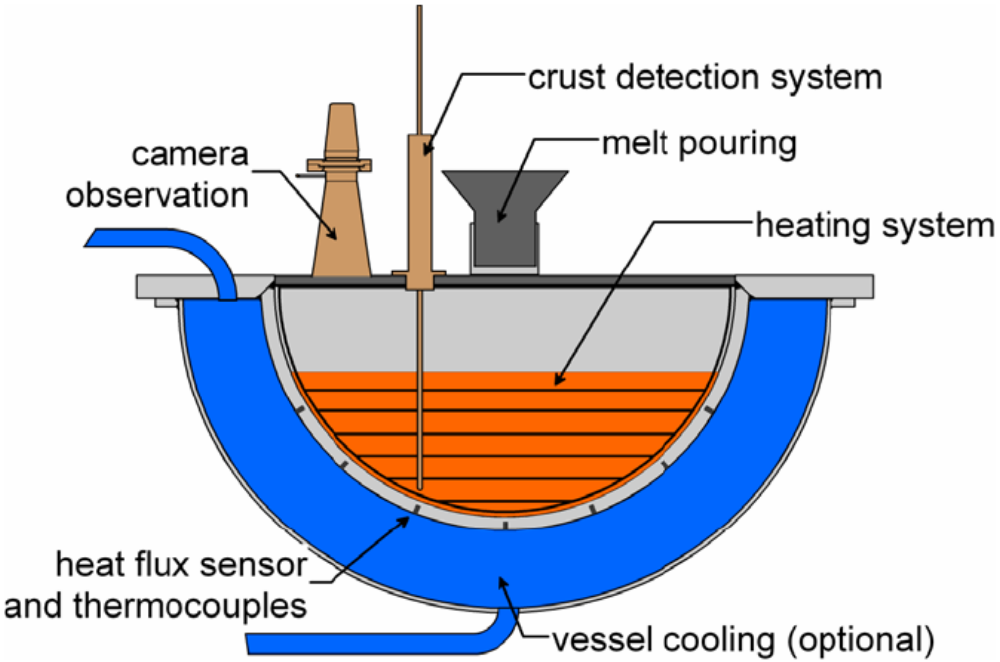


* The SCDAP/RELAP5 Development Team, NUREG/CR-6150, INEL-96/0422, Revision 1 Volume II, U.S. Nuclear Regulatory Commission, 1997.

Description of the LIVE experiment

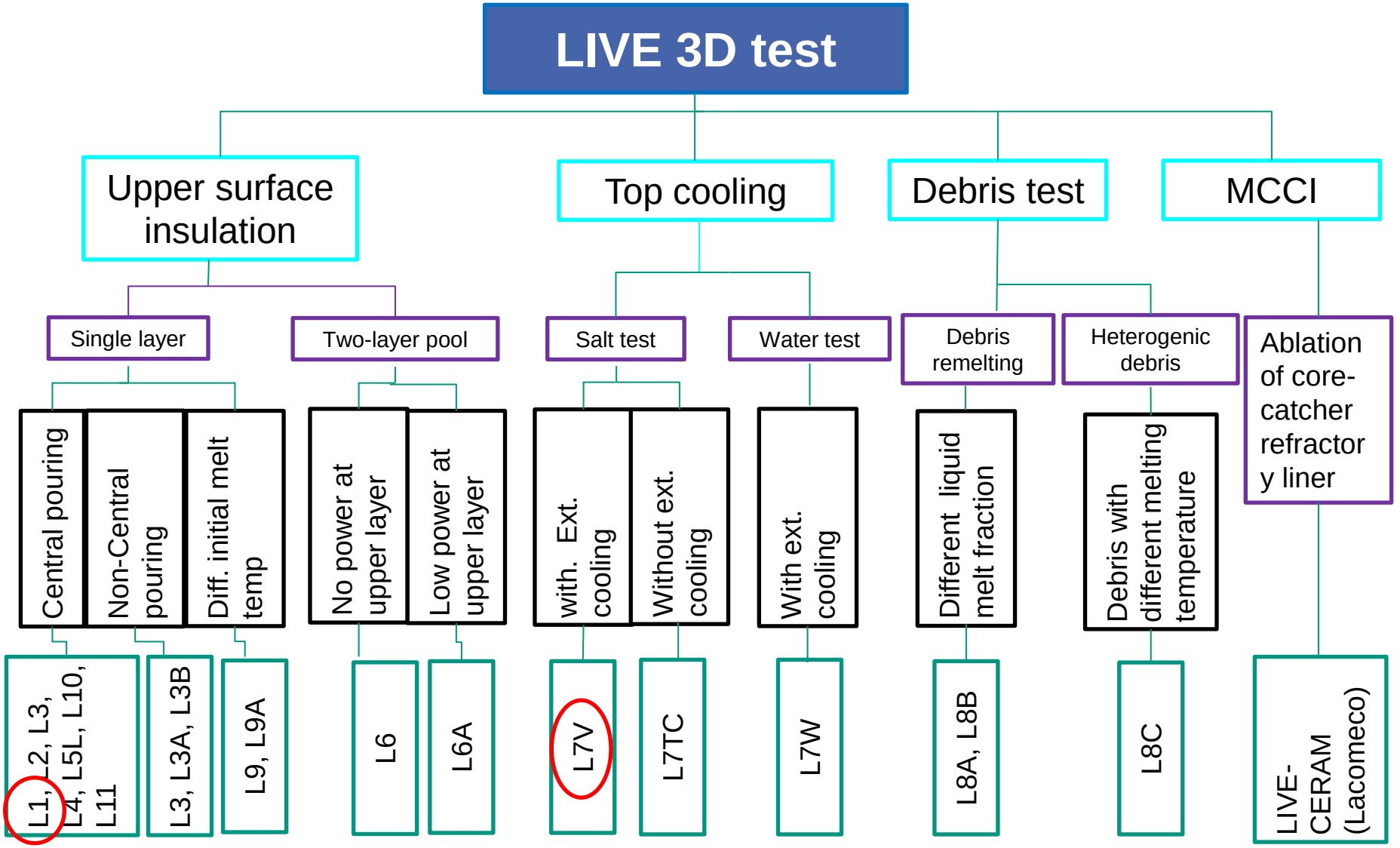
- LIVE test series at Karlsruhe Institute of Technology (KIT)
  - Investigate thermal behavior in RPV
  - 1/5 PWR RPV
  - Volumetric heating system to simulate decay heat
  - Simulant debris: 80%KNO_3-20%$NaNO_3$
    - Solidus temp. ~220°C
    - Liquidus temp. ~280°C





* B. Fluhrer et al, “The LIVE-L1 and LIVE-L3 Experiments on Melt Behavior in RPV Lower Head,” FZK, FZKA-7419, 2008.

Description of LIVE experiment

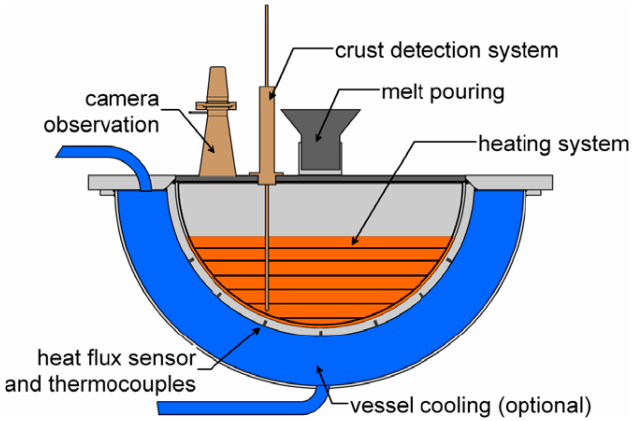


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Description of the LIVE experiment

- Test matrix

	Upper boundary	Sidewall cooling	Power level	Note
L1	Insulation	Air → Water	10kW 7kW	
L7V	Cooling lid	Water	29kW 24kW 18kW 9kW	Water inlet/outlet temperature difference 10ºC

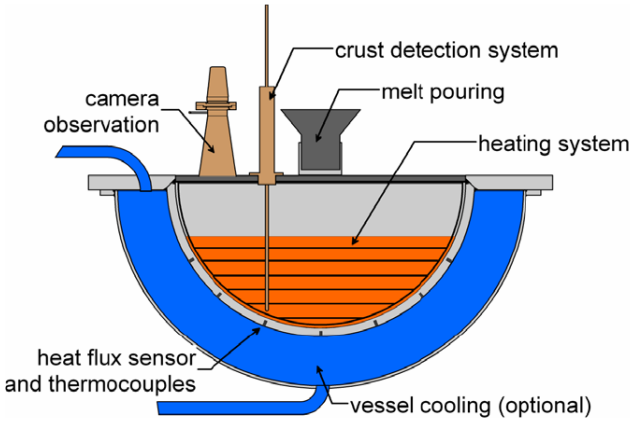


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Description of the LIVE experiment

- Test matrix

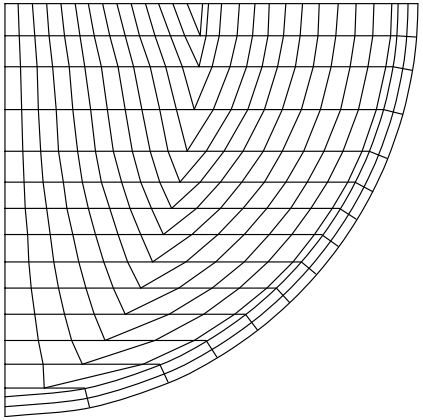
	Upper boundary	Sidewall cooling	Power level	Note
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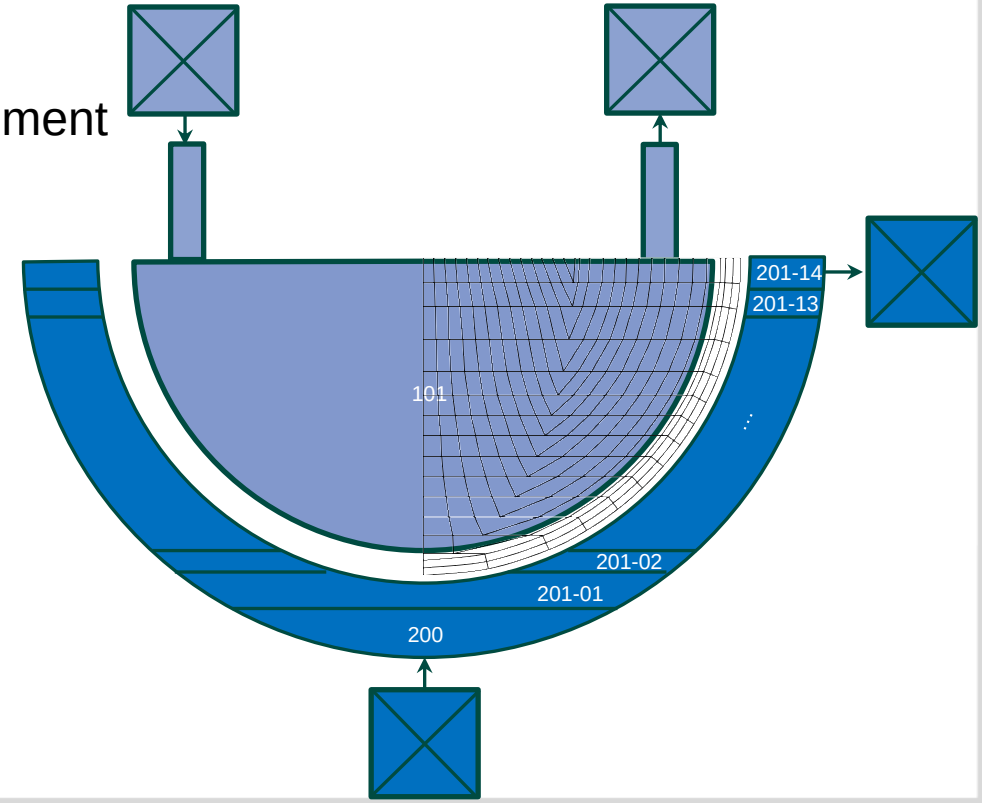
LIVE-L7V analysis

- **Change 1: multiple volumes for the external cooling**
- Original model
  - 1 vol. for the containment, Saturated water
  - Multiple volumes, convection heat transfer
- Modified model
  - Multiple volumes for the containment
  - Convection + radiation



COUPLE nodalization

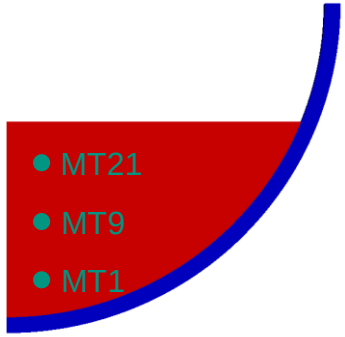
RELAP5 nodalization

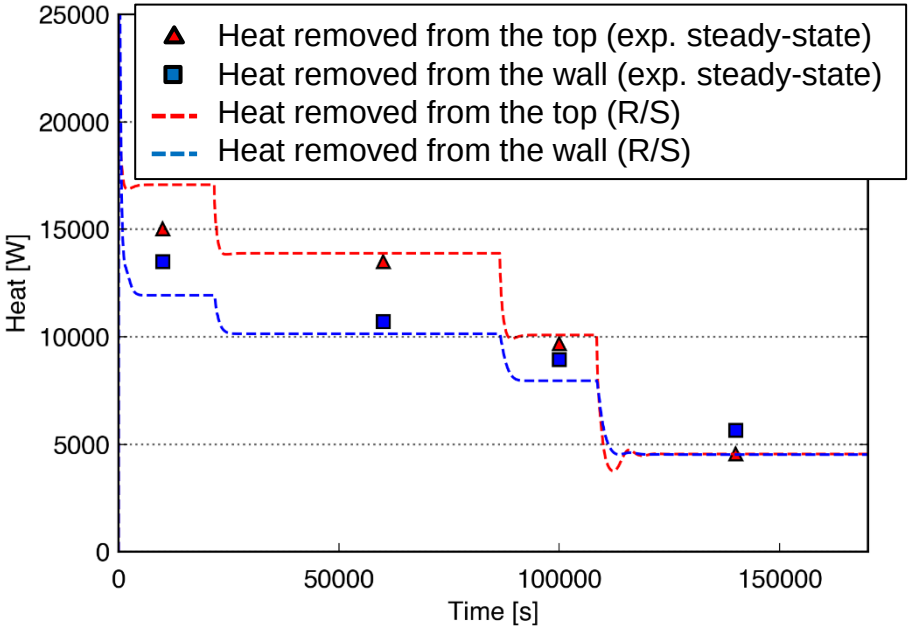


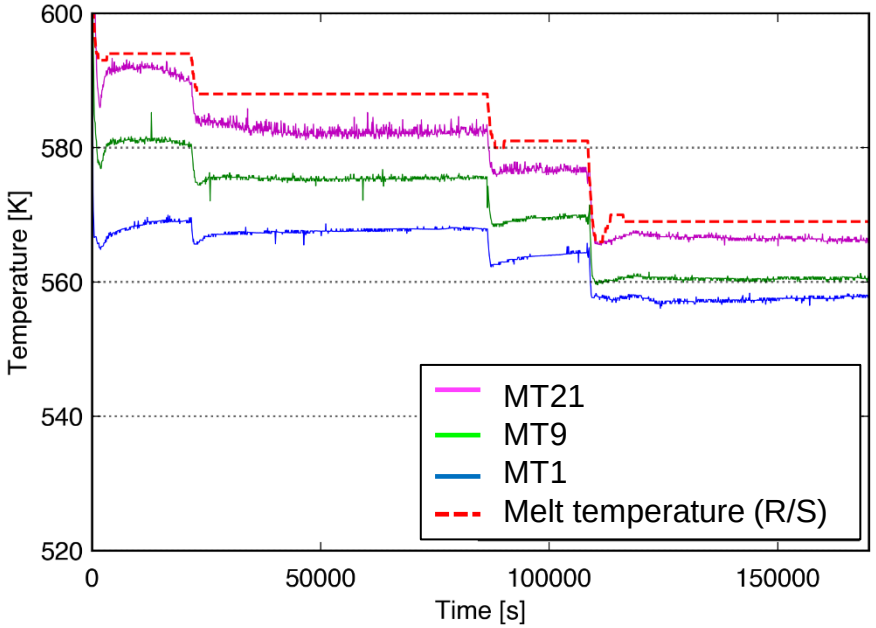
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LIVE-L7V analysis

- Cooling condition
  - Water cooling lid is installed above the melt surface
  - External wall is constantly cooled by supplied water
- Removed heat from melt surface and wall agree well with the experiment
- Melt temperature is slightly overestimated





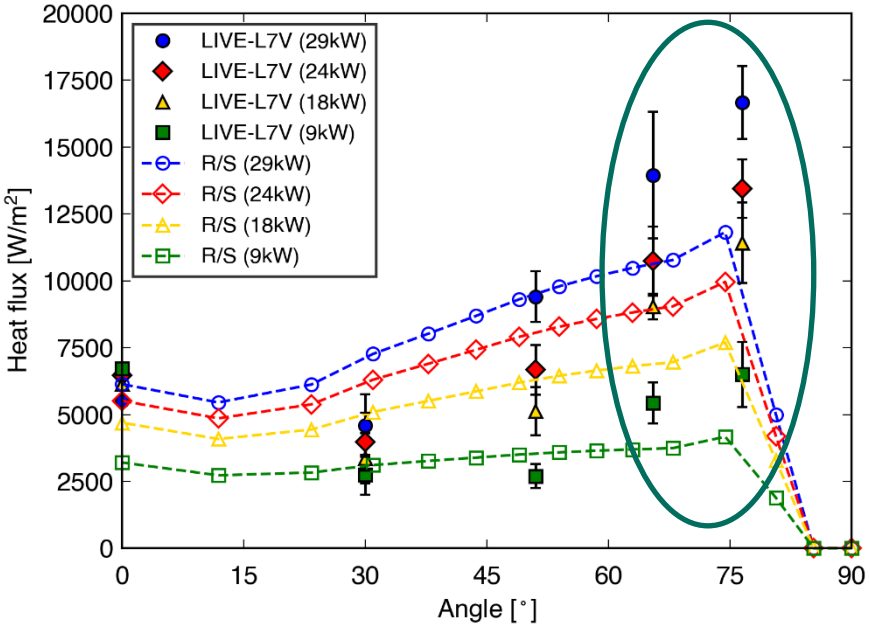


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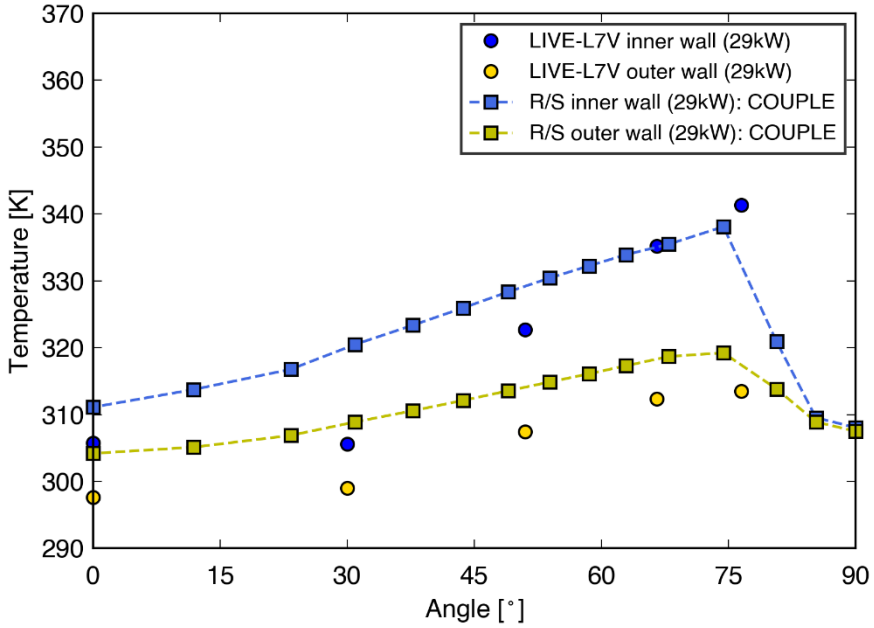
LIVE-L7V analysis

- Heat flux and temperature along the vessel wall are compared in each steady state
  - Heat flux is underestimated at higher region of the vessel
  - Calculated wall temperature profile is rather flat compared to the measured value

Heat flux profile along the vessel wall

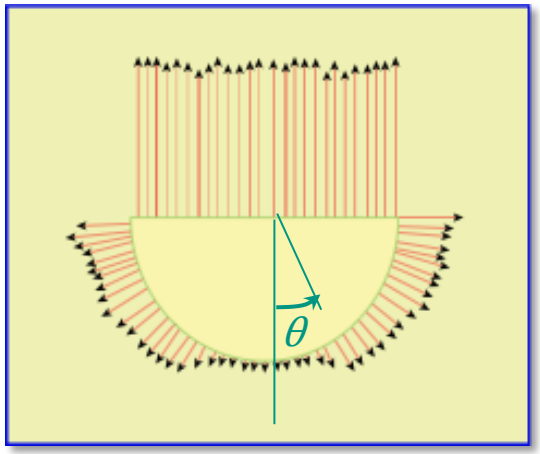


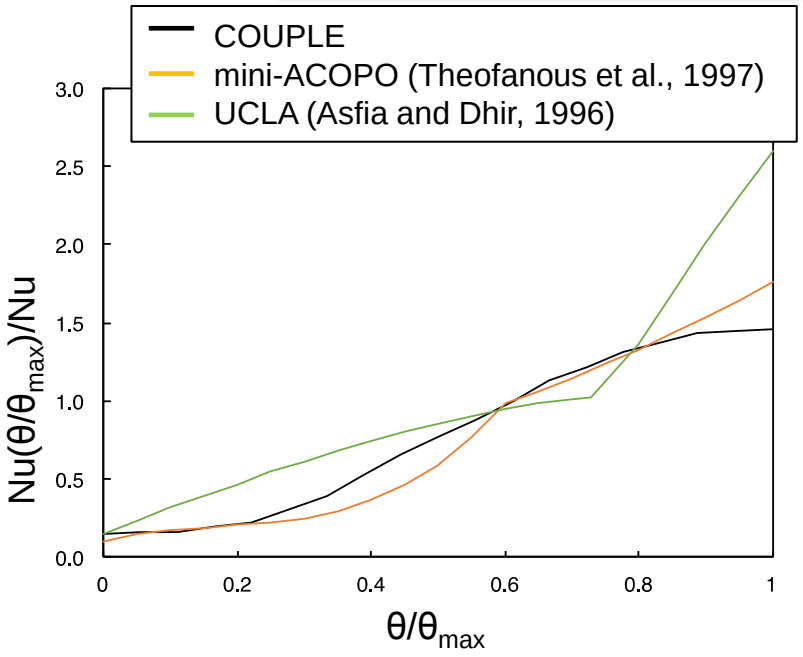
Wall temperature profile along the vessel wall at power level of 29kW



LIVE-L7V analysis

- Heat transfer along external wall is not uniform
  - Local Nu can be obtained by weighing according to normalized polar angle from the center line
- Recent studies show the maximum normalized downward Nusselt number is between 1.7 and 2.6
  - For higher Rayleigh number cases, this value can be even higher
- **Change 2: other correlations from recent studies:**
  - mini-ACOPO (Theofanous et al., 1997)
  - UCLA (Asfia and Dhir, 1996)

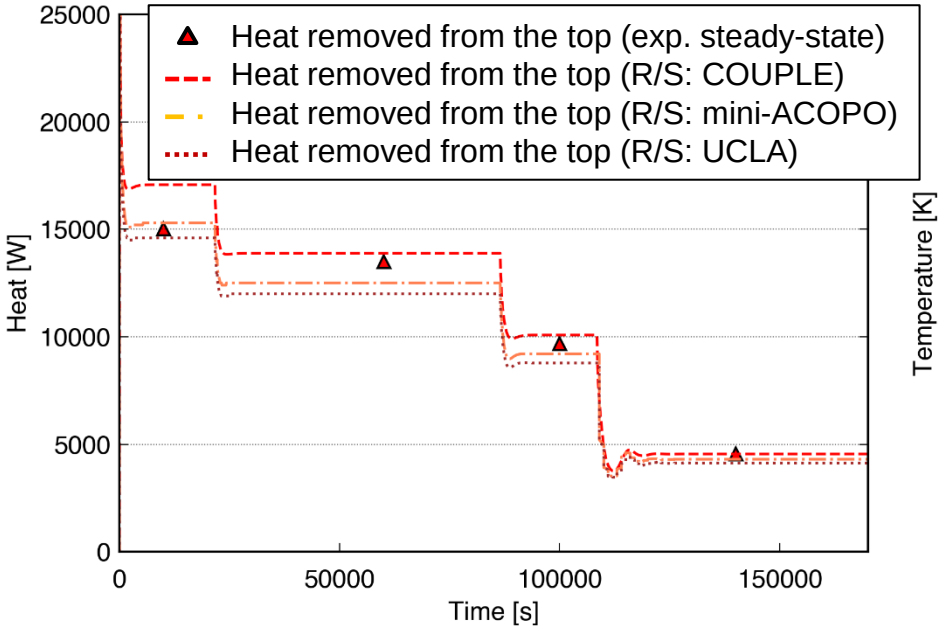


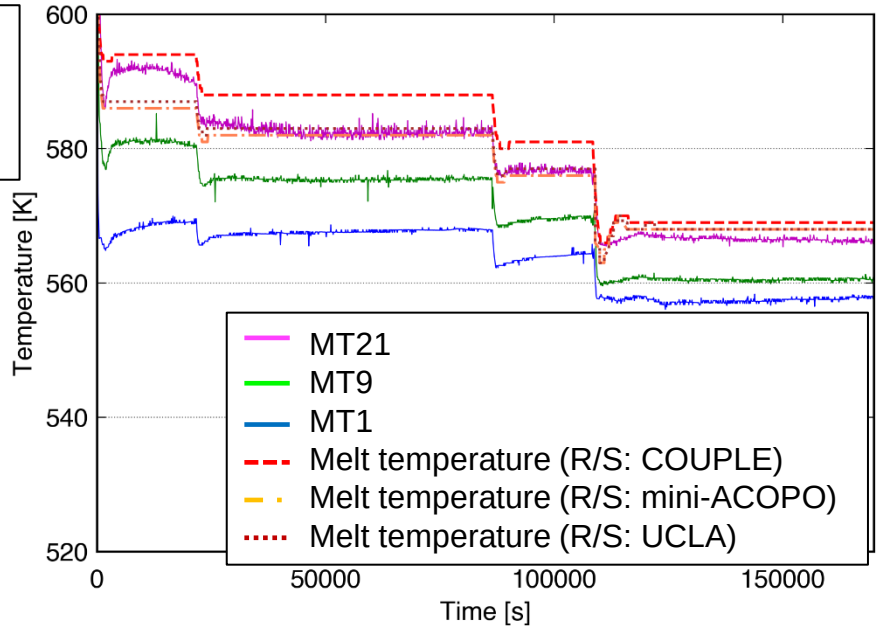


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LIVE-L7V analysis

- Comparison of different correlations

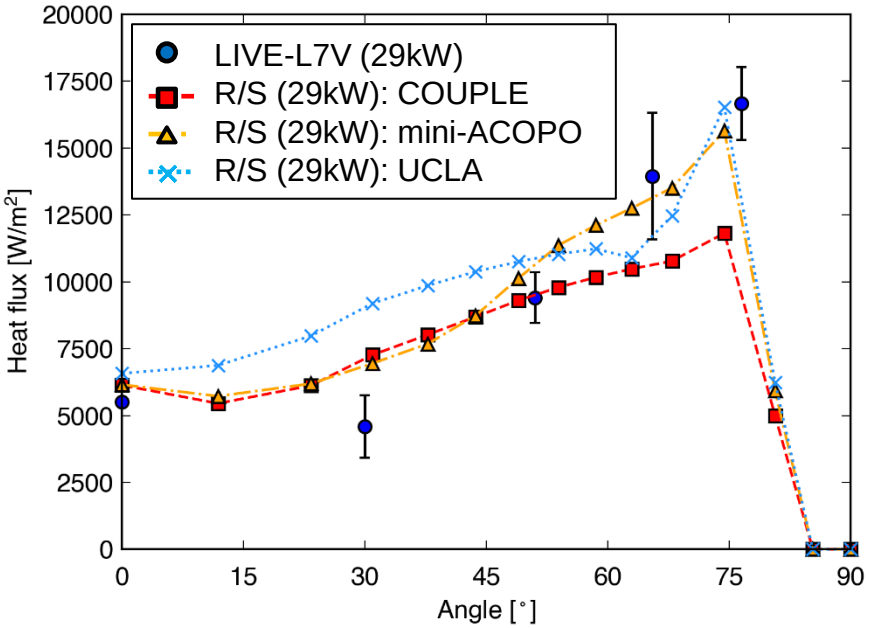


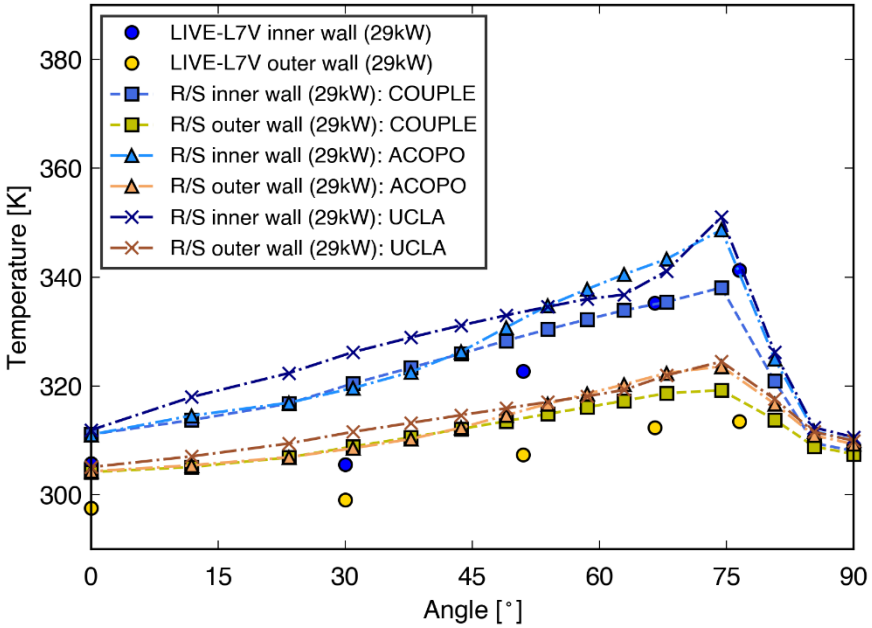


- mini-ACOPO and UCLA correlations give better results of heat removed from the top, especially at heat level of 29kW
- Melt temperatures with mini-ACOPO and UCLA are lower than the one original model

LIVE-L7V analysis

- Comparison of different correlations





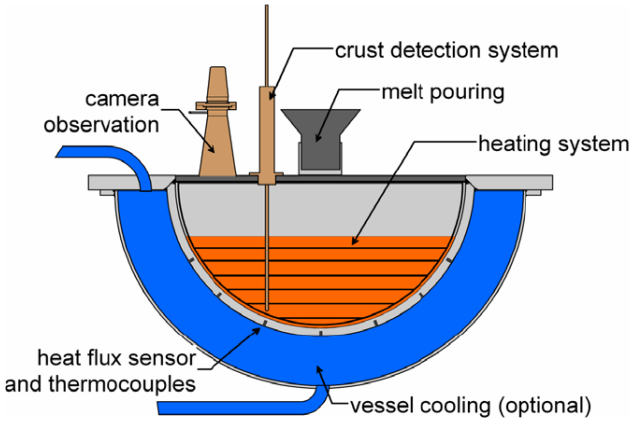
- mini-ACOPO correlation gives reasonable results
  - In the following calculation, the mini-ACOPO correlation is used

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Description of the LIVE experiment

- Test matrix

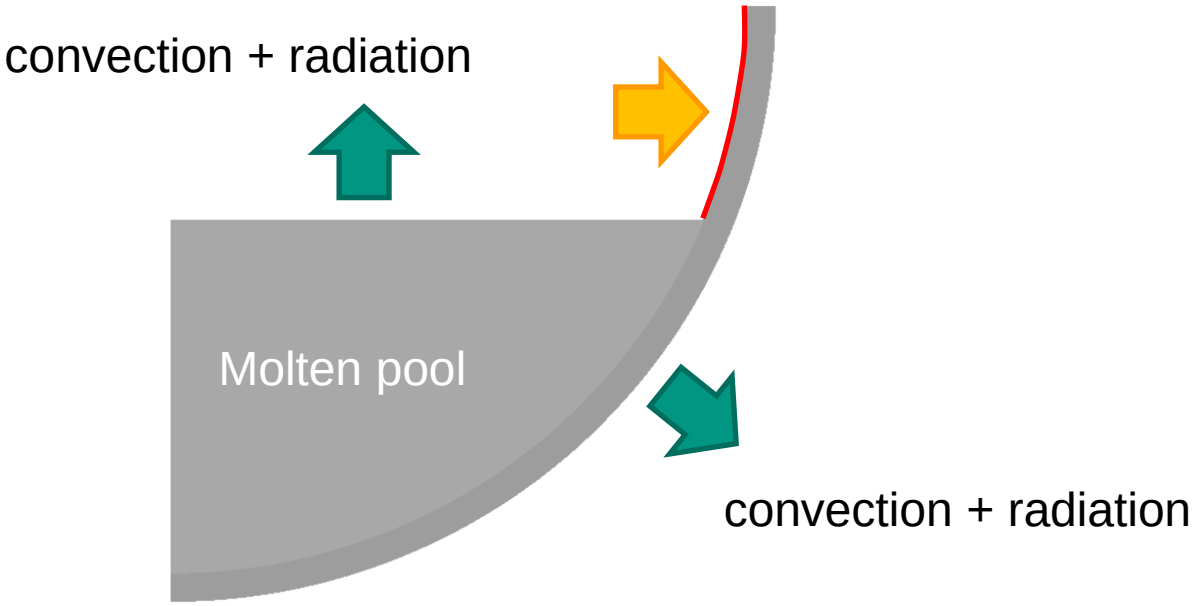
	Upper boundary	Sidewall cooling	Power level	Note
L1	Insulation	Air → Water	10kW 7kW	
L7V	Cooling lid	Water	29kW 24kW 18kW 9kW	Water inlet/outlet temperature difference 10ºC



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LIVE-L1 analysis

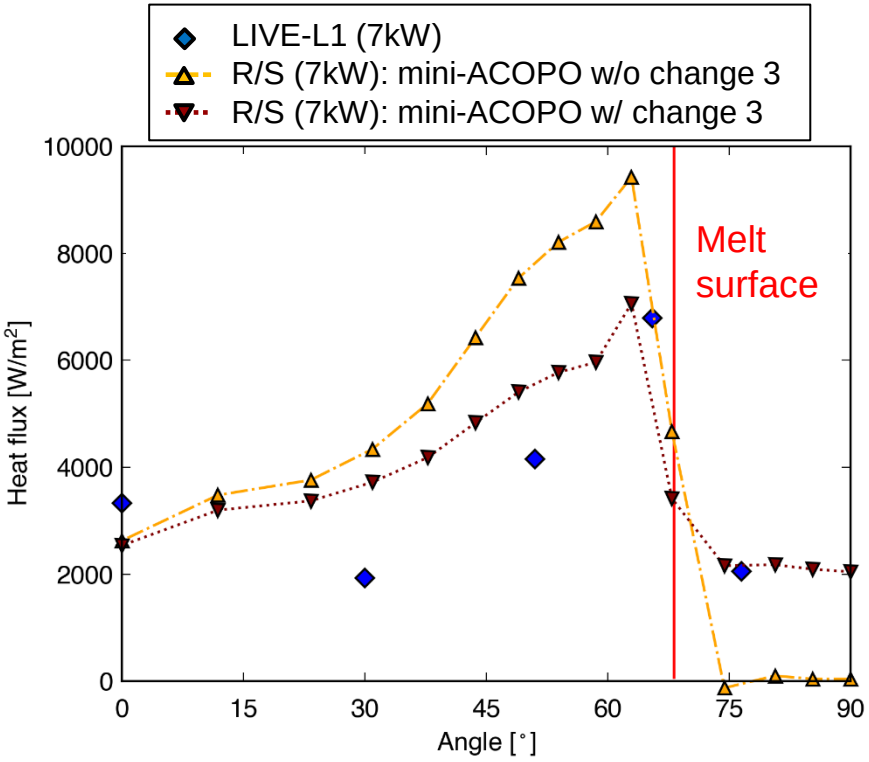
- **Change 3: consideration of convective + radiation heat transfer on inner vessel wall above melt surface**

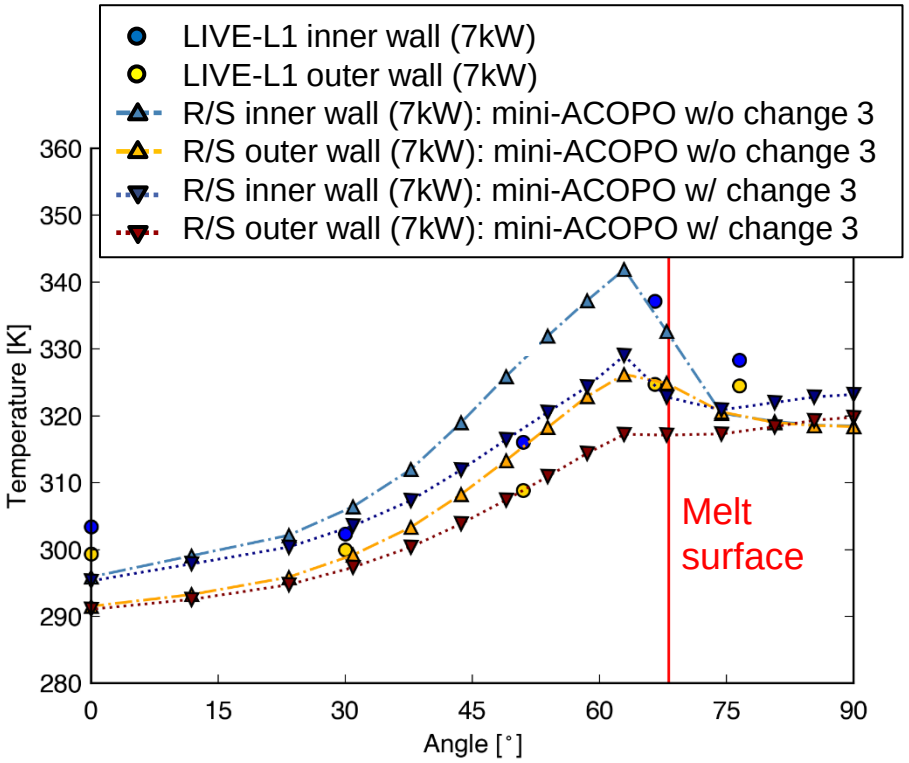


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LIVE-L1 analysis

- Comparison of original and modified models





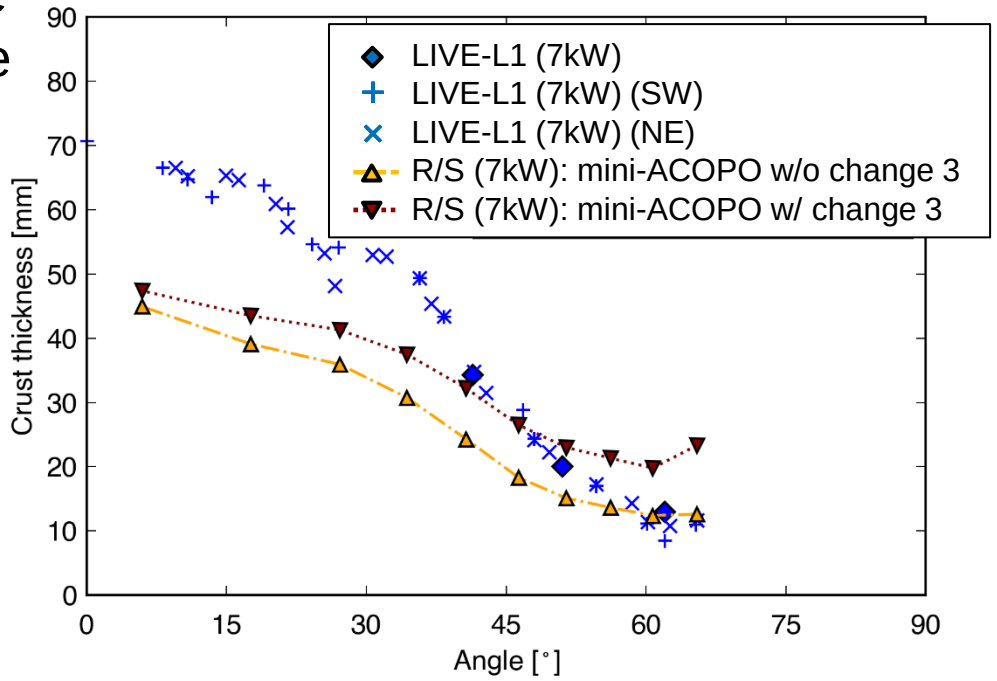
- Heat flux above the melt surface is better predicted with modified model

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LIVE-L1 analysis

- Calculated thickness near the melt surface is slightly thicker than the measured value

- Due to lower heat transfer in the crust, the inner wall temperature is underestimated in the calculation
- Further investigation on solidification calculation should be conducted



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Conclusion

- An input model for the LIVE facility is created
- Radiation heat transfer is considered for external volumes
- LIVE-L7V
  - The current ability of COUPLE is shown
  - Two other correlations for downward Nusselt number from recent studies have been tested →mini-ACOPO correlation gives reasonable results
- LIVE-L1
  - Convective and radiation heat transfer on the inner vessel wall above the melt surface is now considered
  - Crust thickness affects the wall temperature due to lower heat transfer in the crust
    - Difference is significant at the top corner of molten pool
  - Solidification model needs further investigation

❑ Assessment using other experiments

❑ Application to real-scale reactor analysis

Karlsruher Institut für Technologie

Thank you for your attention!

For further questions:

hiroshi.madokoro@kit.edu

Appendix

- Empirical correlations for upward and downward Nusselt number
- COUPLE default (Mayinger et al., 1975)
  - $Nu_{up} = 0.36 Ra_i^{0.23}$
  - $Nu_{down} = 0.54 Ra_i^{0.18}$
- mini-ACOPO and ACOPO (Theofanous et al., 1997)
  - $Nu_{up} = 1.95 Ra_i^{0.18}$
  - $Nu_{down} = 0.3 Ra_i^{0.22}$
- UCLA (Asfia&Dhir, 1996)
  - $Nu_{up} = 0.403 Ra_i^{0.226}$
  - $Nu_{down} = 0.55 Ra_i^{0.2} (H/R)^{0.25}$

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Investigation of thermodynamic phases of Ag-Cd-In system at high temperature and its assessment by CALPHAD

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[a] Institut de Radioprotection et Sureté Nucléaire, B.P. 3, 13115 Saint Paul-lez-Durance Cedex, France.
[b] IM2NP, UMR7334, CNRS, Aix-Marseille Université, Campus de Saint Jérôme, Avenue Escadrille Normandie Niémen – Case 251, 13397 Marseille Cedex 20, France.
[c] Université Grenoble Alpes, CMTC, SIMAP, 38000 Grenoble, France.

The 80 wt.% Ag, 15 wt.% In, 5 wt.% Cd alloy is used as absorber material in most PWR reactors. During a severe accident, the control rod failure occurs early during the sequence. At 1200°C, Cd vapour and helium present in the rod cause internal pressurization leading to rupture of the control rod. As a result, Ag-In-Cd vapours are released into the core. They may interact with the fission products and may significantly affect their speciation and transport in the primary circuit reactor coolant system and finally their behaviour in the containment. This phenomenon is of prime importance for the evaluation of source term and particularly iodine chemistry. The thermodynamic study of the Ag-In-Cd liquid phases is a necessary step for a reliable estimate of the vaporization of the chemical elements of the absorber material but also for the corium progression.

The melting point for the control rod composition was determined by several authors [1] [2] [3]. Additionally two isopleth sections of the ternary phase diagram were partly investigated by thermal analysis so far [2]. Consequently the Ag-Cd-In ternary diagram remains largely unknown, particularly for equilibria involving the liquid phase. In this work, thermal analysis (DTA) has been performed for various compositions using a Differential Scanning Calorimeter (DSC) instrument to check and extend the existing data. It is shown that the classical procedure has some limitations in determining the liquidus temperatures for the ternary compositions. To improve the accuracy of the measurements, a new DTA protocol has been applied and validated on Ag-In binary composition whose liquidus point is well established in the literature and the procedure is extended to 9 ternary compositions.

These new liquidus data as well as our previous measurements [4] of mixing enthalpy in liquid [4] are included in a CALPHAD modelling of the Ag-Cd-In ternary system. This modelling allows performing a reliable estimate of Ag-In-Cd releases in severe accident conditions. The perspectives of this work will be to extend our investigation to study the In_2O_3-ZrO_2, the Ag-Zr, In-Zr and Ag-In-Zr systems for which some preliminary results will be presented.

[1] A. N. B.R Bowsher, Ag-In-Cd Control Rod Behaviour During a Severe Accident in a Pressurized Water Reactor, High Temp. Mater. Sci., vol. 33, 1995.

[2] P. J. Horrocks, PhD Thesis, University of Manchester, Phase Diagram and Thermodynamics of the Ag–Cd–In Ternary Alloy Systems," 1991.

[3] M. Steinbrück and U. Stegmaier, "Experiments On Silver-Indium-Cadmium Control Rod Failure During Severe Accident Sequences," Annu. Meet. Nucl. Technol. 4-6 2010.

[4] P. Benigni, S. Hassam, A. Decreton, G. Mikaelian, K. Gajavalli, M. Barrachin, E; Fischer, J. Rogez, Enthalpy of Mixing in the Ag-Cd-In Ternary Liquid Phase, J. Chem. Therm. 107 (2017) 207-215



Experimental investigation of Ag-Cd-In system at high temperature

Kasi GAJAVALLI

Supervisors: Marc BARRACHIN, Pierre BENIGNI I Director: Jacques ROGEZ

IRSN– Cadarache

Partners:
IM2NP – Aix Marseille University, Marseille
SIMAP - Grenoble INP, Grenoble
SYMME- Savoie Mont Blanc University, Annecy

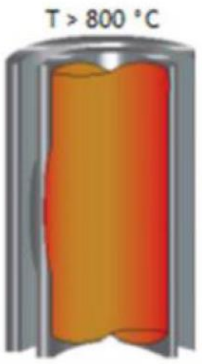
QUENCH Workshop I 17-19 October 2017 I KIT, Germany

Context : Silver-Indium-Cadmium (SIC) control rod degradation and associated releases

Ag-80%, In-15%, Cd-5%



Initial Conditions



Internal pressure increase due to He and Cd



Contact Zry-4/SS Eutectic liquefaction

T>1200°C



Material release in core and vaporization

Phenomenology of Ag-Cd-In control rod degradation in a low pressure scenario

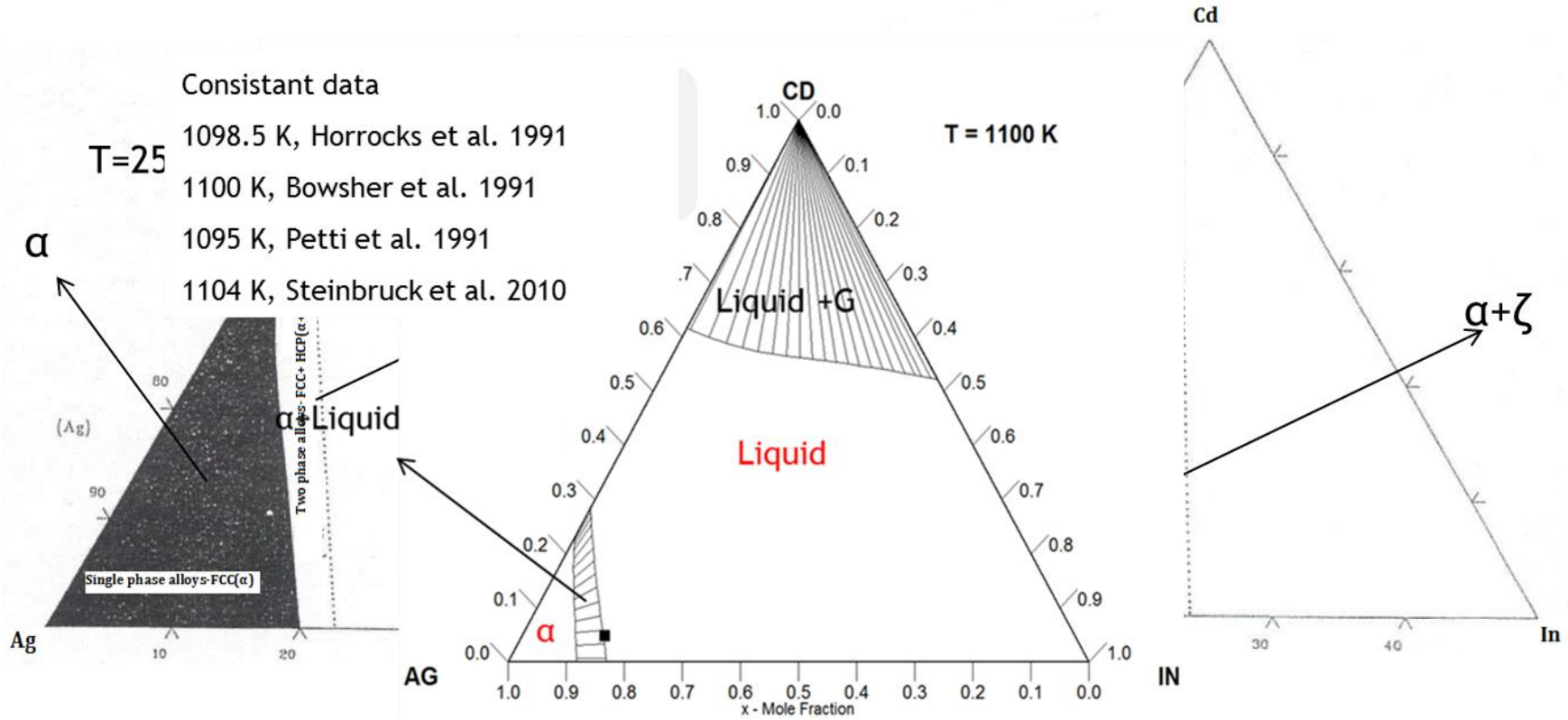
- In a severe accident, the reactor control rods partially liquefy,
- Molten SIC interacts with Zry-4 cladding due to failure of SS cladding,
- Large amount of Ag-Cd-In aerosols generated,
- Ag, Cd and In released into the primary circuit and the containment,
- Impact on the fission products and iodine chemistry.

Issue: quantification of the Ag-Cd-In releases and of their impact on FP chemistry.

State of the art of Ag-Cd-In ternary diagram

Primary solid solution phase boundary for Ag base ternary alloys (Snyder et al. 1967).

Melting points for the nominal control rod composition



Bowsher B.R., Jenkins R.A., Nichols A.L., Rowe N.A., Simpson J.A.H., United Kingdom Atomic Energy Authority, 1986.

Horrocks P.J., Thesis. University of Manchester, 1991.

Snyder, H. J., AIME Metall. Soc. Trans. 239, 1385–1391 (1967).

Petti, D. A., Nucl. Technol. 84, 128–151 (1988).

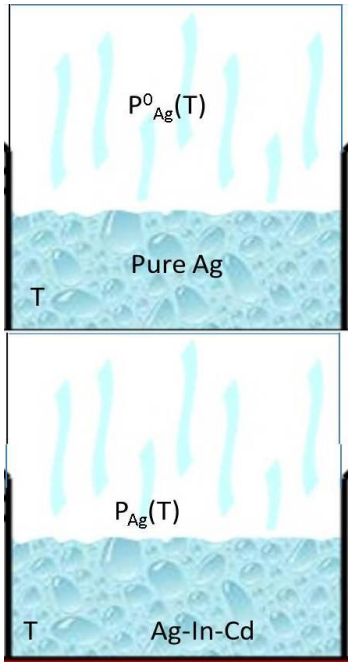
Steinbrück M., Stegmaier U., Annual Meeting on Nuclear Technology. Berliner Congress Center: Jahrestagung Kerntechnik, 2010. .

Evaluation of the SIC release : What we need ?

- Equations governing the mass releases in Ag, In, Cd (ASTEC modelling)

$$\frac{dm_i}{dt} = S\beta_i M_i \frac{P_i(T)}{RT}$$

S= evaporating surface of the melt,
β_i = mass transfer of 'i',
'i' = Ag, Cd or In,



P_i = partial pressure of 'i' in the vicinity of the melt evaporating surface

$$P_i(T) = a_i^{liq} P_0^i(T)$$

$P_0^i(T)$: pressure of pure 'i' at T,
a_i^{liq}: activity of 'i' in the liquid phase

- No thermodynamic data for Ag-Cd-In liquid phase,
- We need to measure the activity of Ag, In and Cd in liquid phase

Experimental methods

Objective : to determine the activity of Ag, In, Cd in the liquid phase

- Chemical potential of element (Ag) in liquid phase;

$$\mu_{Ag}^{liq}(T, x^{liq}) = \left[\frac{\partial G^{liq}(T, n_i)}{\partial n_{Ag}}\right]_{T, n_{In}, n_{Cd}} = \mu_{Ag}^{\circ}(T) + RT\ Ln\ a_{Ag}^{liq}(T, x^{liq})$$

$$G^{liq}(T, n_i) = \underbrace{G^{id.,liq}(T, n_i)}_{\text{Ideal mixing}} + \underbrace{G^{exc.,liq}(T, n_i)}_{\text{Excess mixing}}$$

- Measurements

$$RTLn\frac{a_{Ag}^{liq}(T,x^{liq})}{x_{Ag}} = \left[\frac{\partial G^{exc.,liq}(T,n_i)}{\partial n_{Ag}}\right]_{T,n_{In},n_{Cd}} = \underbrace{\left[\frac{\partial H^{exc.,liq}(T,n_i)}{\partial n_{Ag}}\right]_{T,n_{In},n_{Cd}}}_{\text{Calorimetry}} - T\underbrace{\left[\frac{\partial S^{exc.,liq}(T,n_i)}{\partial n_{Ag}}\right]_{T,n_{In},n_{Cd}}}_{\text{Not directly accessible}}$$

Tian-Calvet Calorimetry to measure the mixing enthalpy of liquid phase,

Differential Thermal Analysis to measure the respective stabilities of the liquid and solid phases.

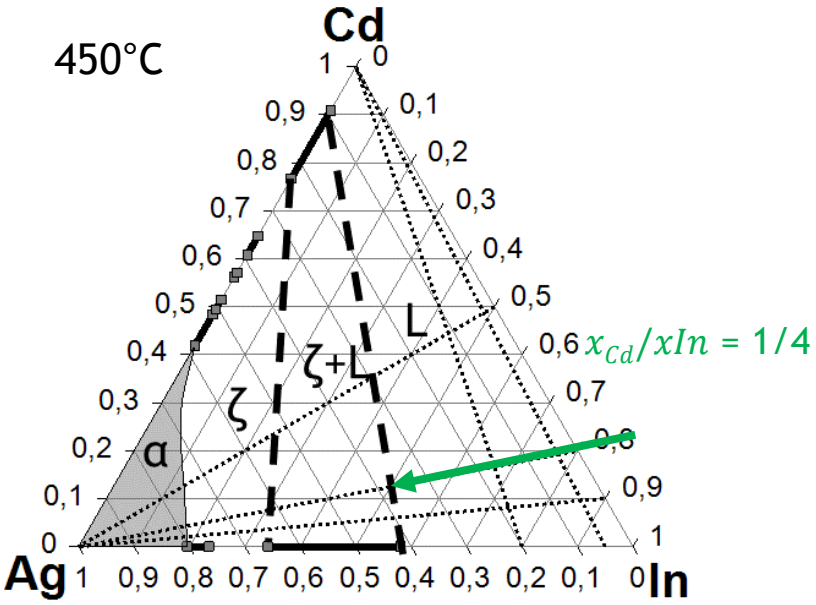
Partial enthalpy of mixing in the Ag-Cd-In ternary liquid phase

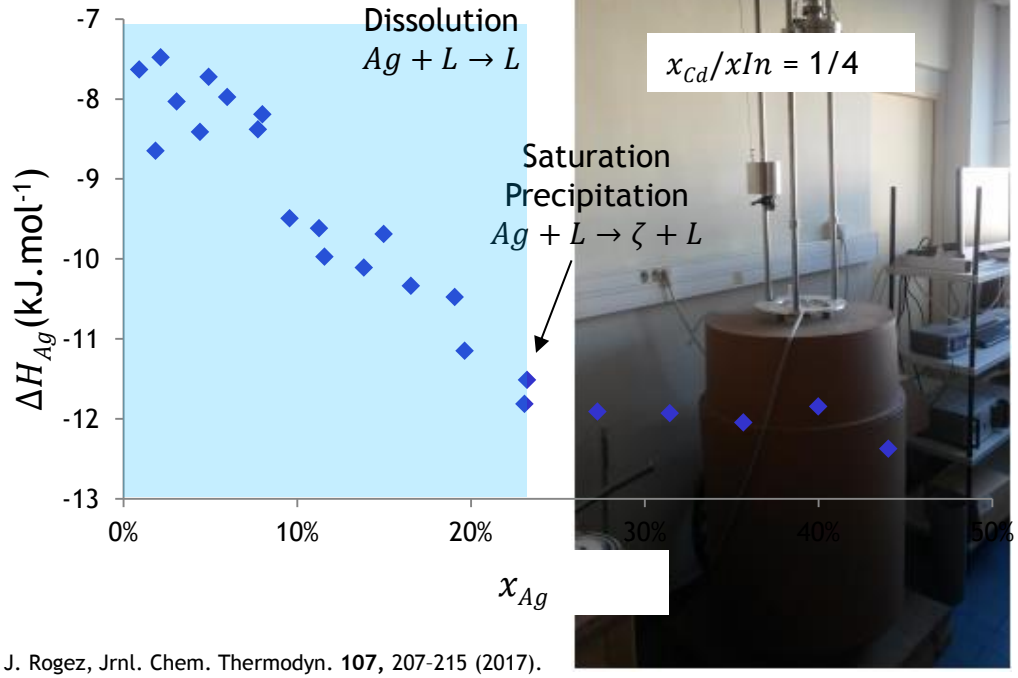
For Ag: measurement of $\left[\frac{\partial H^{exc.,liq}(T,n_i)}{\partial n_{Ag}}\right]_{T,n_{In},n_{Cd}}$ by isothermal drop calorimetry

- successive drops of δn_{Ag} in a pool initially containing (n_{In}+n_{Cd}).
- at each drop, measurement of the thermal effect linked with the mixing.

Working temperature :450°C

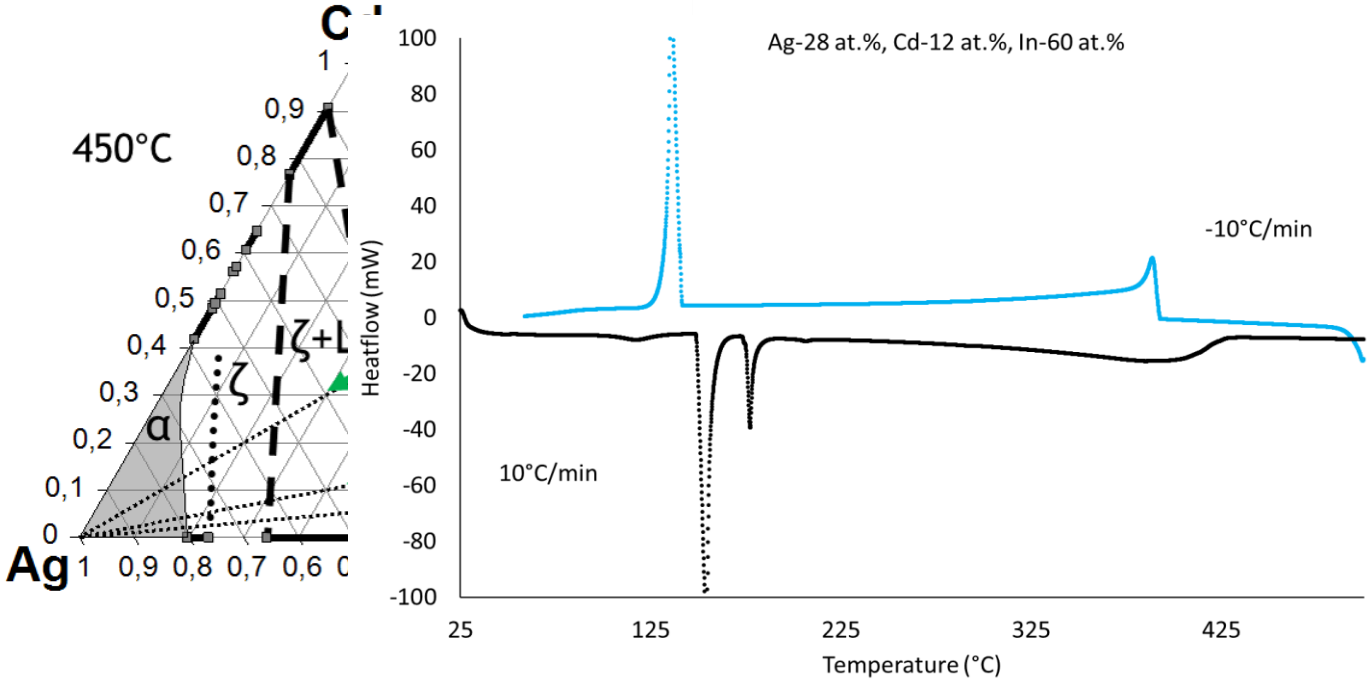
→ Prevent evaporation of Cd and enhancing dissolution





P. Benigni, S. Hassam, A. Decreton, G. Mikaelian, K. Gajavalli, M. Barrachin, E. Fischer, J. Rogez, Jrnl. Chem. Thermodyn. **107,** 207-215 (2017).

Enthalpy of mixing in the Ag-Cd-In ternary liquid phase



Classical DTA

hase over 5 sections,

ıe Cd-rich region,

etermine liquidus from

ıprecise in practicle

measured by DTA.

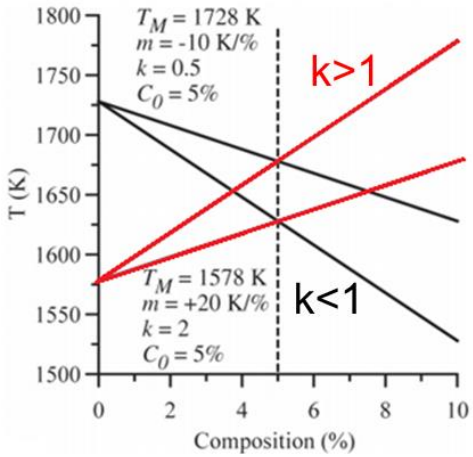
- Classical DTA experiments were performed on various compositons on

  → x_{Cd}=60 and 70 at.% → x_{In}=60 and 55 at.% sections to measure the liquidus

Protocol:

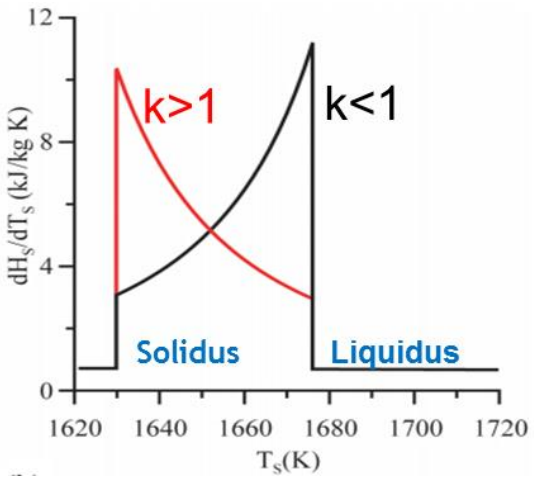
- Heating the sample to a T_{max} at 10, 5, 2, 1, 0.5 °C/min.
- Cooling the sample from T_{max} to T_{room} in same manner.
- True liquidus = extrapolating the liquidus measured at each rate to zero heating rate.

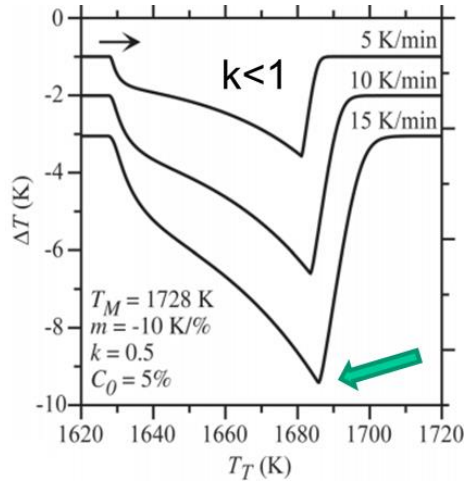
Liquidus detection on heating in classical DTA



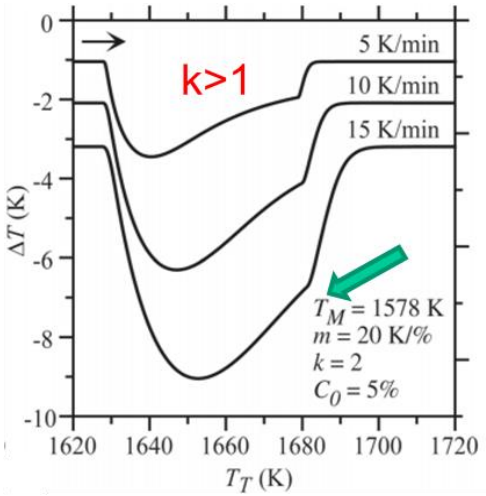
$$k = \frac{Solid\ composition}{Liquid\ compositon}$$

Temperature interval where main part of release heat occurs, depends on ‘k’.





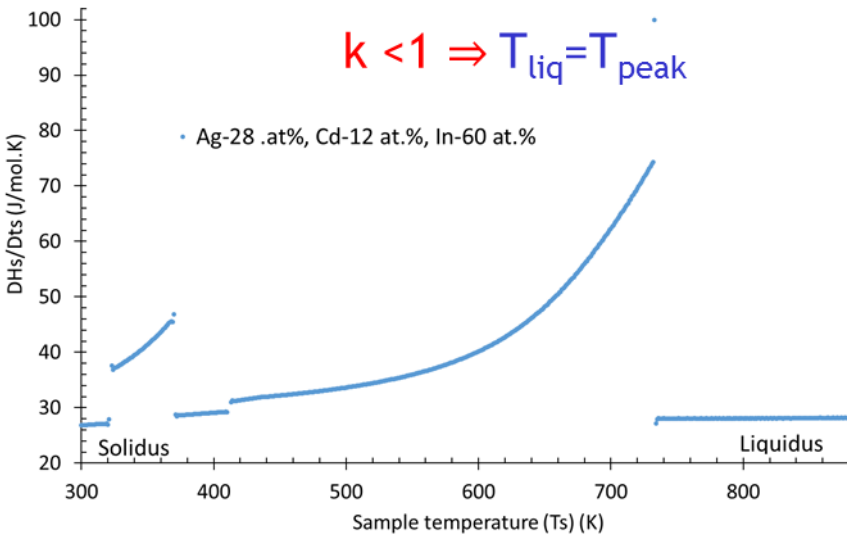
Two kinds of thermal signatures for liquidus detection depending on ‘k’.



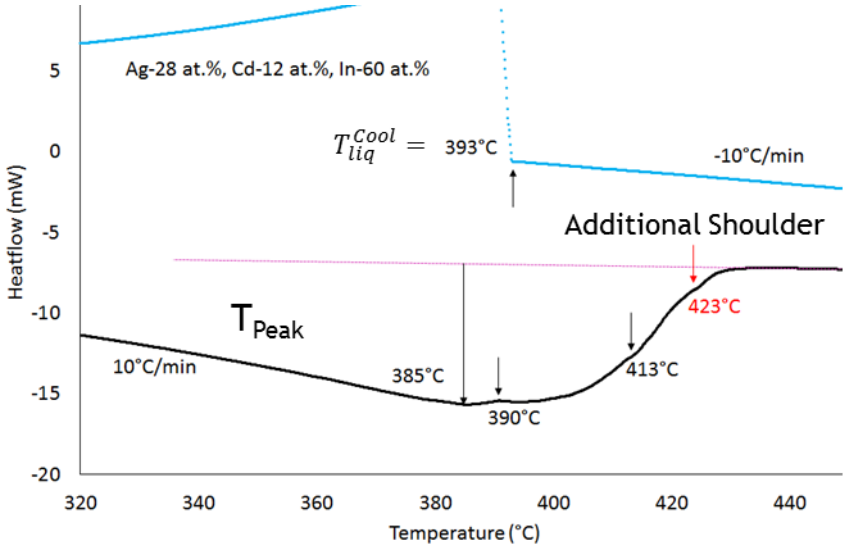
Boettinger, W. J., Kattner, U. R., Moon, K.-W. & Perepezko, J. H., *NIST Spec. Publ.* 960–15, 90 (2006).

Complications in detecting liquidus for Ag-Cd-In ternary alloys

Initial thermodynamic equilibrium calculation



Liquidus detection from DTA thermograms



- Smeared peak over a temperature range around 394°C and additional shoulder at 423°C,

  → $T_{Peak} < T_{liq}^{Cool}$

  → difficulty to unambiguously identify true liquidus.

Complications in detecting liquidus for Ag-Cd-In ternary alloys

- Equilibrium for the (ζ +L) phase, not attained at continuous heating rates:
  - Large compositional stability for the (ζ) solid solution phase – Sluggish diffusion

    → Alloys are prone to microsegregation
  - Undissolved Ag particles observed in calorimetric baths[1] –Slow dissolution kinetics

[1] P. Benigni, S. Hassam, A. Decreton, G. Mikaelian, K. Gajavalli, M. Barrachin, E. Fischer, J. Rogez, Jrnl. Chem. Thermodyn. 107, 207-215 (2017).

Resolution of liquidus detection problems by thermal cycling near the liquidus

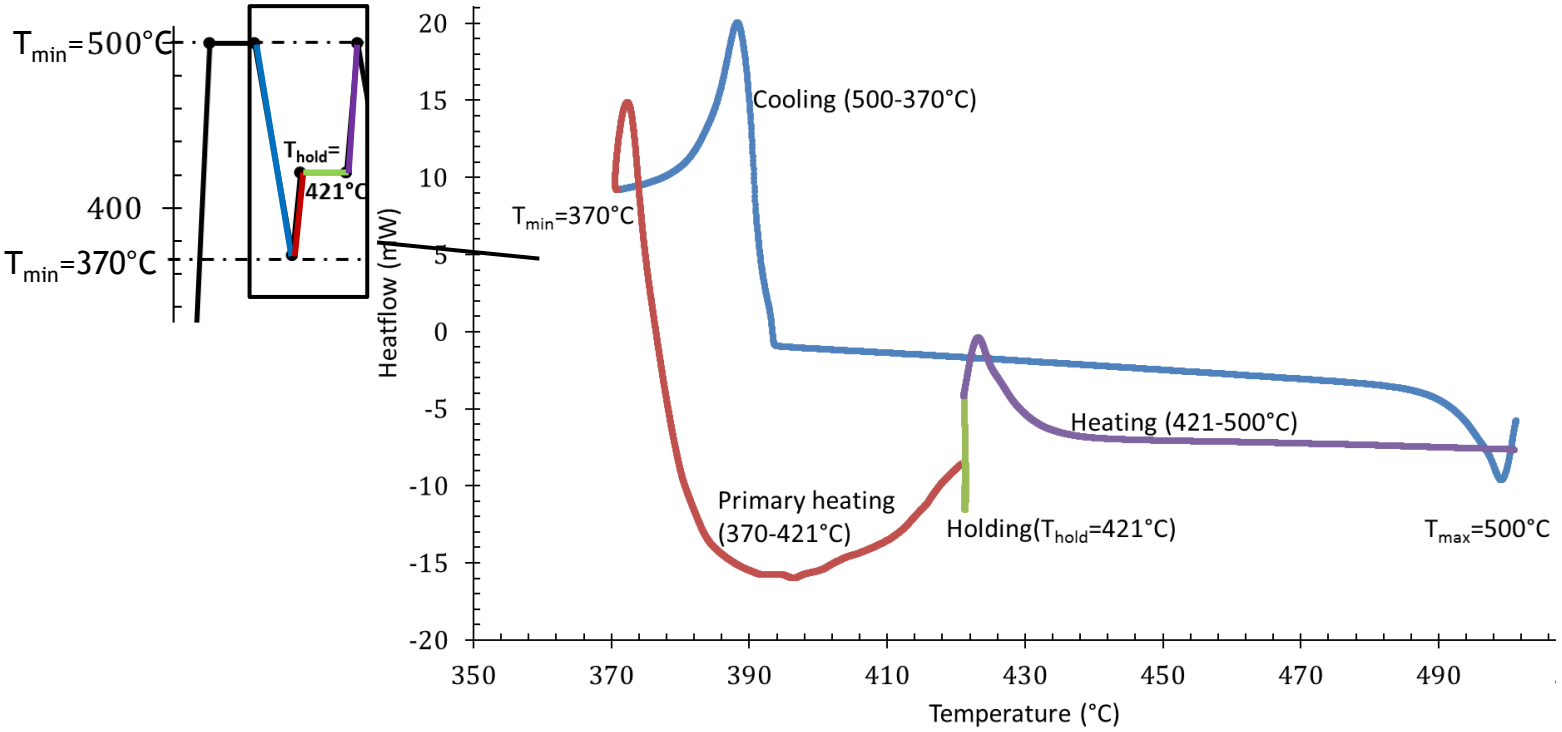
⇓

Interrupted Heating DTA

(R.I. Wu and J.H. Perepezko, Metallurgical and materials transactions 31(2000), 497)
(K.W.Moon, W.J.Boettinger, U.R.Kattnet, F.S.Baincaniello, C.A.Handwerjer, Journal of Electronic Materials, Vol. 29(2000), 10)

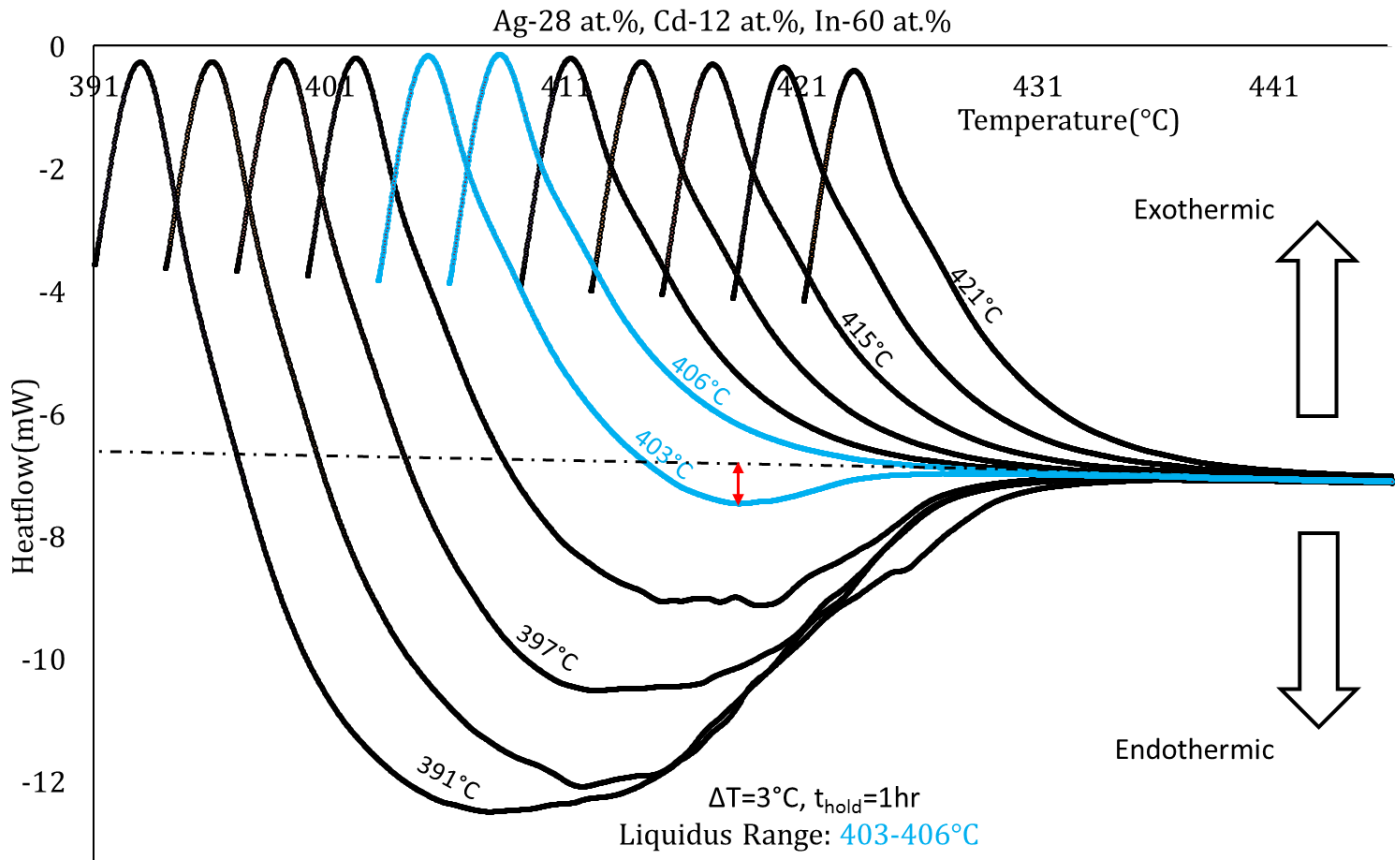
Interrupted heating DTA Protocol

The temperature program for the Ag-28 at.%, Cd-12 at.%, In-60 at.%.



Analysis of heating curves after holding

- Signs of endothermic deviations are sought
- If absent, $T_{Hold} > T_{Liquidus}$ and a fully liquid state is implied



Comparison of the results from various protocols

Composition. (at %)			Classical DTA	Interrupted Heating DTA	Literature Survey
Ag	Cd	In	$T_{liquidus}$	$T_{liquidus}$	$T_{liquidus}$
40	0	60	457 ± 3	451-454	455.9 [1], 455[2]
28	12	60	412 ± 3	403-406	423 [1]
7.5	23.1	69.4	333± 3	319-321	244 [1]

- Liquidus for 11 compositions measured.
- Significant difference in liquidus values.
  - → Upto 20°C, if we consider Additional shoulder temperature and,
  - → Upto 35°C, if we consider T_{peak} as liquidus.
- Quicker manipulation time for Interrupted DTA.

[1] Horrocks, P. J. Phase Diagram and Thermodynamics of the Ag-Cd-In Ternary Alloy Systems. (1991).
[2] Moser, Z. et al. Studies of the Ag-In phase diagram and surface tension measurements. J. Electron. Mater. 30, 1120–1128 (2001).

CALPHAD Optimisation

- **Description of solution phases ($\phi = \alpha$, ζ or L)**

Gibbs energy G^{ϕ} for each solution phase $\quad G^{\phi} = G^{\phi,ref} + G^{\phi,id} + G^{\phi,exbin} + G^{\phi,exter}$

$G^{\phi,ref} = x_{Ag}\,{}^{0}G^{\phi}_{Ag} + x_{Cd}\,{}^{0}G^{\phi}_{Cd} + x_{In}\,{}^{0}G^{\phi}_{In}$ Gibbs energy of Ag, Cd and In (reference)

$G^{\phi,id} = RT\left(x_{Ag}\ln x_{Ag} + x_{Cd}\ln x_{Cd} + x_{In}\ln x_{In}\right)$ Ideal Gibbs energy of mixing

Excess binary terms, adjusted to reproduce the established information for Ag-Cd, Ag-In and Cd-In binaries

$$G^{\phi,exbin} = x_{Ag}x_{Cd}\sum_{v} {}^{v}\boldsymbol{L}^{\boldsymbol{\phi}}_{\boldsymbol{Ag,Cd}}\left(x_{Ag}-x_{Cd}\right)^{v} + x_{Ag}x_{In}\sum_{v} {}^{v}\boldsymbol{L}^{\boldsymbol{\phi}}_{\boldsymbol{Ag,In}}\left(x_{Ag}-x_{In}\right)^{v} + x_{Cd}x_{In}\sum_{v} {}^{v}\boldsymbol{L}^{\boldsymbol{\phi}}_{\boldsymbol{Cd,In}}\left(x_{Cd}-x_{In}\right)^{v}$$

- **Adjustment of ternary excess parameters**

$$G^{\phi,exter} = x_{Ag}x_{Cd}x_{In}\left(x_{Ag}\,{}^{\boldsymbol{Ag}}\boldsymbol{L}^{\boldsymbol{\phi}}_{\boldsymbol{Ag,Cd,In}} + x_{Cd}\,{}^{\boldsymbol{Cd}}\boldsymbol{L}^{\boldsymbol{\phi}}_{\boldsymbol{Ag,Cd,In}} + x_{In}\,{}^{\boldsymbol{In}}\boldsymbol{L}^{\boldsymbol{\phi}}_{\boldsymbol{Ag,Cd,In}}\right)$$

α Phase

$${}^{Ag}L^{\alpha}_{Ag,Cd,In} = {}^{Ag}a^{\alpha} + {}^{Ag}b^{\alpha}T$$
$${}^{Cd}L^{\alpha}_{Ag,Cd,In} = {}^{Cd}a^{\alpha} + {}^{Cd}b^{\alpha}T$$
$${}^{In}L^{\alpha}_{Ag,Cd,In} = {}^{In}a^{\alpha} + {}^{In}b^{\alpha}T$$

ζ Phase

$${}^{Ag}L^{\zeta}_{Ag,Cd,In} = {}^{Ag}a^{\zeta} + {}^{Ag}b^{\zeta}T$$
$${}^{Cd}L^{\zeta}_{Ag,Cd,In} = {}^{Cd}a^{\zeta} + {}^{Cd}b^{\zeta}T$$
$${}^{In}L^{\zeta}_{Ag,Cd,In} = {}^{In}a^{\zeta} + {}^{In}b^{\zeta}T$$

Liquid Phase

$${}^{Ag}L^{L}_{Ag,Cd,In} = {}^{Ag}a^{L} + {}^{Ag}b^{L}T$$
$${}^{Cd}L^{L}_{Ag,Cd,In} = {}^{Cd}a^{L} + {}^{Cd}b^{L}T$$
$${}^{In}L^{L}_{Ag,Cd,In} = {}^{In}a^{L} + {}^{In}b^{L}T$$

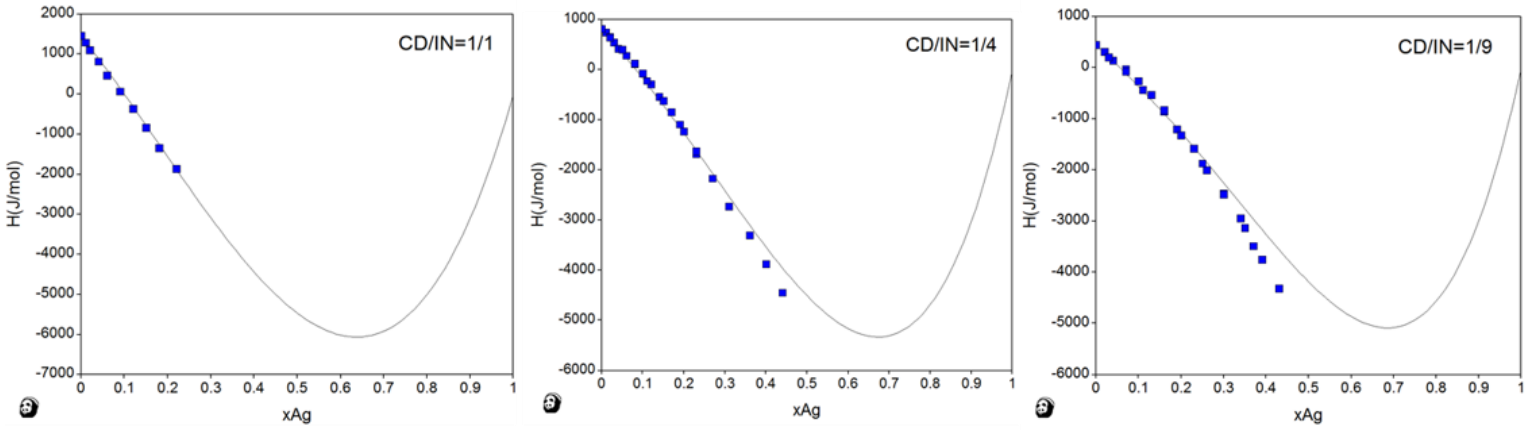
18 adjustable parameters

9 optimized parameters

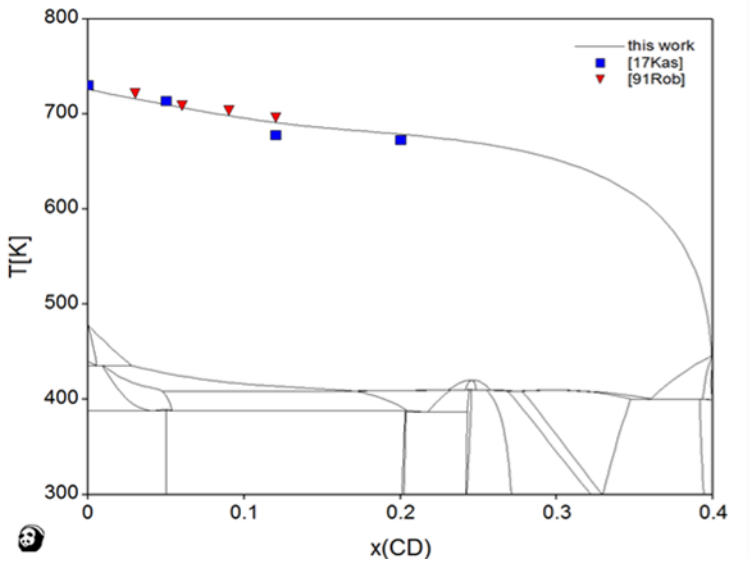
IRSN

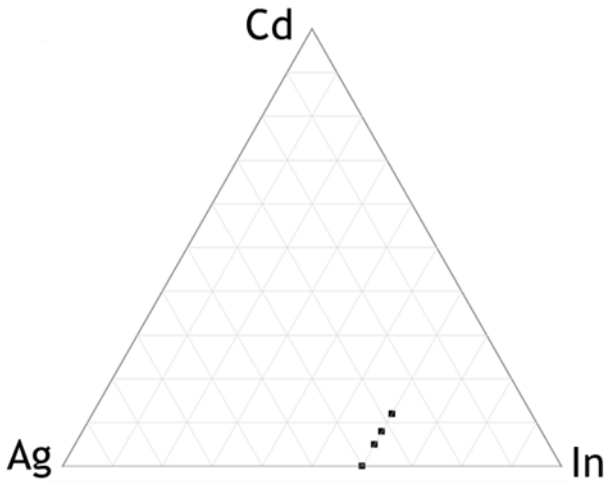
CALPHAD Optimisation of Ag-Cd-In system

Ag-Cd-In liquid at 450°C modeled from the mixing enthalpy measurements



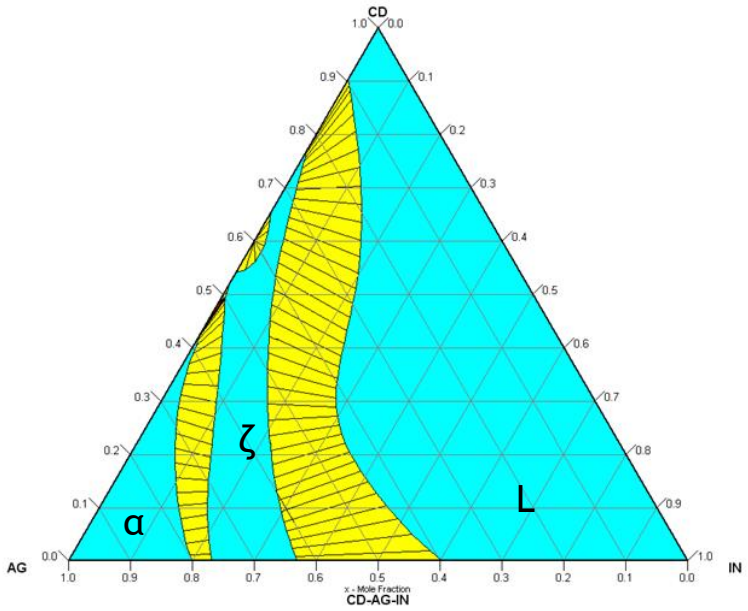
x_{In} section = 60 at.% isopleth section of the Ag-Cd-In ternary diagram from the DTA experiments



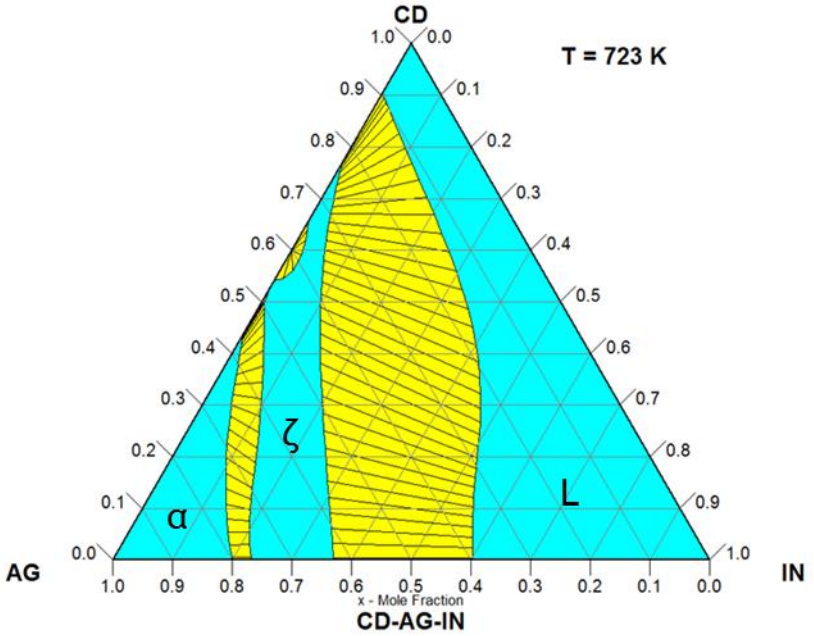


CALPHAD Optimisation of Ag-Cd-In system

Without excess ternary term, only from the binary systems



Ternary diagram optimization with excess ternary parameters



Conclusions

Objectives achieved

- For the first time, complete investigation of thermodynamics Ag-Cd-In ternary system at high temperature.

Experiments performed

- Enthalpy of mixing measurements in the liquid at 450 °C by calorimetry,
- Temperature measurements of liquidus by interrupted DTA protocol to succeed in dealing with the inconsistencies in liquidus measurements by classical DTA

CALPHAD optimisation of the system

Work performed and Perspectives

- Ag-Zr (PhD, A. Decreton, Aix Marseille University, 2015)
- In-Zr (in course)
- In_2O_3-ZrO_2: (next future collaboration with JRC-Karlsruhe, Dario MANARA)

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Dario MANARA
Rudy KONINGS

WE ACKNOWLEDGE AND THANK EVERY MEMBER FOR THEIR UNWAVERING SUPPORT

Questions ??

Accident Progression Analysis Based on Experiments QUENCH-16 and QUENCH-10

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E-mail: pivryashkova@inrne.bas.bg

Keywords: Severe accident, Hydrogen generation, Oxygen consumption, Temperature escalation, Reflooding

Both experiments addressed air ingress into an overheated core following earlier partial oxidation in steam. QUENCH-10 was performed with extensive preoxidation and moderate-high air flow rate, while QUENCH-16 was performed with limited preoxidation and low air flow rate. In both experiments the bundles were reflooded with water. In each simulation the same input model, assumptions and code version of MELCOR 2.1 are used for both experiments, differing only in respect of the boundary conditions. Series of simulations using new model for oxidation for air at low and high temperatures of MELCOR 2.1 for QUENCH-16 and QUENCH-10 were performed to examine influence of oxygen concentrations to improve the results of hydrogen generation during the quench phase. The QUENCH-16 and QUENCH-10 tests were conducted successfully at KIT (Karlsruhe Institute of Technology).
The analyses of the two similar quench air ingress experiments with MELCOR 2.1 code have improved the understanding of cladding oxidation by air and hydrogen generation during the reflood. The quench air ingress experiments show the magnitude of different behaviour of oxidation in steam and air environments on based of initial and boundary conditions. There is a significant range of calculated oxidation rates depending on the boundary conditions of the test. The simulation of the two related experiments conducted with different boundary and initial conditions provides an opportunity to assess the progression of a nuclear accident under different conditions.


Petya Vrochkova
Institute for Nuclear research and Nuclear energy
Bulgaria

23RD INTERNATIONAL QUENCH WORKSHOP

”Accident Progression Analysis Based on Experiments QUENCH-16 and QUENCH-10”

Petya Vryashkova, Pavlin Groudev

17-19 October 2017, Karlsruhe, Germany

CONTENT

- Introduction
- Scenario of the QUENCH-16, -10 experiment
- Description of Quench facility
- Quench-16, -10 facility MELCOR model
- Oxidation model of MELCOR 2.1
- Analysis of results of Quench-16, -10 with MELCOR 2.1
- Conlusions



Introduction

- Analyses are presented with the version of the MELCOR 2.1 computer code of the QUENCH-16 and QUENCH-10 experiments.
- As opposed to steam, the air ingress is expected to increase the damage of the fuel rods
- In each simulation the same input model, assumptions and code version of MELCOR 2.1 are used for both experiments, differing only in respect of the boundary conditions and initial conditions.
- It also provides an opportunity to assess the progression of a nuclear accident under different conditions.



Introduction

- The analyses of the two similar quench air ingress experiments with MELCOR 2.1 code have improved the understanding of cladding oxidation by air and hydrogen generation during the reflood.
- QUENCH-10 was performed with extensive preoxidation and moderate-high air flow rate, while QUENCH-16 was performed with limited preoxidation and low air flow rate.
- In both experiments, reflooding is before reaching the melting temperature of the cladding the fuel rods to avoid strong oxidation.

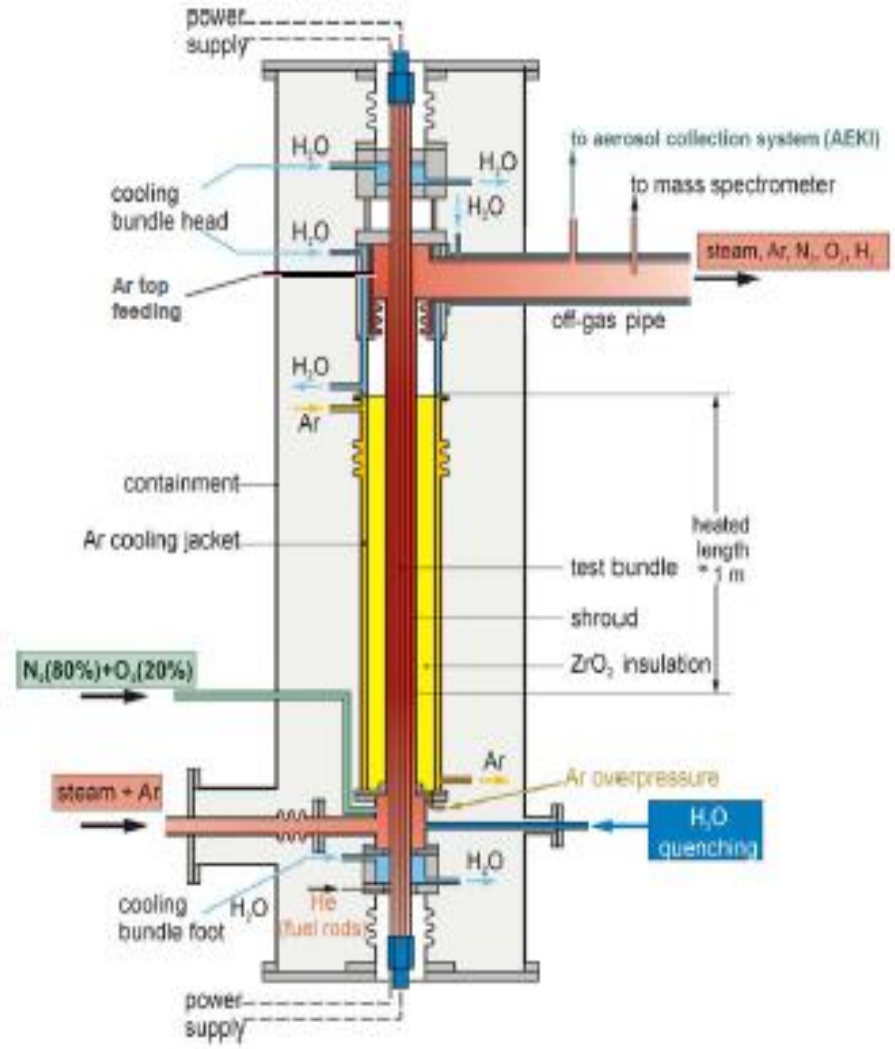


SCENARIOS OF THE EXPERIMENTS

Phase		QUENCH-10	QUENCH-16
I	T_{max}, steam phase	**1620 K**	**≈1430 K**,
	power increase to	13 kW	10-11.5 kW
	duration	30 min	1 h 15 min
	steam flowrate	3.0 g/s	3.4 g/s
	argon flowrate	3.0 g/s	3.0 g/s
II	Pre-oxidation of the test bundle for	**113 min**	**≈ 30 min**
	at constant temperature of	1620-1690 K	1430K,
	power:	10-13 kW	11.5 kW,
	steam flowrate:	3.0 g/s	3.4 g/s,
	argon flowrate:	3.0 g/s	3.0 g/s
III	Intermediate cooling from	**≈1690 K**	**≈1430 K**
	to	≈1190 K	≈990 K
	duration:	**38 min**	**17 min**
	power:	7 kW	4 kW
	steam flowrate:	**3.0 g/s**	**3.4 g/s**
	argon flowrate:	3.0 g/s	3.0 g/s
IV	Air ingress and slow heat-up from	≈1190 K	≈990 K
	to	**≈2200 K**	≈1823 K
	duration:	30 min	67 min
	oxygen starvation for	≈1.5 min	14 min
	power:	7-8 kW	4 kW
	steam flowrate:	0.0 g/s	0.0 g/s
	argon flowrate:	3.0 g/s	1.0 g/s
	air flowrate:	**1.0 g/s**	**0.2 g/s**
V	Quenching of bundle at	**13400 s**	**11341 s**
	water flowrate:	50 g/s	50 g/s

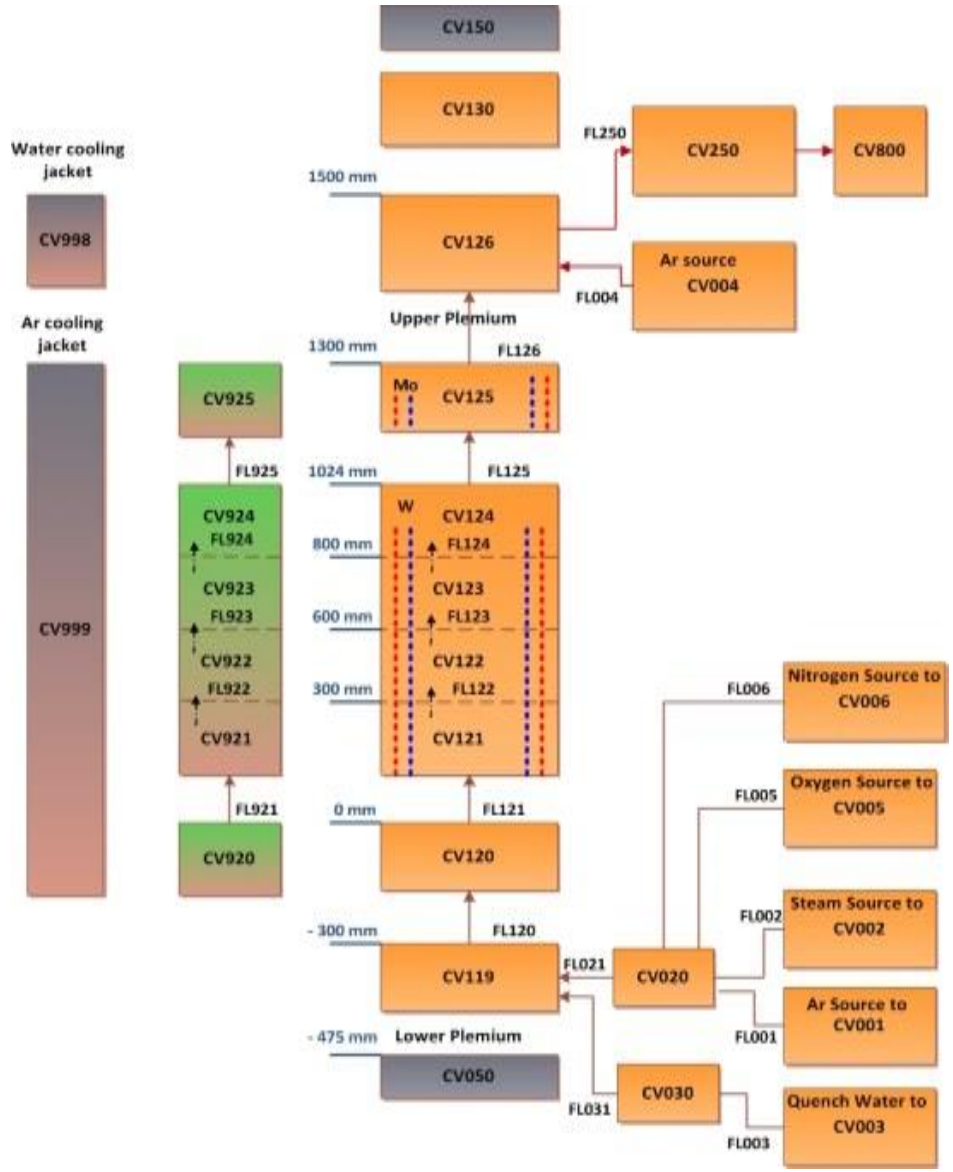


DESCRIPTION OF QUENCH-10, -16 FACILITY



- 20 fuel rod simulators, electrically heated over 1 m length (PWR type of reactor)
- 1 central unheated fuel rod simulator
- rods filled with He (~0.22 MPa, cladding failure)
- 5 grid spacers (1 Inconel, 4 Zry)
- 4 corner rods
- shroud: Zr, insulation ZrO2
- annulus shroud / cooling jacket filled with Ar at 0.22 MPa

QUENCH- 10, -16 FACILITY MELCOR MODEL



- twenty-two axial core levels subdivided in four radial rings
- Sixteen hydrodynamic volumes were used to represent the core;
- The cooling systems were represented as a fixed temperature boundary.
- Additional volumes were used to represent the lower and upper plenum.

OXIDATION MODEL OF MELCOR 2.1

The MELCOR 2.1 was used for the analysis:

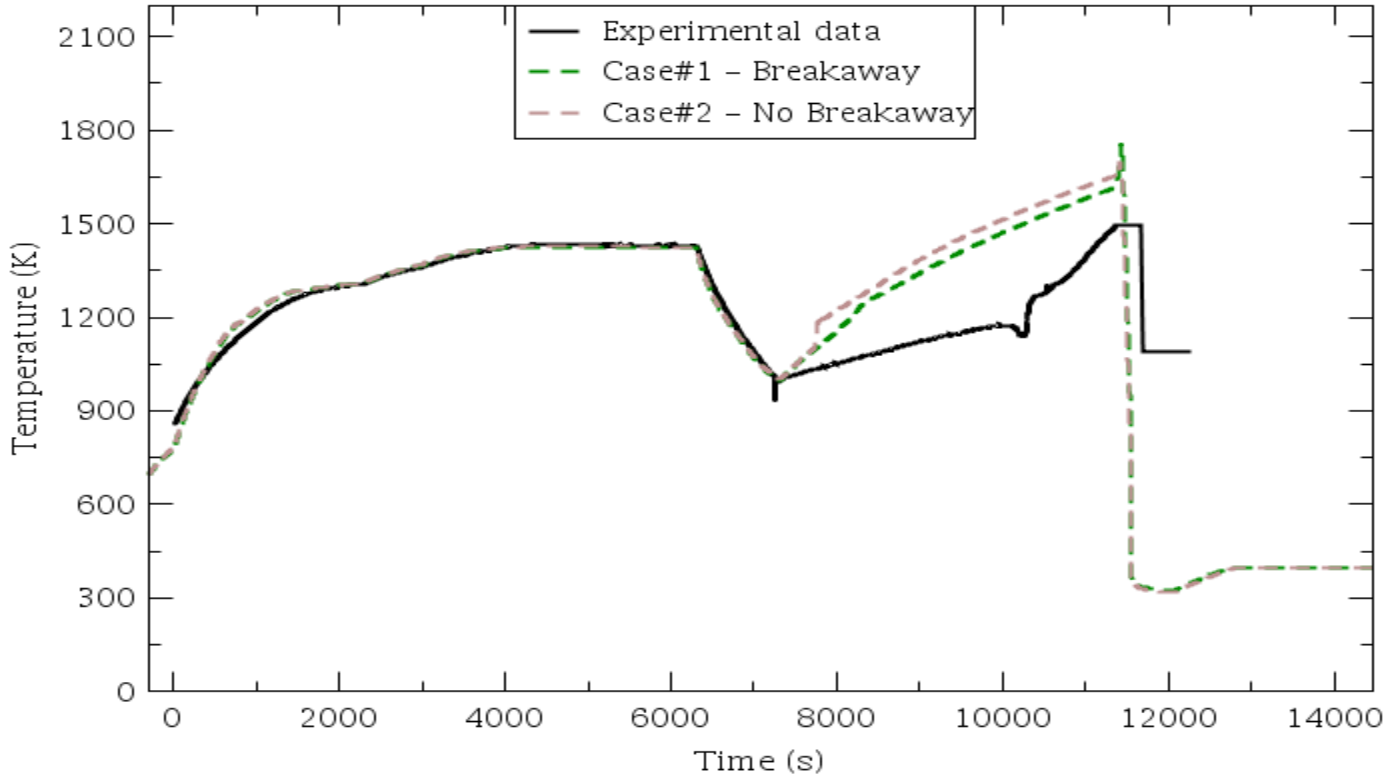
- For the Zircaloy-oxygen reaction is used breakaway model.

The notable feature of the model is a transition from parabolic to linear kinetics.

- For the Zircaloy-steam reaction, the oxidation constant is evaluated using the **Urbanic-Heidrich constants**, which are implemented (along with the transition temperatures of 1853 K and 1873 K) as sensitivity coefficients.

ANALYSIS OF QUENCH-16 WITH MELCOR 2.1

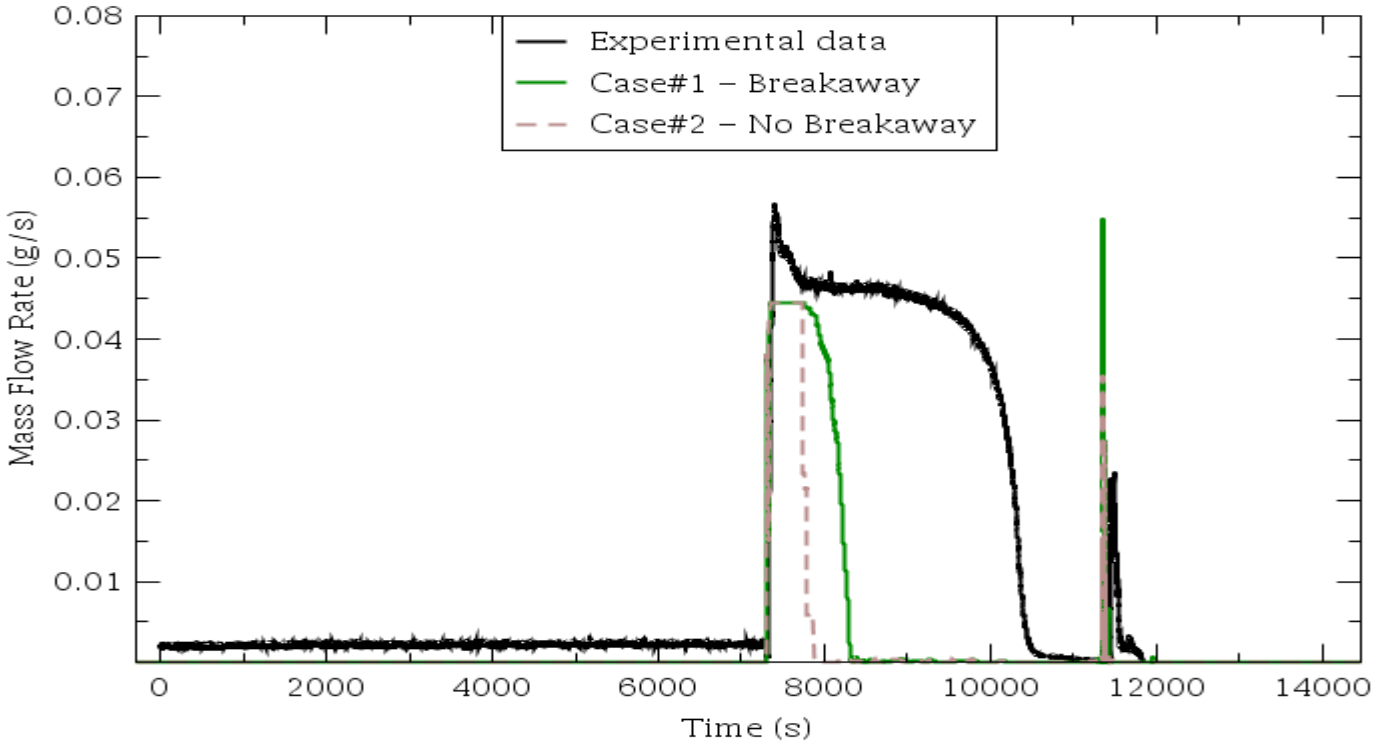
Temperature inner rod at elevation 950 mm



- During the air ingress phase the calculated temperatures are higher compare with experimental data.
- The hottest temperature location has changed from the 950 mm to lower elevations due to rapid oxygen consumption during air ingress phase.

ANALYSIS OF QUENCH-16 WITH MELCOR 2.1

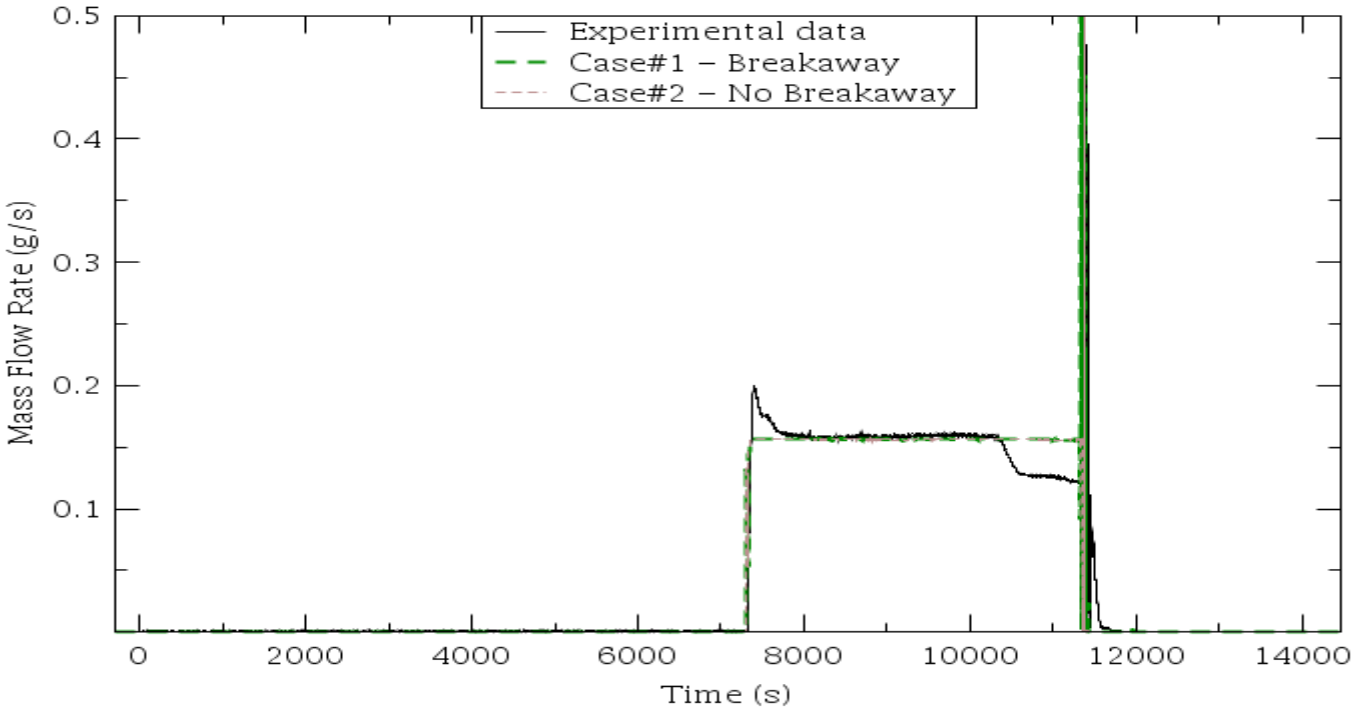
Oxygen consumption



- Early oxygen consumption was corresponding on increasing in the temperature.
- All of the calculations showed an earlier than measured oxygen consumption with MELCOR 2.1 with or without breakaway model.

ANALYSIS OF QUENCH-16 WITH MELCOR 2.1

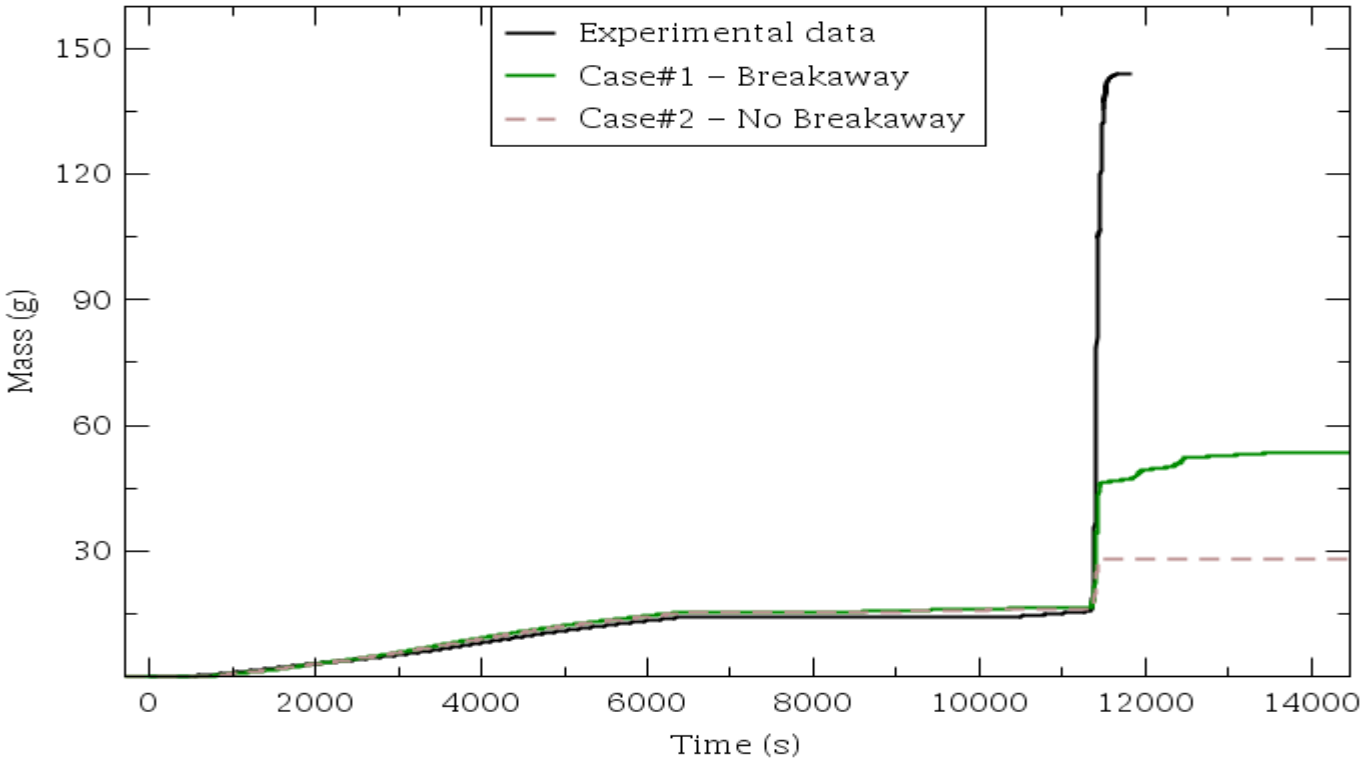
Nitrogen consumption



- The case#1 with breakaway model estimation was very close to the experimental results.
- The case#2 has underestimated it, following a late occurrence of nitrogen reaction.

ANALYSIS OF QUENCH-16 WITH MELCOR 2.1

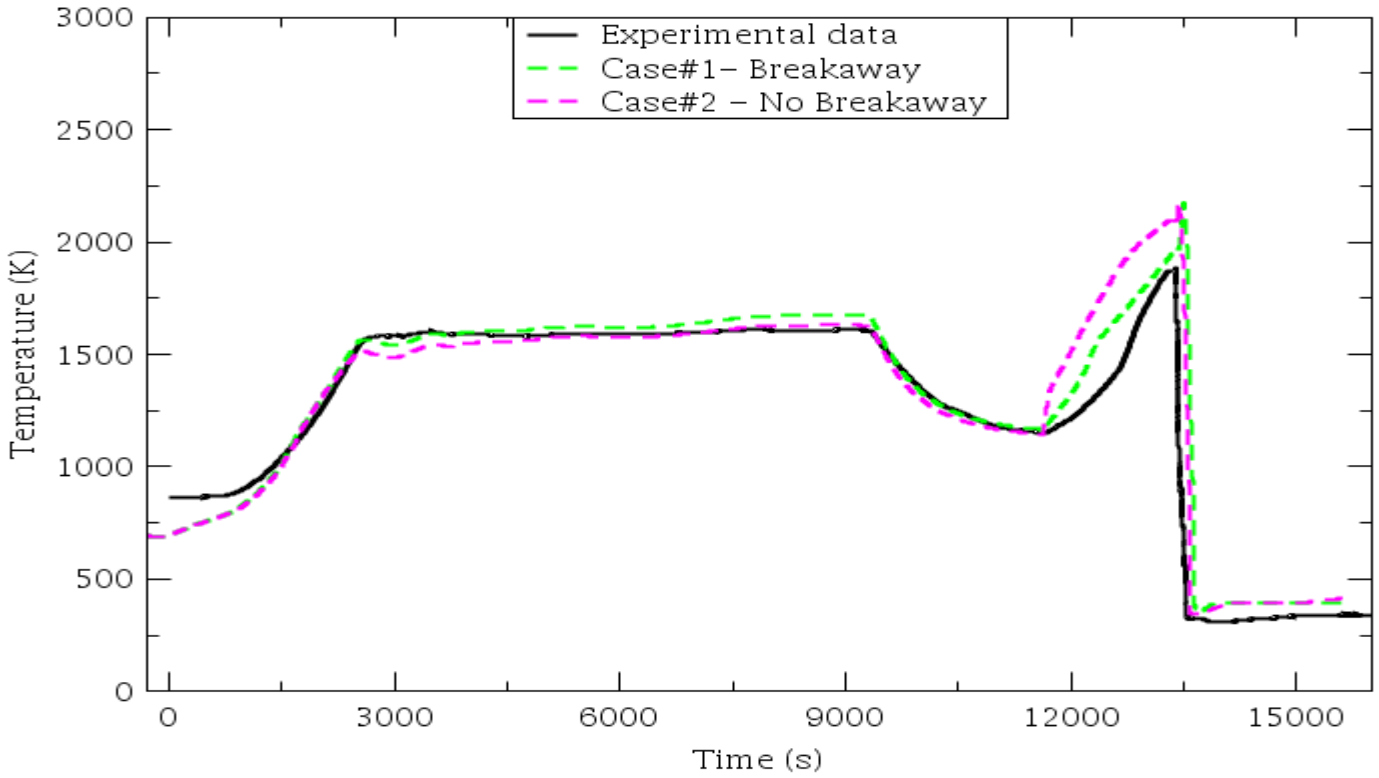
Total hydrogen generation



- The calculations gave a closer prediction on the time when the hydrogen started to be generated (during the heat-up phase) compare with experimental data.
- The hydrogen generation during reflood was underestimated by all cases.

ANALYSIS OF QUENCH-10 WITH MELCOR 2.1

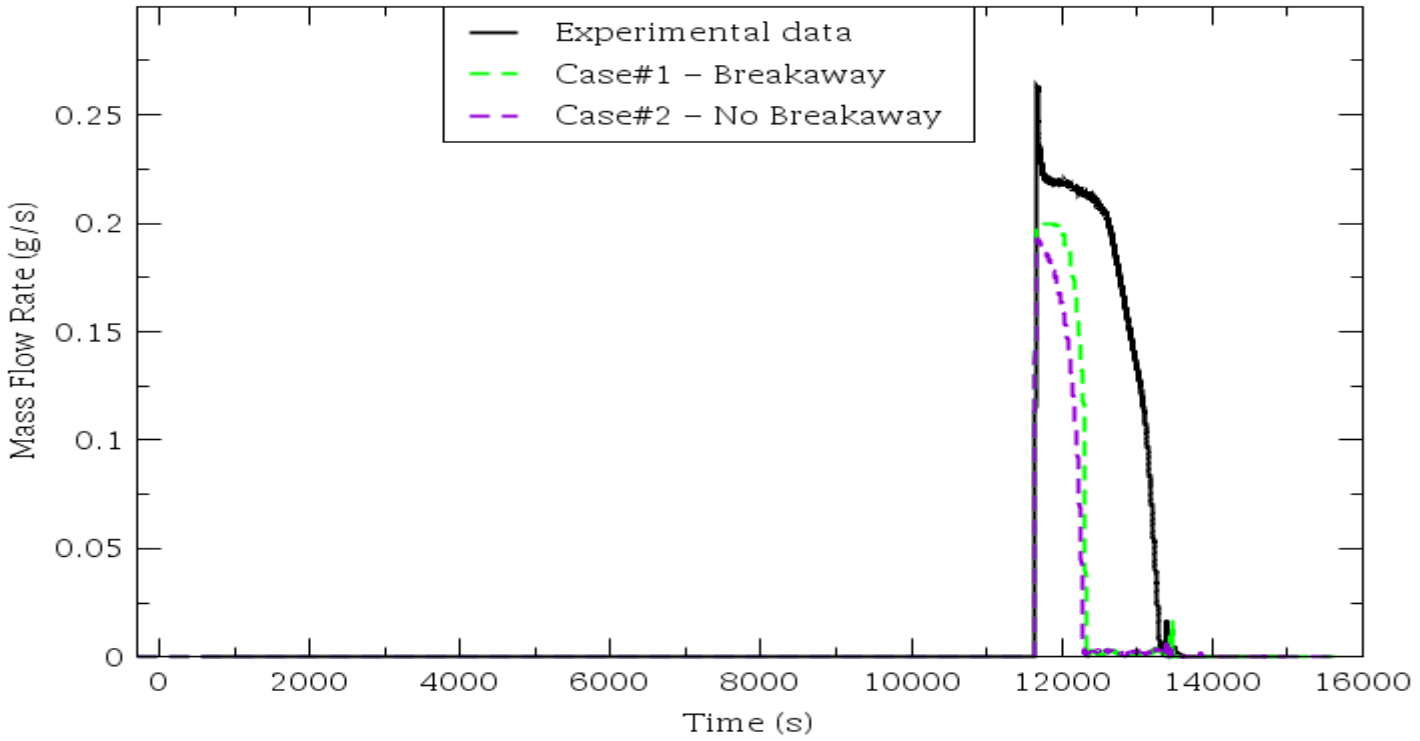
Temperature inner rod at elevation 950 mm



- The calculations showed a better temperature in good agreement with the experimental data.
- During the air ingress phase the temperature slighty overpredicted with breakaway or without breakway.

ANALYSIS OF QUENCH-10 WITH MELCOR 2.1

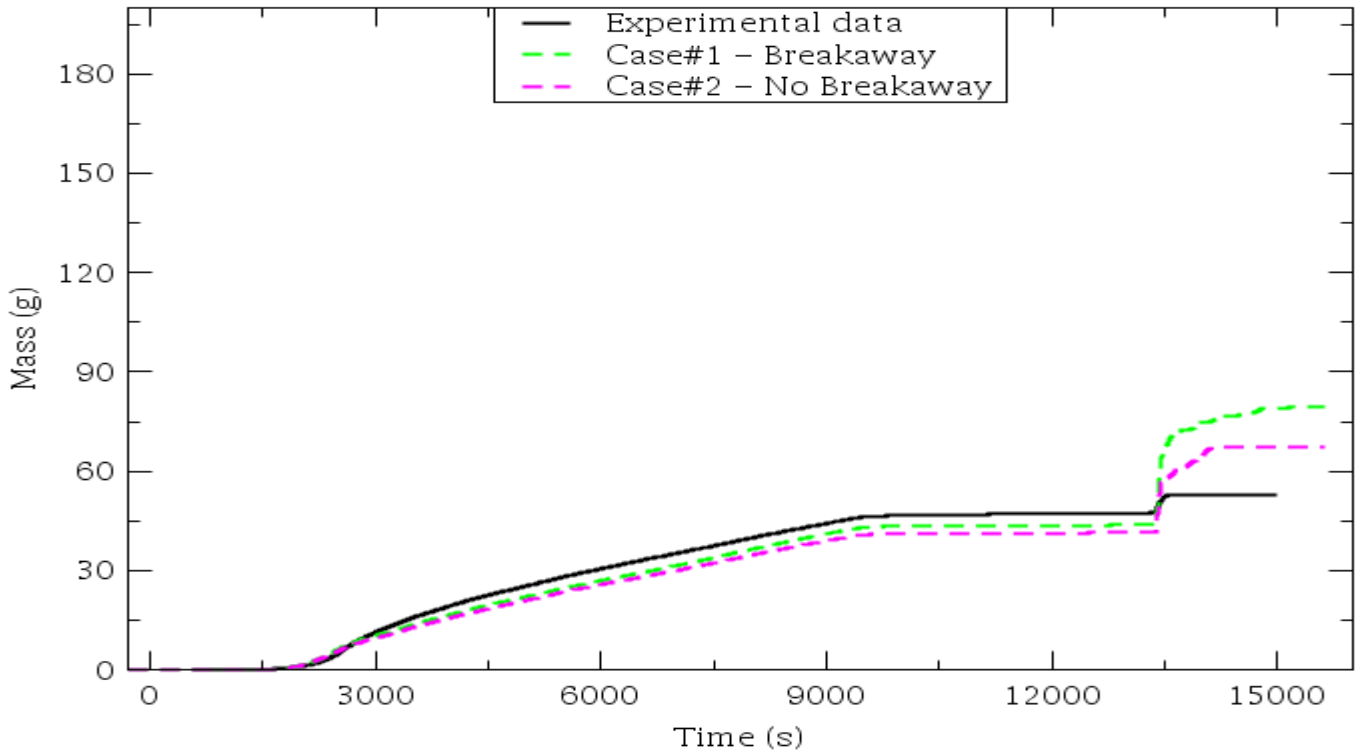
Oxygen consumption



- The calculated oxygen consumption in the all cases was lower than in the experiment.
- According to the model of the oxidation the onset of oxygen consumption may be quite sensitive to the prior oxidation.

ANALYSIS OF QUENCH-10 WITH MELCOR 2.1

Total hydrogen generation



- The hydrogen generation during reflood was overpredicted by all cases for QUENCH-10 after a long period of oxygen starvation.

Conclusions

- The quench air ingress experiments confirmed the different behaviour of oxidation in steam and air environments.
- The combined effect of air and steam can lead to an increased rate of damaged of fuel rods, which is major importance for the safety of a nuclear power plant.
- The QUENCH tests aims not only to determine the amount of hydrogen released during reflood of a test bundle with core materials as cladding and spacer grids, but also to investigate the related high-temperature interactions of the core materials.



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Acknowledgements

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Thank you for your attention!



ACCIDENT TOLERANT MATERIALS – IDEAS and RESEARCH DIRECTIONS

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ABSTRACT

Zirconium has very good water corrosion and radiation resistance at normal working conditions of nuclear reactors. Due to these properties it is commonly used as cladding material for fuel elements. However, in the case of LOCA conditions, the extremely fast oxidation of zirconium at steam atmosphere or and air/steam mixture results in intense hydrogen generation and possible hydrogen-oxide mixture explosion. The development of the solution to minimize the risk in such of accidents is needed. The concept of Accident Tolerant Fuels (ATF) and Accident Tolerant Materials (ATM) has been developed recently for this purpose.

Advanced materials that are intended to be more tolerant of severe accident conditions with the safe operation under normal conditions are looking for. There are two main ideas for the increasing of claddings' corrosion resistance:

(i) developing of new materials for claddings production (SiC composities, FeCrAl alloys)
(ii) modification of the surface layer of claddings (incorporation of rare earth elements, formation of the protective layer on the base of silicon, ceramics MAX, FeCrAl alloys).

Scientific works are carried out in the frame of IAEA CRP on Analysis of Options and Experimental Examination of Fuels for Water-Cooled Reactors with Increased Accident Tolerance, acronym ACTOF. INCT proposal "Silicide/silicate coatings on zirconium alloys for improving the high temperature corrosion resistance" is aimed to develop the silicon-based coatings on zirconium alloys claddings and evaluation of their properties at accident scenario as well as in the reactor normal working conditions. The main goal of this program consist in developing of surface layer of zirconium claddings modification by different physical and chemical methods. Among the applied modifications methods the following techniques have been used: alloying with high-intensity pulsed ion-plasma beams treatment with the chosen electrodes material incorporation, coatings deposition by magnetron sputtering of zirconium silicide or chromium targets, melting of the deposited silicon particles using TIG technique, coating with sol-gel method, plasma electrolytic oxidation (PEO).

The results of the experimental works concerning synthesis method, coatings characterization, autoclave and furnace tests results will be reported.

Acknowledgements
This work has been partially supported under IAEA Research Contract No. 19026/R0 and by Polish Ministry of Science and Higher Education under decisions 3689/IAEA/2017/0



ACCIDENT TOLERANT MATERIALS – IDEAS and RESEARCH DIRECTIONS

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23 International Quench Workshop, Karlsruhe, 17-19.10.2017

BACKGROUND

- Severe accidents such as those at three Mile Island and Fukushima Daiichi have shown that under such extreme conditions, nuclear fuel will fail and the high temperature reaction between zirconium alloys and water will lead to generation of hydrogen with the potential for explosions, damaging the plant further.

  - There have been many improvements to the original design and materials used. However , the basic concept of uranium oxide feel pellets clad with zirconium alloy tubing has reminded the choice for the majority of commercial nuclear power plants

- Development of the solution to minimize the risk related to the design and unforseable situations is urgently needed

- The concept of Accident Tolerant Fuels **(ATF)** and Accident Tolerant Materials **(ATM)** - it means materials with increased accident tolerance - has been developed recently for this purpose

CLADDINGS

Zirconium alloys due to their good water corrosion and radiation resistance at normal working conditions of nuclear reactors are commonly used as cladding material

In the case of normal working conditions (cooling water temperature 360°C for PWR) the slow zirconium oxidation occuers due to presence of water absorbed at the claddings surface or the oxygen from water radiolises reaction

In the case of LOCA conditions, the **extremely fast oxidation** of zirconium at steam atmosphere or at air/steam mixture at temperatures above 800°C results in intense hydrogen generation and possible hydrogen-oxide mixture explosion

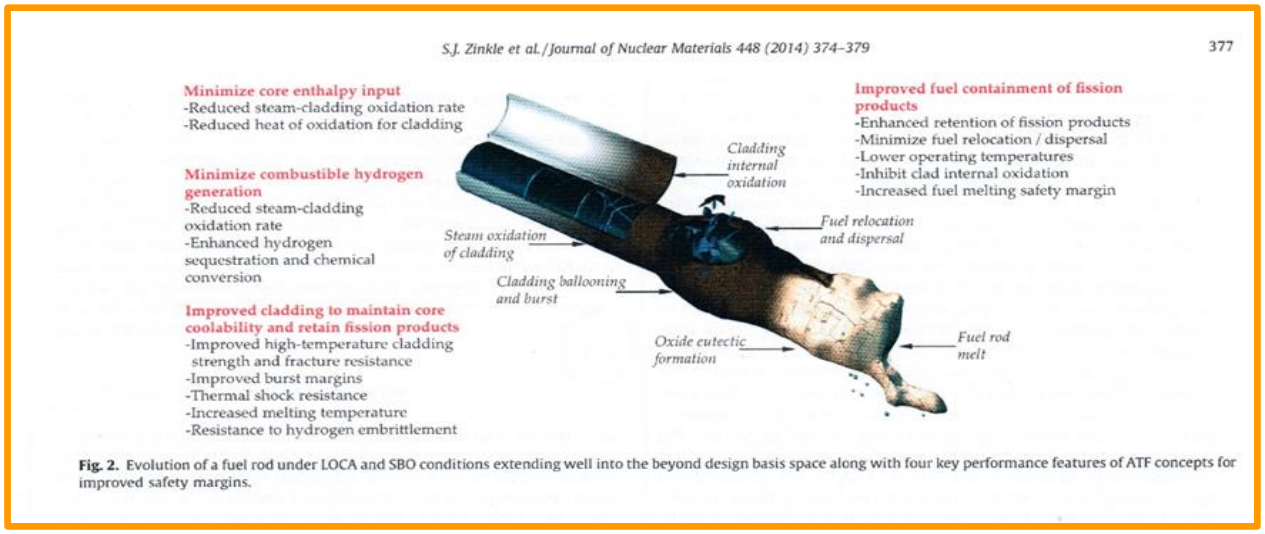


Fig. 2. Evolution of a fuel rod under LOCA and SBO conditions extending well into the beyond design basis space along with four key performance features of ATF concepts for improved safety margins.

DEVELOPMENT AND INVESTIGATIONS DIRECTIONS

A variety of approaches are being considered as potential accident-tolerant alternatives to Zr alloy cladding. The approaches can be grouped into three general categories

Non-zirconium cladding with high strength and oxidation resistance

Issues to be resolved:

- effect of higher thermal neutron absorption cross section
- the impact of radiation hardening and embrittlement on cladding
- pellet-cladding mechanical interaction
- oxidation behavior in steam at high temperature
- potential enhanced susceptibility to stress corrosion cracking
- cost for fabrication of tubing

Zr alloy cladding with improved high temperature oxidation resistance and/or strength

Challenges:

- coating could be easily regenerated if a small portion spalled off (fuel assembly handling or normal reactor operation)
- careful matching of the thermal expansion coefficient in order to minimize interfacial stresses and delamination during temperature cycling (RT to 300oC – normal or 300o C to high temperature - off-normal conditions)
- coating must be compliant enough to withstand the diametrical compression

The durability of coatings to protect the exposed cladding to a sufficient extent after burst also needs to be considered because after burst, the inner, uncoated surface of the cladding is exposed to the steam, leading to rapid oxidation

Altenative fuel forms with improved properties compared to monolithic UO2

(not in this talk)

Highly adherent oxidation-resistant coatings can be alternative to bulk materials for cladding

NON-ZIRCONIUM CLADDING

Structural alloys

Oxidation – resistant structural alloys offer the potential for improved strenght and oxidation resistance to existing Zr alloys over a broad temperature range

As for example:

- austenitic stainless steels: AISI 304, AISI 316, AISI 347
- austenitic nickel based alloys: Inconel 600, Incoloy 800
- ferritic steel with chromium AISI430

[Terrani, J.Nucl. Mater. 448,1-3,2014, 420]



Characteristic of Ferritic Stainless Steels for ATF Cladding. *[IAEA-TECDOC-1797]*

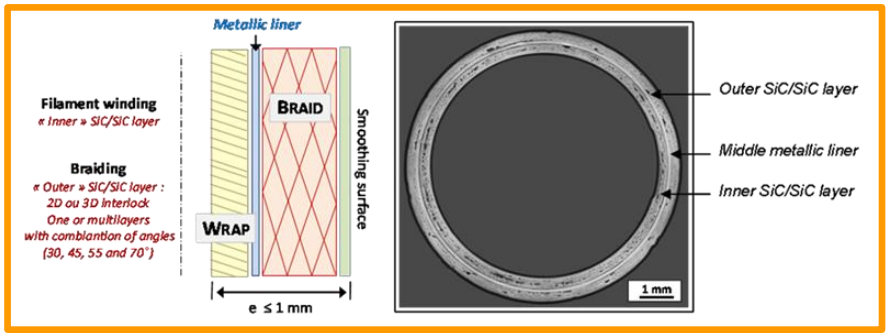
SiC

SiC/SiC composite cladding was investigated at first for Generation IV application in Gas Fast Reactors due to its very good high temperature properties and dimensional stability

Silicon carbide fiber-reinforced SiC matrix ceramic composites (SiC/SiC) have been proposed as a potential cladding material

- retention of strength up to very high temperatures
- good radiation resistance
- good oxidation resistance in air and steam up to temperatures of at least 1600o C.

[Yueh K. et al., Nucl. Eng. Int. 55 (2010) 75]



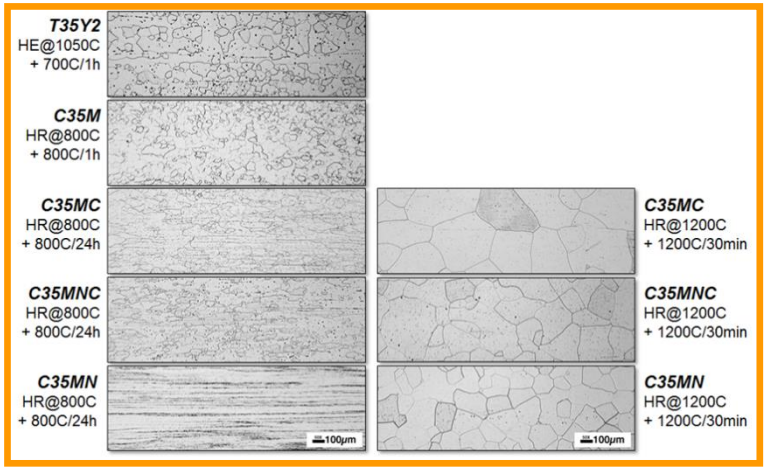
The CEA sandwich concept: thin (50-100 µm) refractory metal liner in between two SiC composite layers [IAEA-TECDOC-1797, 2016]

NON-ZIRCONIUM CLADDING

FeCrAl [wt.%]: **Fe: 10-20 Cr: 3-5 Al: 0-0.15**

Nuclear grade, iron-based wrought FeCrAl alloys has been proposed as the candidate materials for accident tolerant fuel cladding

More Cr: better for corrosion and oxidation resistance, but it would increase potential embrittlement [Pint, J.Am.Ceram.Soc.86,2003,686]
More Al: better oxidation resistance at elevated temperatures, but the ductile-brittle transition temperature may be raised

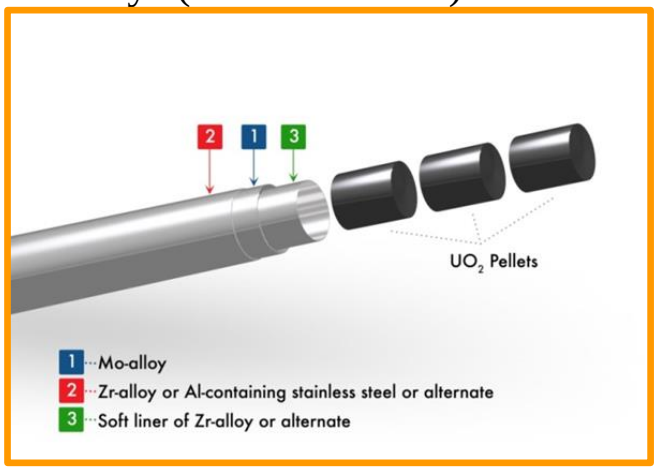


Light optical micrographs showing the FeCrAl alloys processed at 800 (left) and 1200° C (right) [IAEA-TECDOC-1797].

Refractory alloys

Refractory alloys based on Nb, Ta, Mo, or W offer the potential for significant improvements in high temperature strength compared to Zr alloys

For example:
Molybdenum is a refractory metal which melts at a temperature of 2623°C, which is close to the melting point of UO_2 (2850°C) and nearly 1000°C more than zirconium alloys (around 1850°C).



Schematic of EPRI's Mo cladding concept, (Electric Power Research Institute USA) IAEA-TECDOC-1797]

OXIDATION RESISTANT COATINGS

Chromium

Protective oxidation barier due to properties:

- significantly improves resistance to corrosion in the 360oC water test
- reduced corrosion rates in high temperature steam
- increased post-quench mechanical behaviour due to a reduced ingress of oxygen within the metal to form the alfa (O)-Zr phase
- fabricated by PVD method creates a dense adherent layer



Overview of uncoated and coated Zy-4 tubes exposed to steam at 1200°C for 6000 s after quenching. [IAEA-TEODOC-1797]

MAX phases

M: transition metals (Sc, Ti, V, Cr, Zr, Cr, Nb, Mo, Hf, and Ta)

A: elements in the A group (Al, Si, P, S, Ga, Ge, As, In, Cd, Sn, Tl, and Pb)

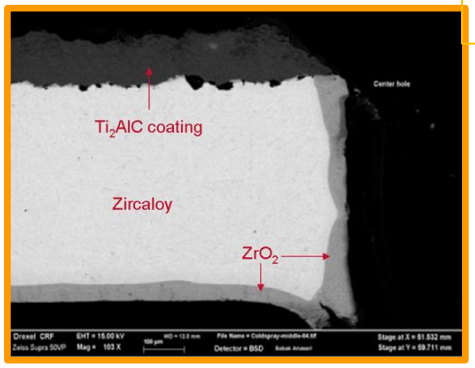
X: carbon or nitrogen

MAX phases have a good:

- Thermal and electrical conductivity
- corrosion resistance
- mechanical properties are maintained at high temperature.

As for example:

Ti_2AlC coating deposited on the sample surface by Cold Spray technique. The coating was adherent to the substrate and shows good high temperature steam corrosion resistance.



Very little oxidation was observed under the coating compared to the un-coated edges of the sample.

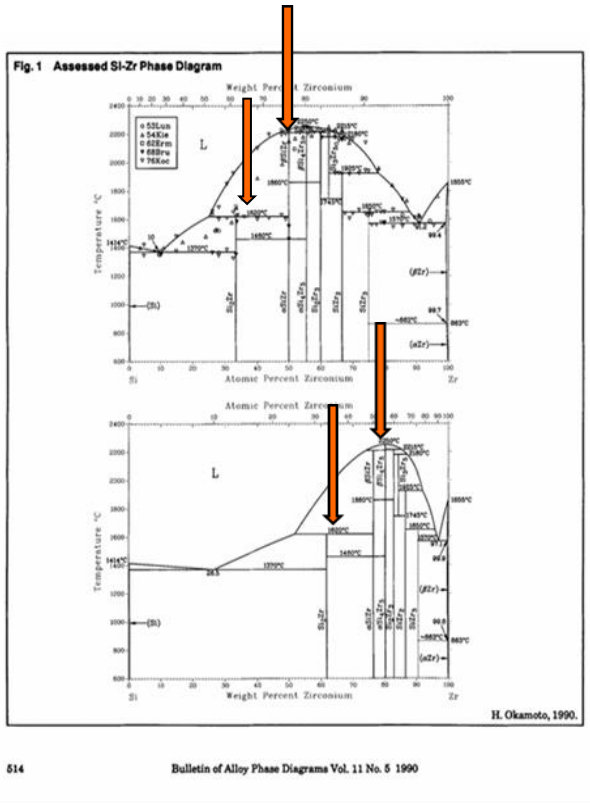
On the base of silicon

WHY SILICON BASE COATINGS

- The **zirconium silicide** or **zirconium silicate** coatings are known for good resistance in high temperature conditions and are applied as environmental barrier coatings for high-temperature gas-turbine components

- **Silicon based coatings** may provide a more protective barrier than the native ZrO2 films formed on alloy cladding during routine nuclear plant operations and may provide an protective barrier during high-temperature accident scenarios

- Both, Zr and Si form their respective oxides at high temperatures and surface layers of ZrO_2 or SiO_2 or even **$ZrSiO_4$** may be formed depending on the stoichiometry



Phase diagram for zirconium-silicon systems: of intermetallic compounds - different Zr/Si ratios and stability regions
For example:
ZrSi2 is stable up to 1620°C
ZrSi is stable up to 2210°C

INCT PROPOSAL
Silicide/silicate coatings on zirconium alloys for improving the high temperature corrosion resistance

⌘ The aim:

to develop the silicon-based coatings on zirconium alloys claddings and evaluation of their properties at accident scenario as well as in the reactor normal working conditions

⌘ The main topics:

- development of deposition methods of silicide or/and silicate coatings on zirconium alloys surface
- determination of corrosion properties of deposited layer in water, steam and air atmospheres at high temperatures.

Materials

Zirconium alloys
(Firmetal, China) in two forms: sheets and rods

Si compounds
$ZrSiO_4$
Si powder
Si, Zr targets
Zr+Si targets
TEOS tetraethylorthosilica

S**ilicide** - any compounds containing silicon bonded to a more electropositive element.
S**ilicate** is a compound containing an anionic silicone compound, the great majority of the silicates are oxides

Characterisations

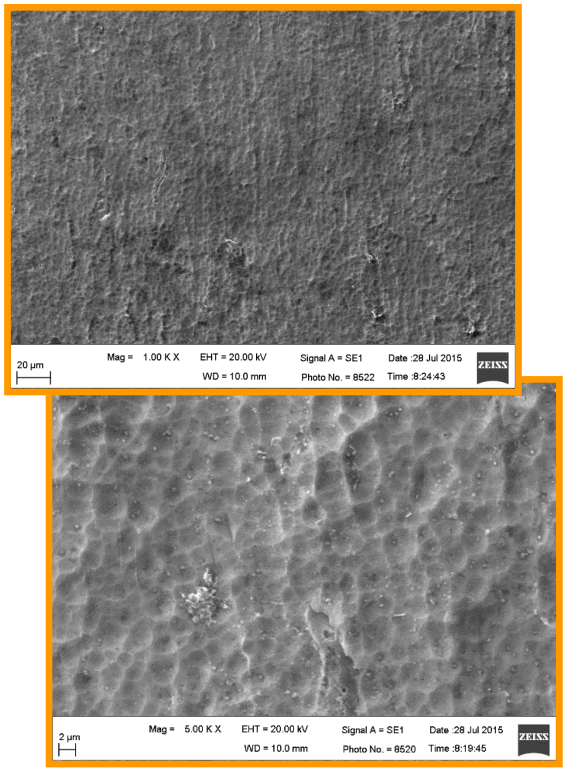
1. Surface morphology: OM, SEMs including HR SEMs
2. Elemental analysis: EDS with Quantax 400 (Bruker, Germany)
3. Phase analysis: X-ray diffractometer D8 Advanced (Bruker, Germany) with geometries Θ-2Θ, ω=5o, ω=10o
4. thermal behaviour: scanning calorimetry + thermogravimetric analysis (DSC / TGA)
5. changes of chemical bonding state of powders: Fourier Transform Infrared Spectroscopy (FTIR)

6. Oxidation tests

6.1. in the furnace up to 1200oC, air, argon
6.2. in the autoclave (360°C, 196 bar)

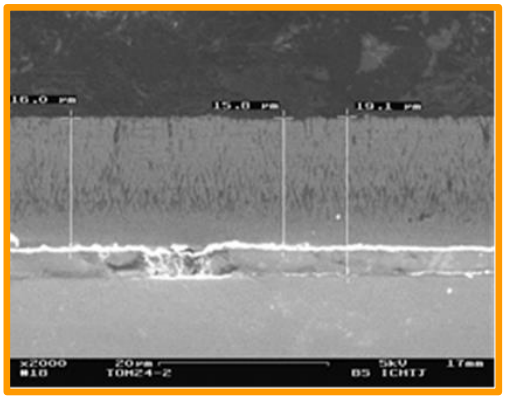
PEO -1

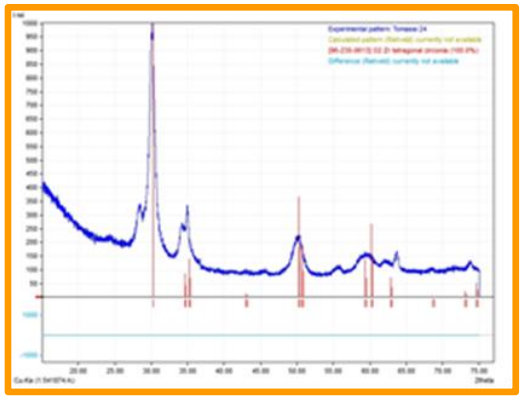
- Plasma electrolytic oxidation (PEO) is an electrochemical surface treatment process for generating oxide coatings on metals.
- PEO experiments with zirconium silicate $ZrSiO_4$ were carried out



Element	norm. C (wt.%)	atom C (at. %)
Zirconium	75.74	33.73
Silicon	**0.42**	**0.6**
Oxygen	15.81	40.13

in progress





Blue line – experiment
Red lines - ZrO_2

Physical Vapour Deposition PVD - 1

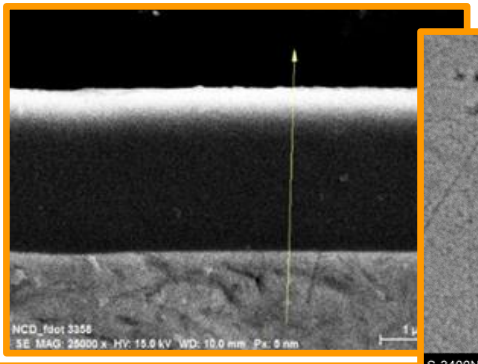


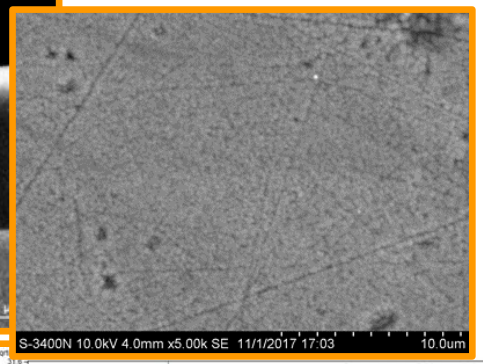


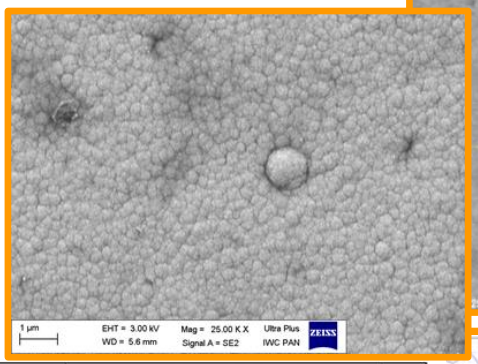
Balzer's system with 3 plasma sources.
Two circular magnetrons were used

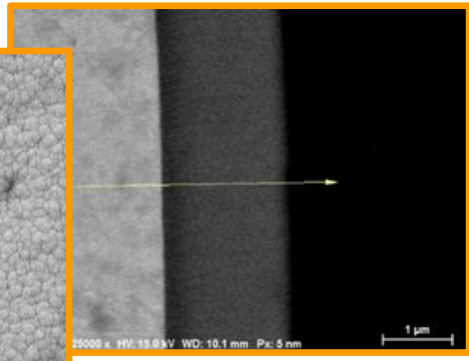
with composite target $ZrSi_2$

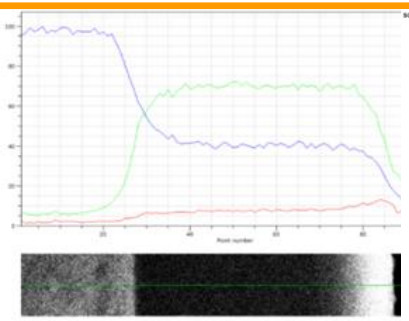
with single element targets Zr and Si

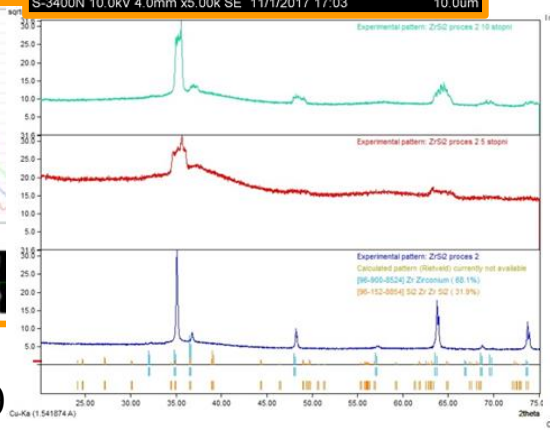


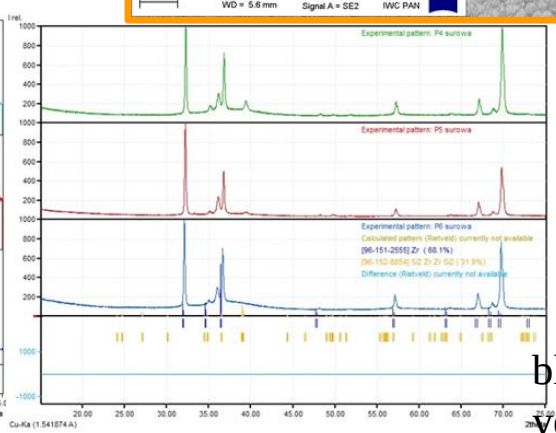


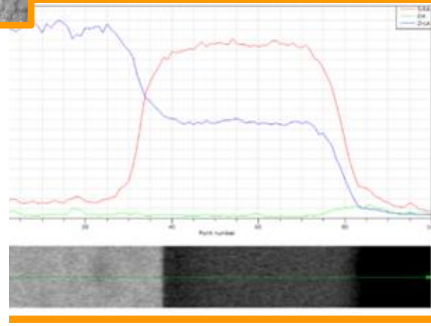










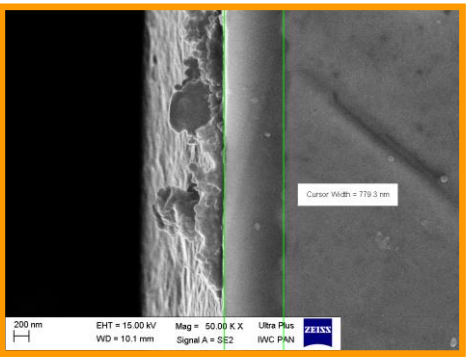


Geometries:
ω=10o, ω=5o, θ-2θ

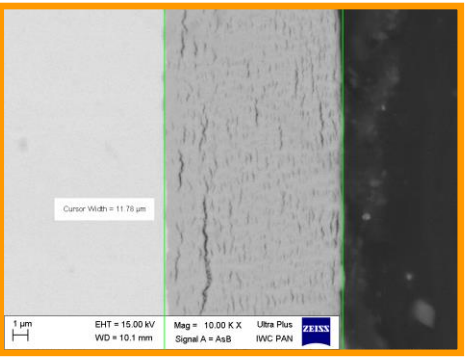
blue – Zr
yellow – ZrSi2

PVD - 3 $ZrSi_2$ target Oxidation tests

autoclave

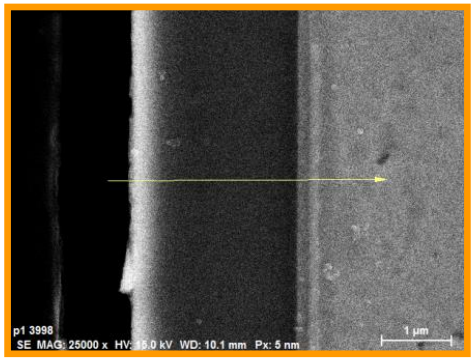


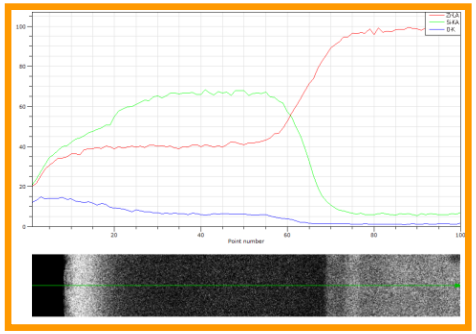
modified surface



unmodified surface
-thicker surface layer

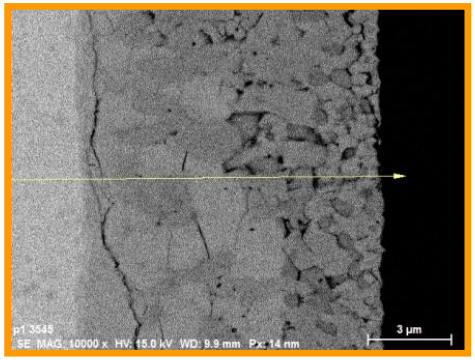
700oC/3h/air

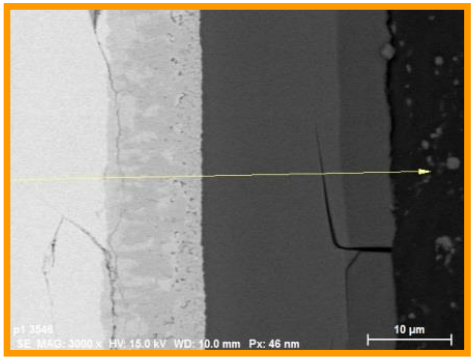


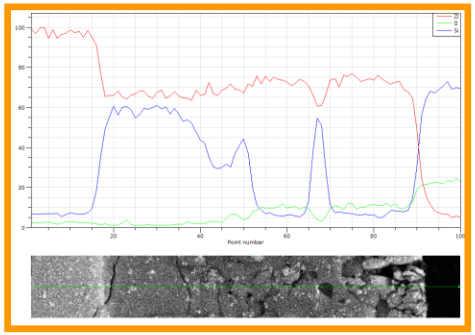


Modified; Si presence confirmed (green)

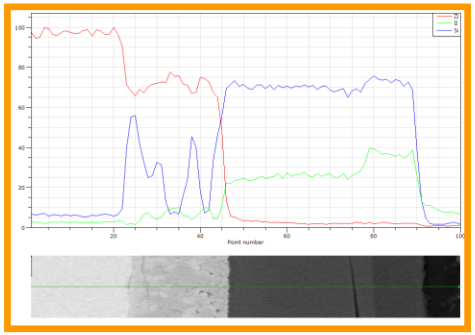
1200oC/1000s/air







Water, Si presence confirmed (blue)



with furnace, Si presence confirmed

PVD - 4

Oxidation T = 700°C/3h/air

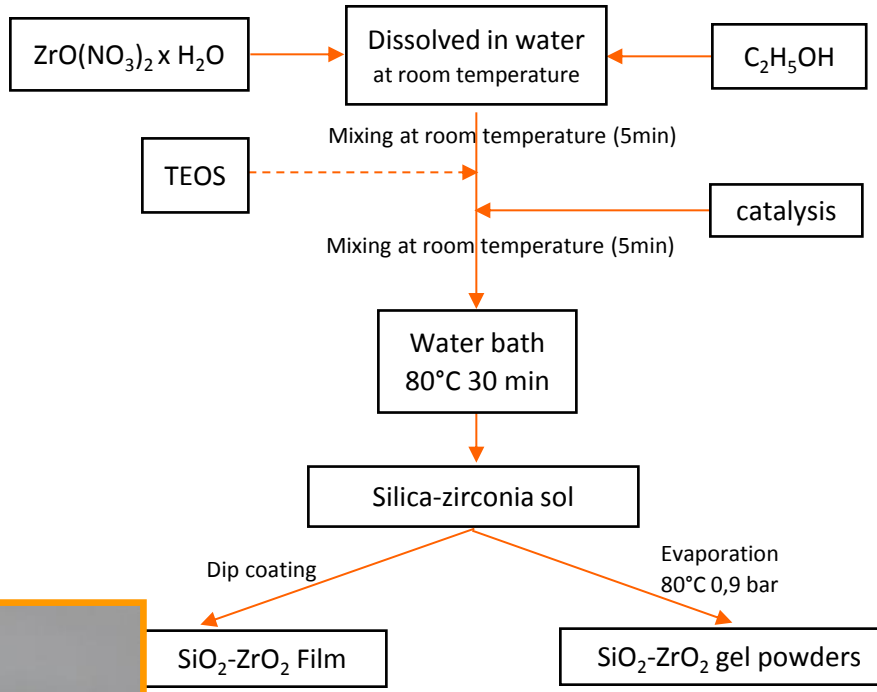
Time [h]	Zr alloy, raw [g]	coated from ZrSi2 target [g]	coated from Zr and Si targets [g]	ZrSi2 powder [g]
1	0.0264	0.0096	**0.0072**	0.3354
2	0.0325	0.0134	**0.0088**	0.3693
3	0.0383	0.0156	**0.0126**	0.3891
4	0.0472	0.0195	**0.0143**	0.4046
5	0.0585	0.0241	**0.0184**	0.4215

Oxidation T=1100°C/1000 s

	Δm
Zr alloy, raw	0.3039
Coated from $ZrSi_2$ target	**0.2159**
Coated from Zr and Si targets	0.2216

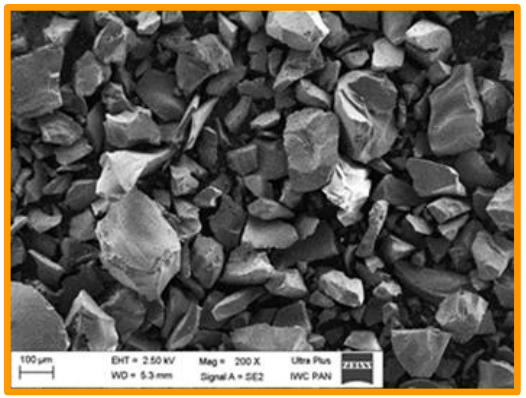
SOL-GEL -1

SiO_2–ZrO_2 Synthesis







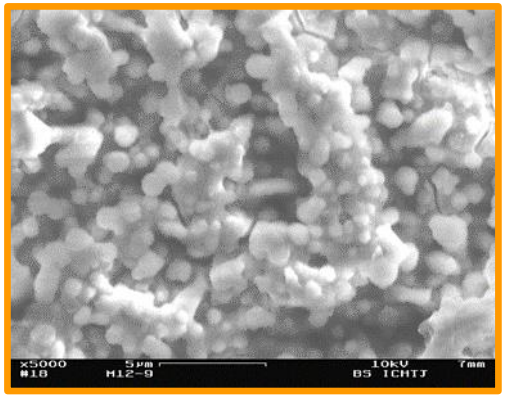


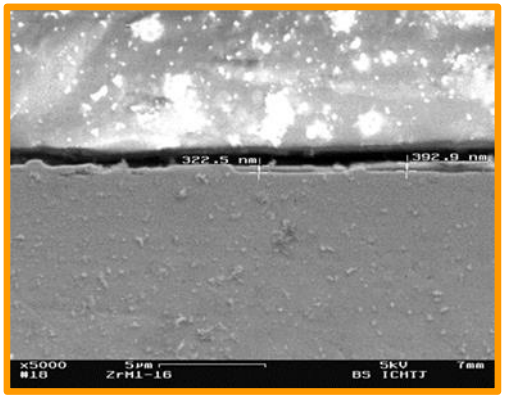
SOL-GEL - 2

Surface and cross-section of the silicon based compound layer on the zirconium alloy

Oxidation tests – autoclave 7 days

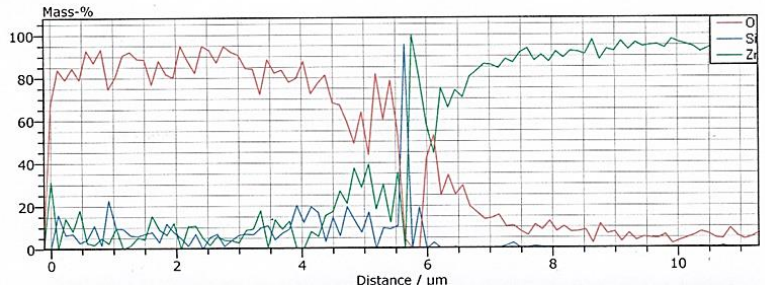
500oC calcined

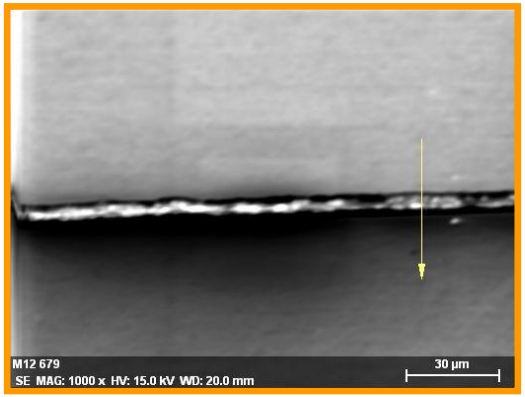


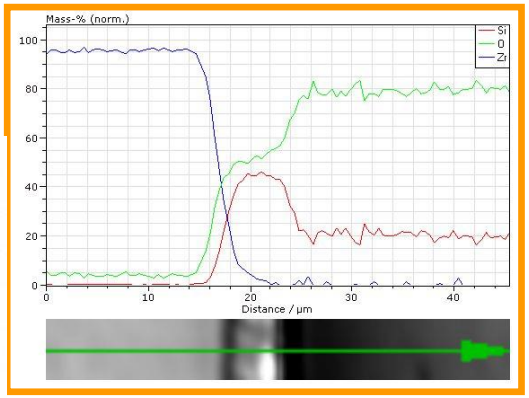


EDS results - surface

Element	Conc. (wt.%)	Conc. (at. %)
Zirconium	9.33	2.14
Silicon	**37.05**	**27.64**
Oxygen	53.62	70.23



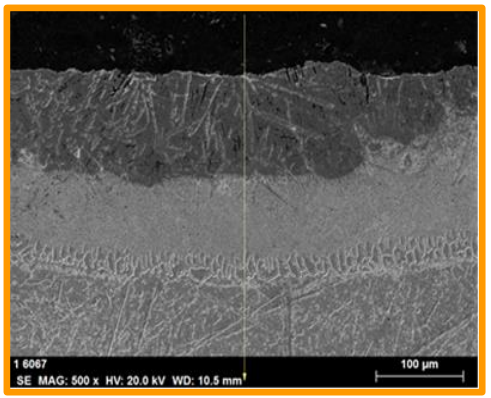


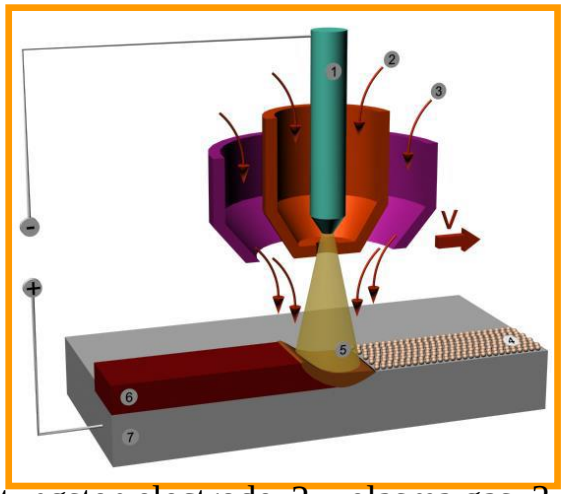


TUNGSTEN INERT GAS (TIG) - 1

Tungsten inert gas (**TIG**) **welding**, is an arc welding process that uses a non-consumable tungsten electrode. The weld area is protected from atmospheric contamination by an inert shielding gas, here: argon.

Re-melting process parameters: electrode from tungsten doped with thorium, angle - 40º; argon as the protective gas with 10 l/min flow; currents in the range of 20 - 90 A; electrode feed in the range of 20 – 40 mm/min.

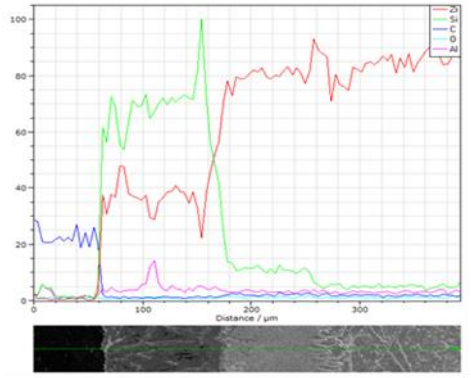




1 – tungsten electrode, 2 – plasma gas, 3 – protective gas, 4 – powder/suspension, 5 – plasma arc, 6 – coating, 7 – remelted base material

Slurries prepared from Si, Zr+Si, $ZrSiO_4$ powders in 10% solution in PVA

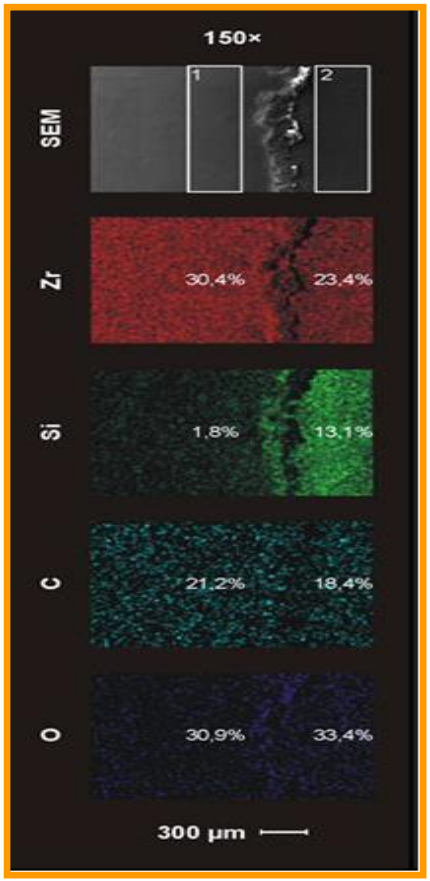
Element	norm. C (wt.%)	atom C (at. %)
Zirconium	96.01	81.89
Silicon	**1.63**	**2.70**
Oxygen	8.38	24.31

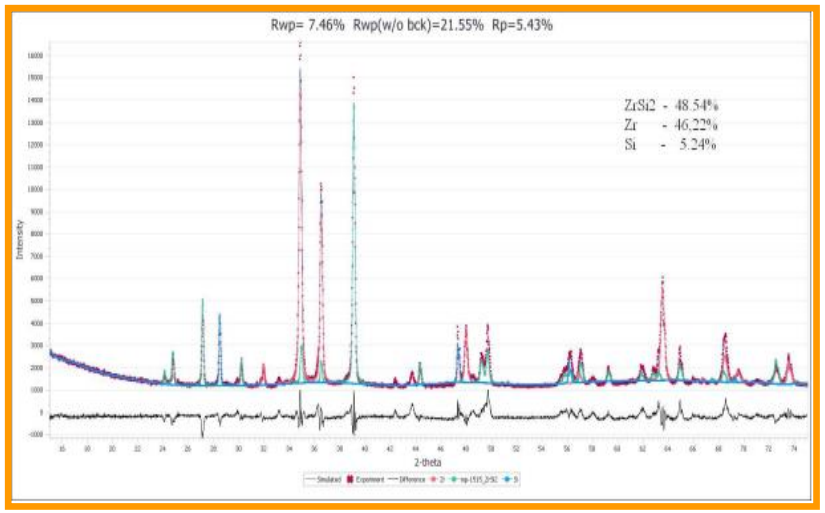


Si – green line

TIG - 2

Elements presence in areas:
(1) coated and remelted
(2) coated

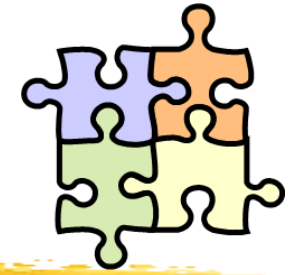




XRD diffraction pattern
sample coated with Si powder suspension in the 10% PVA water solution followed by re-melting with the 40 A current

Presence of Zr, $ZrSi_2$ and Si was confirmed

SUMMARY



- Several potential approaches for development of ATMs exist that could lead to improved accident tolerance in LWR fuel systems
  - Various options are exploring and offering basic guidelines for ATM concepts. Each of these options has known or potential shortcomings
    - Accident progression is dominated by the integral system response during such events. ATMs can affect the sequence and rate of this progression, and providing enhanced coping time during such events. The safety margins for nuclear power systems can be enhanced.

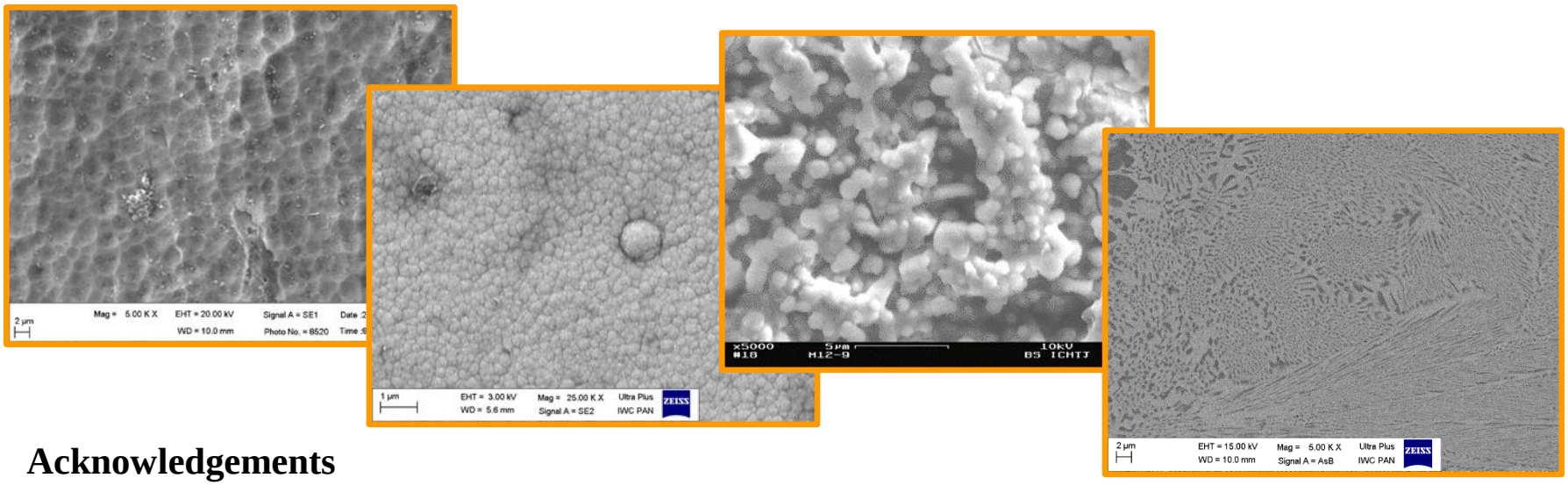
Zirconium coatings with compounds on the base of silicon was obtained using

- PVD
- sol-gel diping technique
- plasma electrolitic oxidation PEO
- TIG

2. Presence of Si in modified surface layer was confirmed
3. First oxidation test showed improvement of oxidation resistance after proposed surface layer modification
4. Next step: more oxidation tests
4. Next step: improvement of the obtain coatings on the base of Si

ACCIDENT TOLERANT MATERIALS – IDEAS and RESEARCH DIRECTIONS

Thank you for your attention



Acknowledgements

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IAEA International Atomic Energy Agency

Ministerstwo Nauki i Szkolnictwa Wyższego

Chromium and Chromium Nitride Coated Cladding for Nuclear Reactor Fuel

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[1]*UJP PRAHA a.s., Czech Republic*
[2]*Faculty of Nuclear Science and Eng., Czech Republic*
[3]*Massachusetts Institute of Technology, Cambridge, USA*
[4]*Faculty of Mechanical Engineering, Czech Technical University in Prague, Czech Republic*

Abstract

Accident tolerant fuels (ATF) have been widely studied since the Fukushima accident in 2011. A coating technology for nuclear fuel cladding has been considered to decrease the high-temperature oxidation rate of zirconium-based alloys and to enhance accident tolerance of the cladding. This study is focused on high-temperature oxidation of Zr-1%Nb cladding alloys coated with Cr and CrN layers by PVD technique. Coated and reference uncoated samples of a Zr-1%Nb alloy were tested in the high-temperature steam environment at temperatures in the range between 1000 and 1400°C. The comparison between both types was made. The high-temperature oxidation tests showed a protective behavior of the coating in the steam environment. Optical microscopy, scanning microscopy (EDS, WDS) were used for the evaluation of microstructure and chemical composition after the high-temperature oxidation tests.

Keywords: *ATF, coatings, chromium nitride, chromium, LOCA.*


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Czech Technical University in Prague

NEW TECHNOLOGIES RESEARCH CENTRE
UNIVERSITY OF WEST BOHEMIA

Experimental Behaviour of Chromium Based Coatings

J. Krejčí[1,2], J. Kabátová[1], F. Manoch[1], J. Kočí[1], L. Cvrček[2], S. Krum[2], J. Kadlec[2], P. Šutta[3], M.Ševeček[2]

[1] UJP, [2] CTU, [3] NTC UWB



23rd International QUENCH Workshop

Karlsruhe Institute of Technology, Germany

October 17-19, 2017

Introduction

- Experimental research of fuel cladding tubes has been carried in UJP PRAHA since 1971
  - Different Zr-alloys: E110 (many variants), E635, Zry-2, Zry-4, ZIRLO
  - Corrosion experiments, high temperature oxidation (steam or air), burst-test, spent fuel dry storage
- Cr-based coatings research
  - In cooperation with CTU in Prague
  - Started in the end of 2016
- Presentation outlines:
  - Material & deposition
  - Long-term corrosion experiment
  - High temperature oxidation
  - Coating/substrate interface
  - Experiments beyond Cr-Zr eutectic point (1330°C)

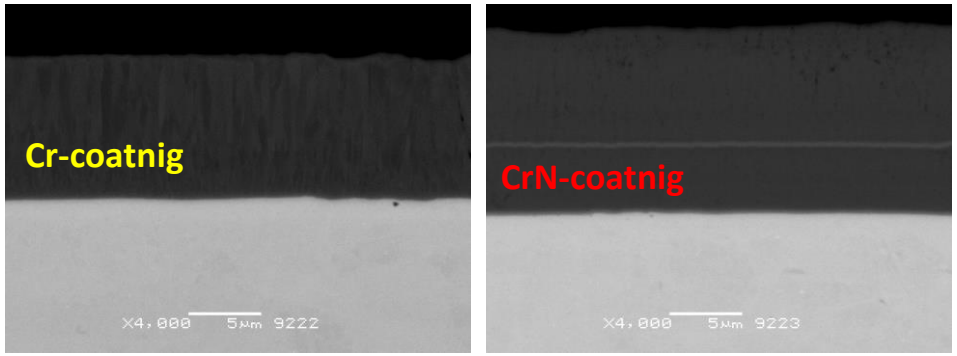
Material & deposition

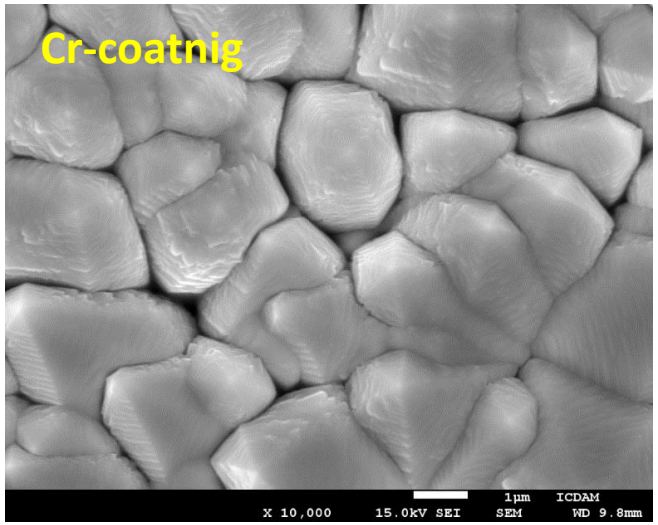
- Speciemens geometry:
  - Double-sided oxidation: ∅ 9.1 × 7.9 mm, length 30mm, 15mm
  - Single-sided oxidation: ∅ 9.1 × 7.9, length 75mm + endplugs
- Substrate:
  - E110 iodide/electrolytic
  - E110 sponge based
- Deposition (CTU):
  - Hauzer Flexicoat 850
  - Coating from outer side of speciemens only
  - **Layers:**
  - Magnetron sputtering: Cr (10-30mic)
  - Reactive magnetron sputtering: CrN (5-15mic), Cr_xN_y (substochiometric, 20mic)
  - Multi-layer: Cr + CrN (20mic)

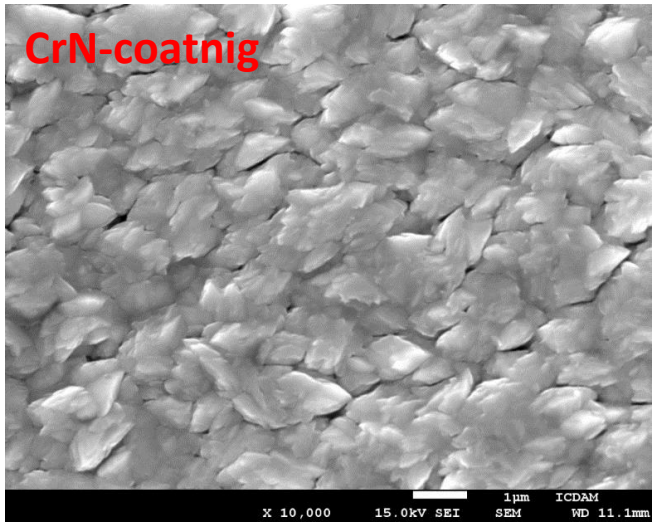


Cr & CrN coatings: as-received state

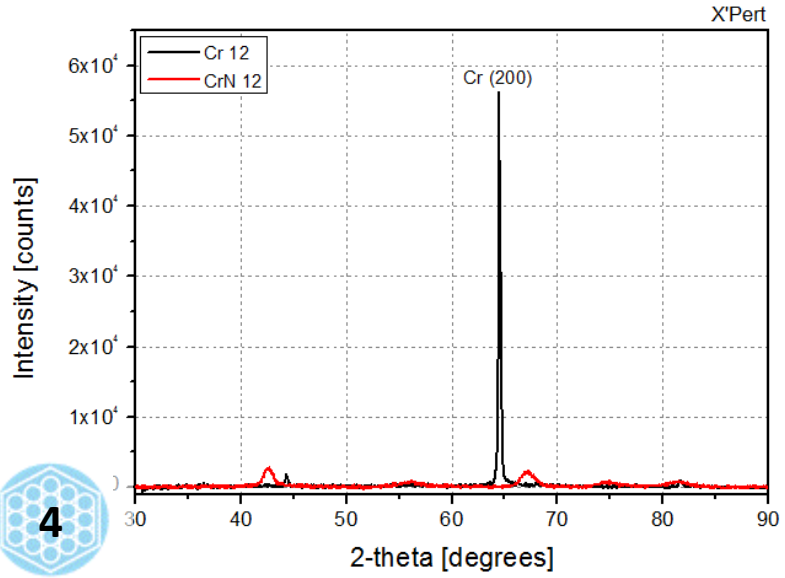
- SEM: polished cross-section, surface, fracture

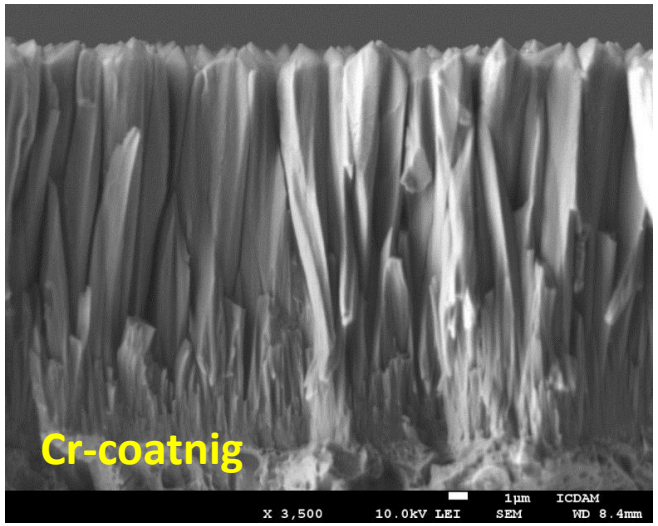


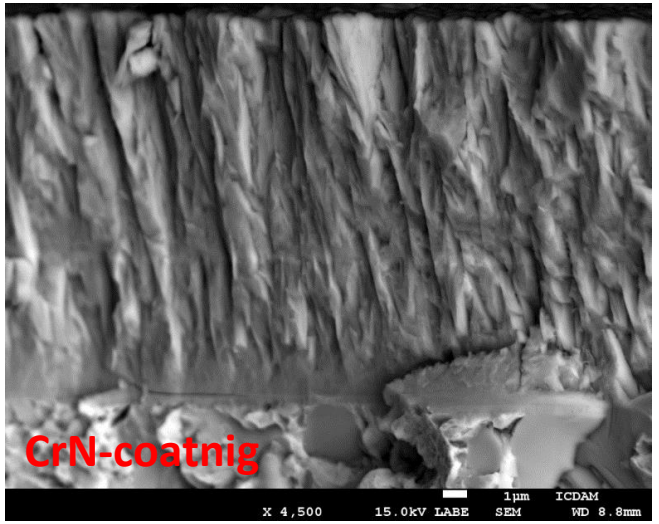




- X-ray diffraction (at RT):
  - Cr-coating: strongly oriented texture





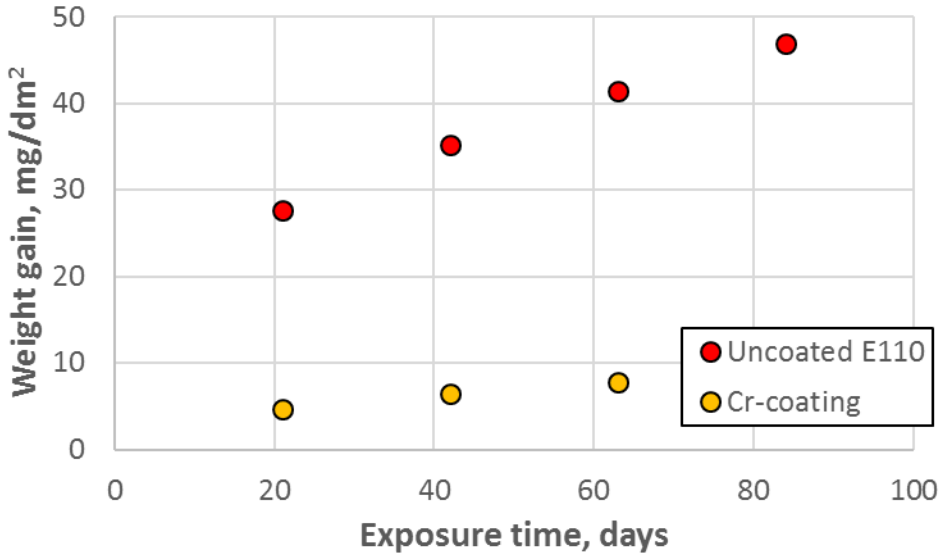


Corrosion experiment

- <u>On-going experiment:</u>
- Autoclave: 4dm^3
- 360°C/197bar, WWER conditions
- Period duration: 21 day
- Single-sided oxidation (75mm long + welded endplugs)
- 10 uncoated samples & 5 Cr-coated samples (63 days)



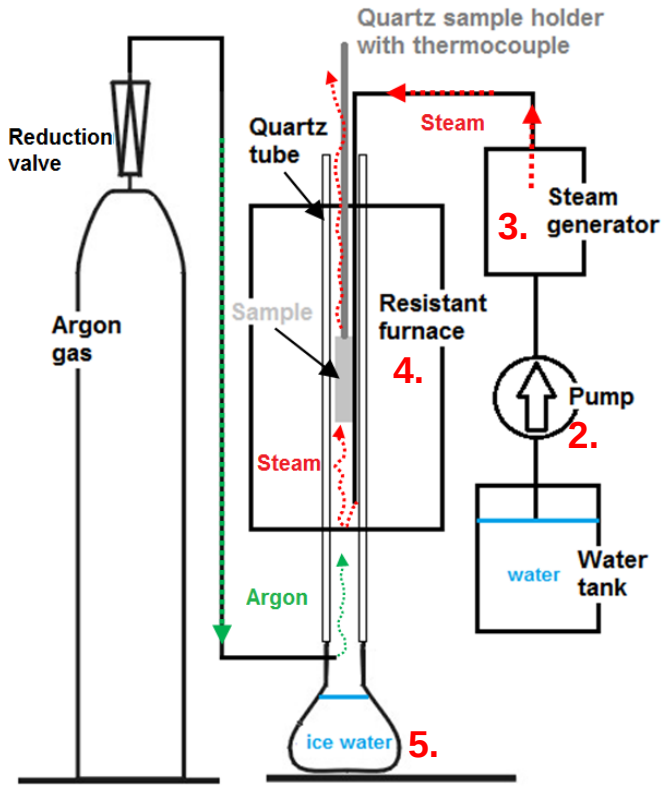


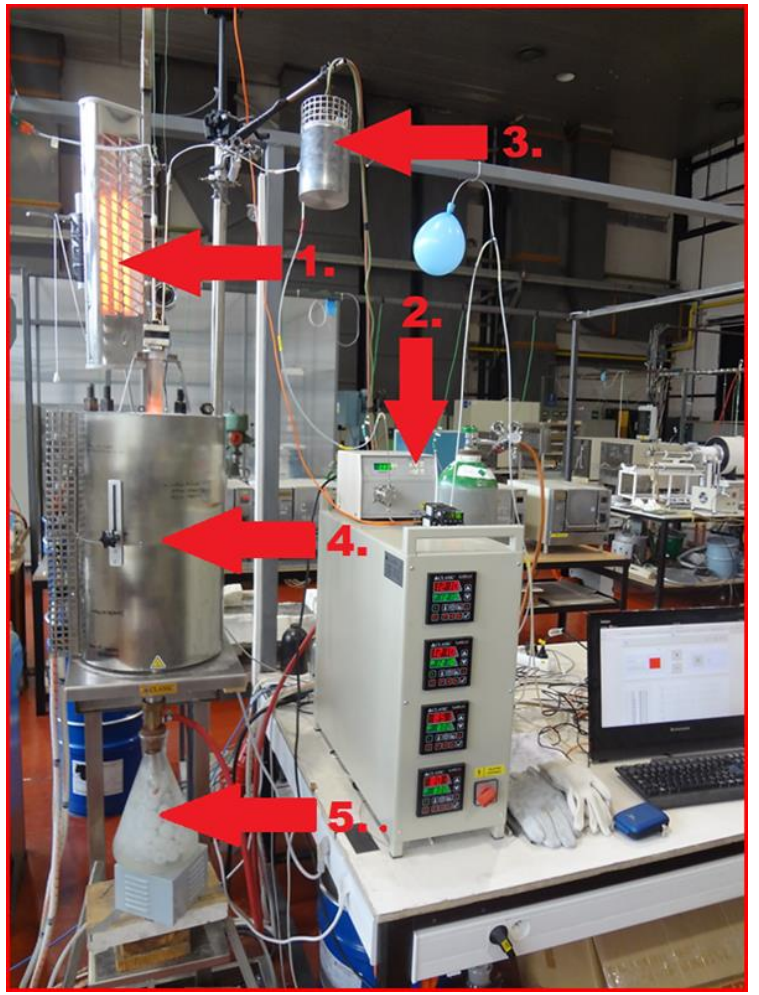


High temperature oxidation facility

UJP PRAHA

- Resistant furnaces up to 1700°C (CLASIC CZ)
- Single-sided & double-sided oxidation
- Steam + Argon environment
- Experiments in range 1000-1450°C







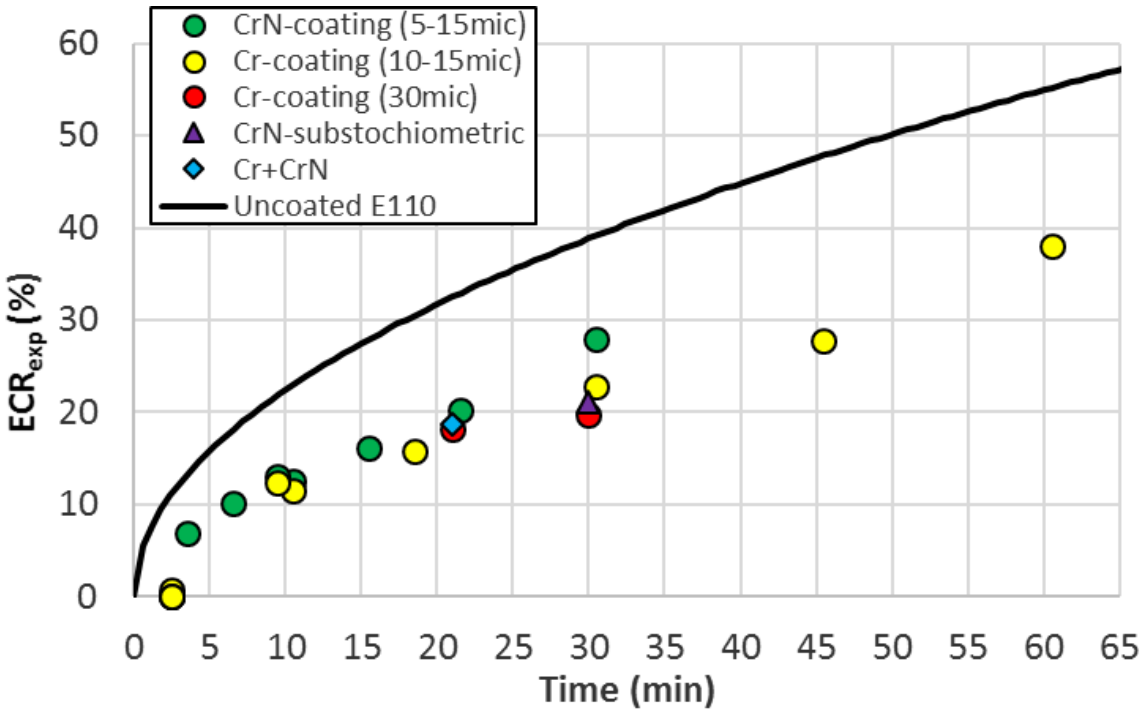
Sample-holder detail



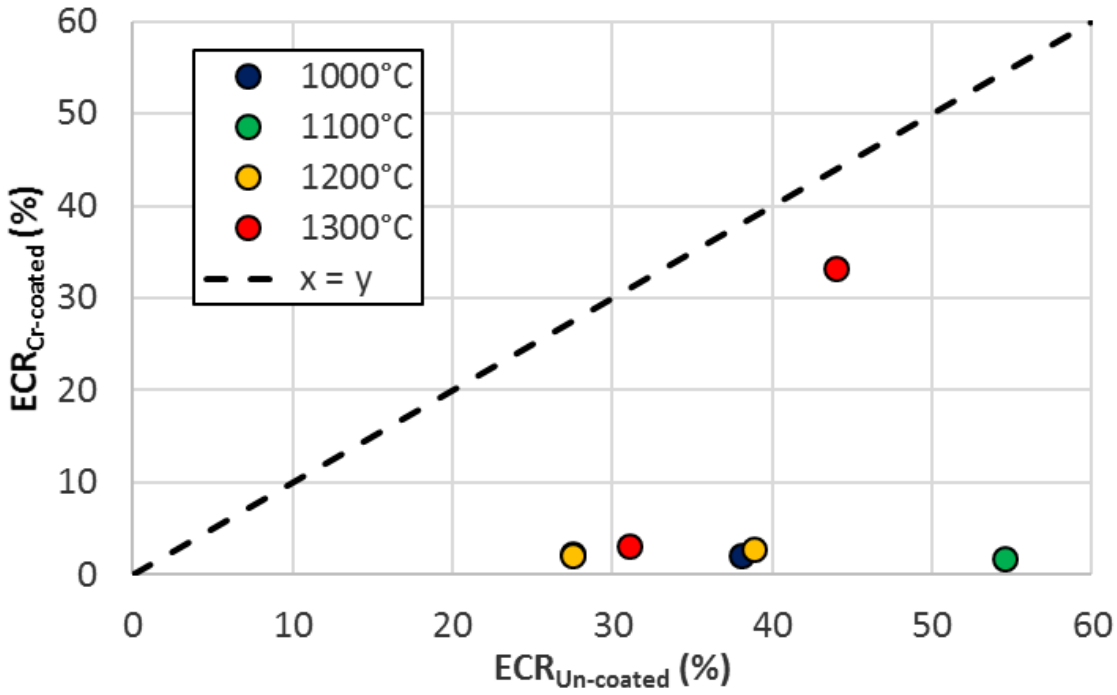
Oxidation kinetics

- Experiments in temperature range 1000-1300°C
- Significantly lower weight gain (ECR_{exp}) for all variants of coated samples

Double-sided oxidation 1200°C (only outer side coated)



Single-sided oxidation

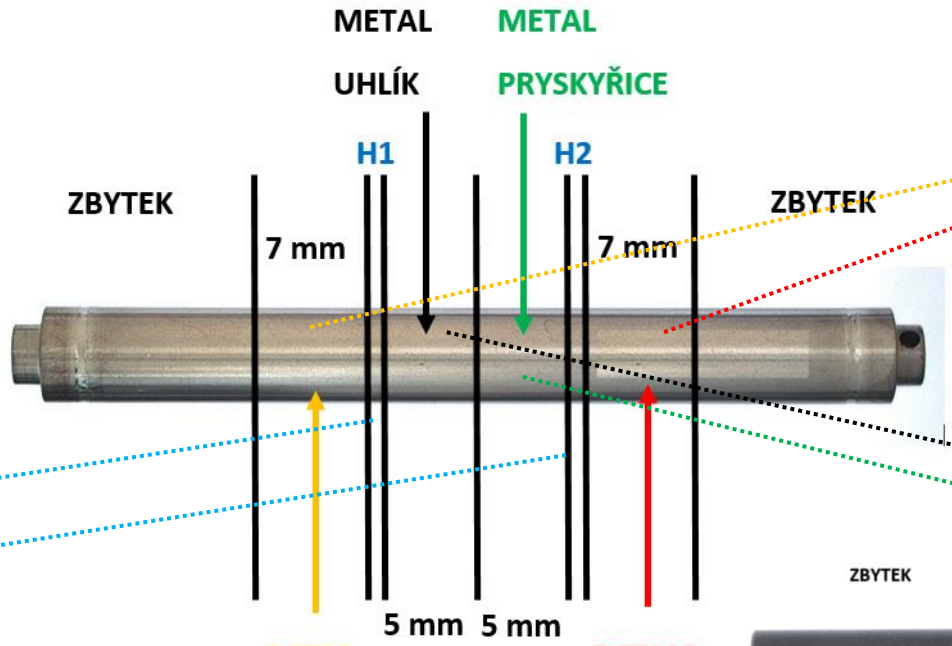


Post-experimental examination

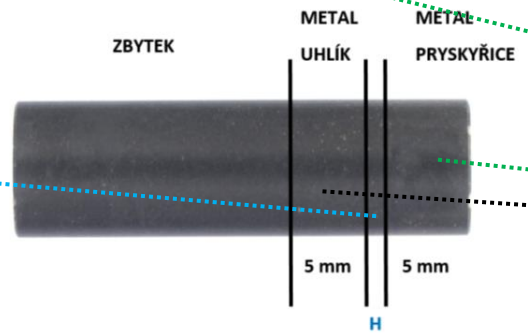
- **Sample cutting:**
- METAL: methalography & microhardness evaluation, EDS, WDS
- RCT20 & RCT135 : ring compression tests at temperatures 20 & 135°C
- H, H1 & H2: hydrogen content measurements
- ZBYTEK: remaining part

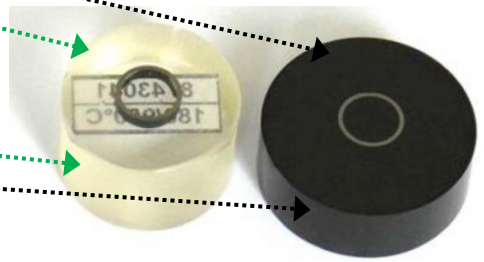






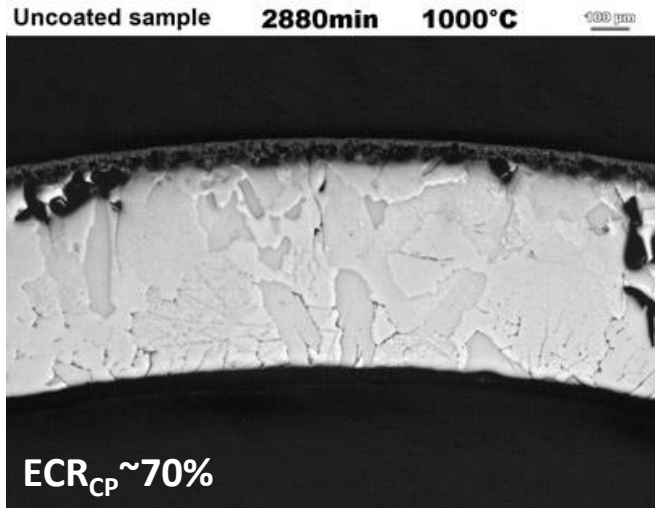


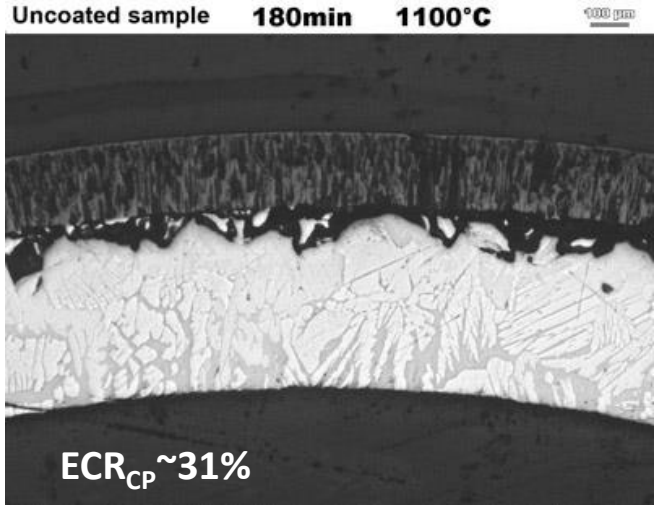


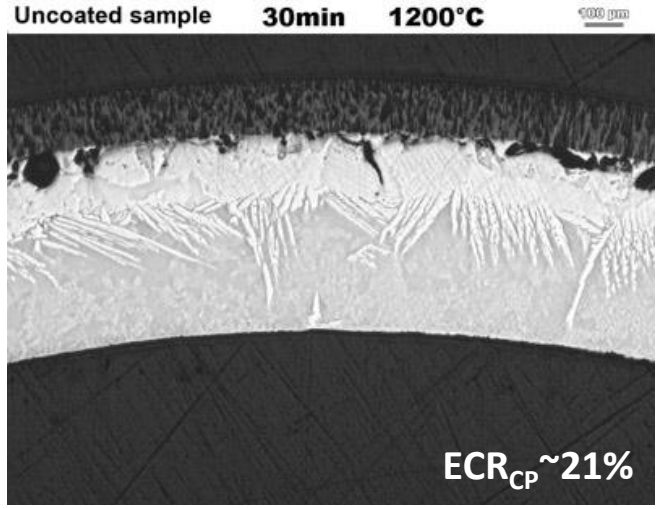


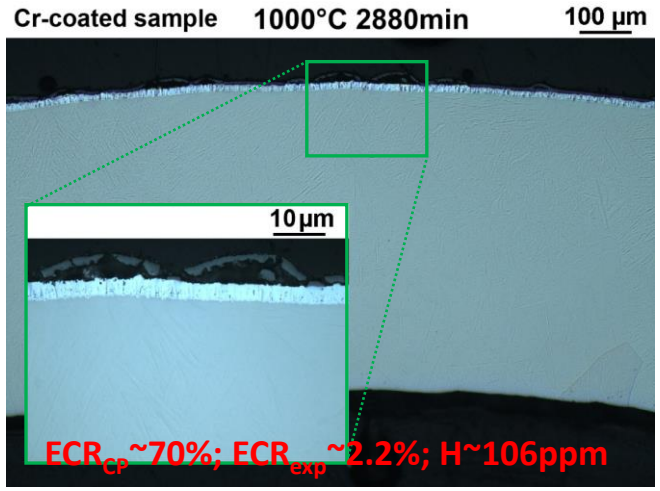
Microstructure after high temperature ox.

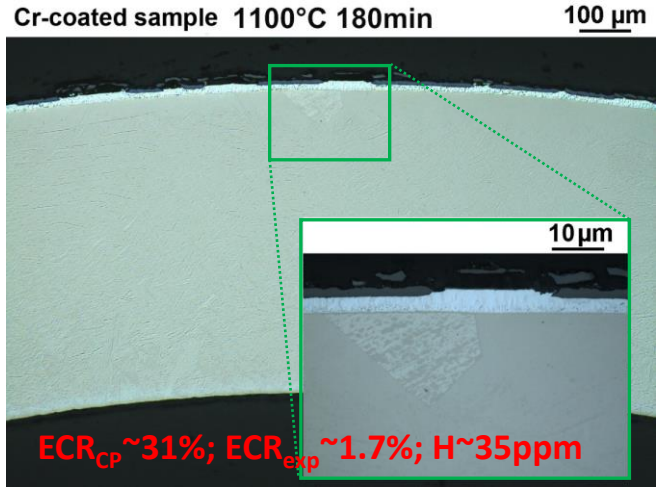
- Single-sided oxidation: Comparison uncoated and Cr-coated samples
  - No visible typical α-Zr(O) grains for Cr-coated samples

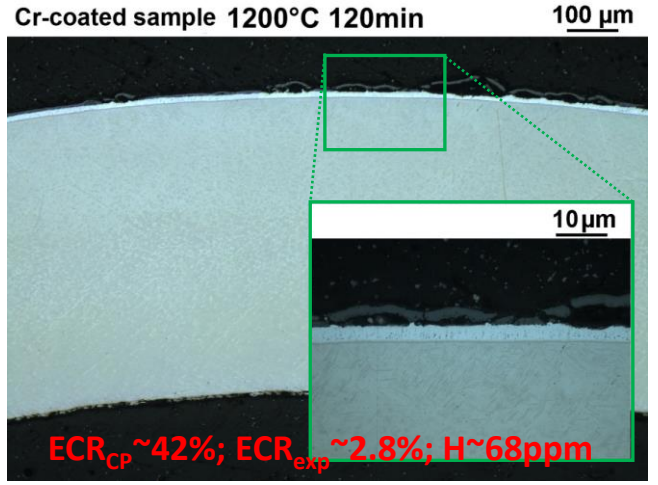








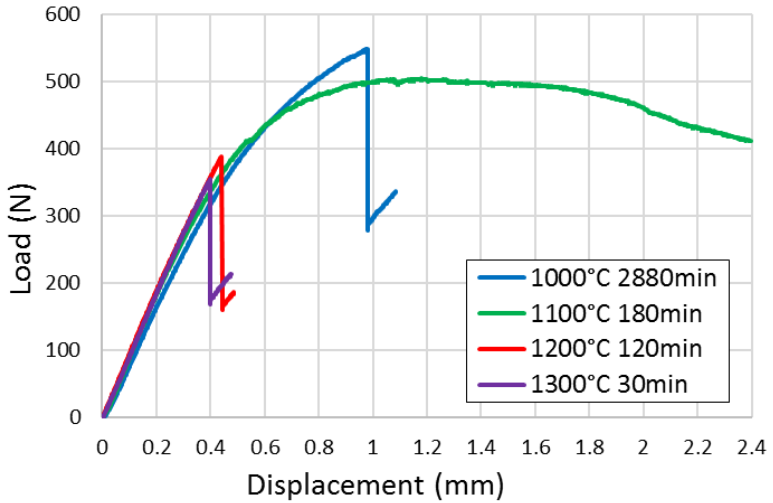




Ring compression test (RCT)

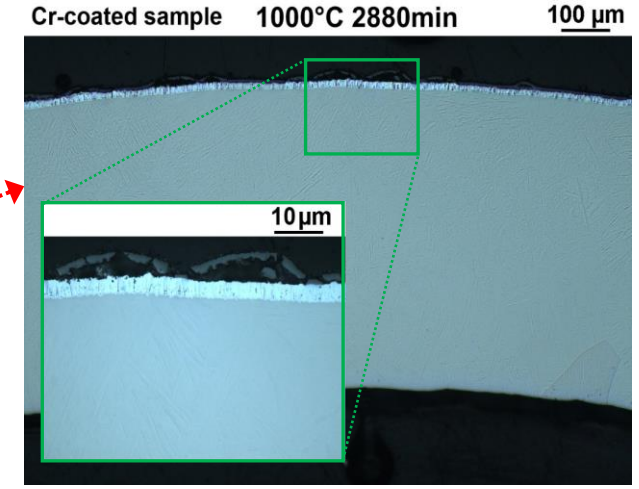
UJP PRAHA

- At temperatures 20 & 135°C



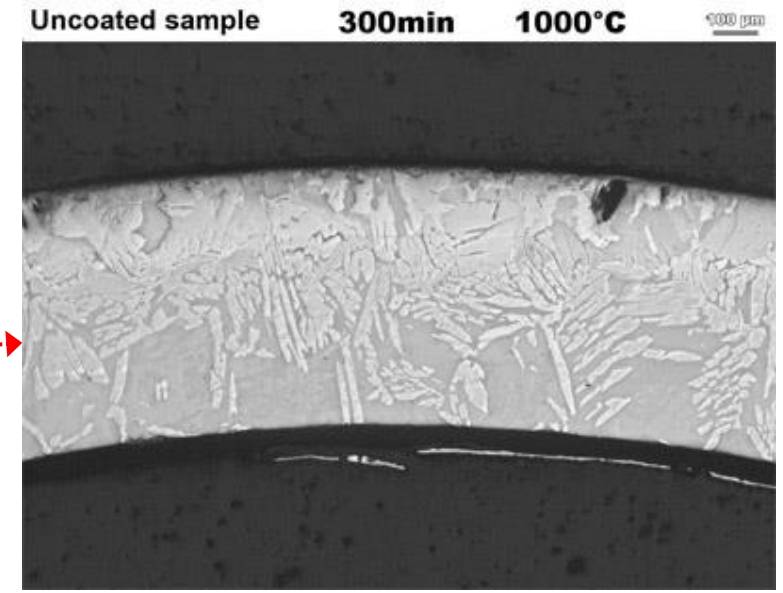
Cr-coated samples

Temperature, °C	1000	1100	1200	1300
Exp. time, min	2880	180	120	30
ECR_{CP}, %	70	31	42	32
ECR_{exp}, %	2.2	1.7	2.8	3.2
Hydrogen, ppm	106	35	68	9
$Ductility_{RCT20°C}$, %	3.7	8.4	0.39	-
$Ductility_{RCT135°C}$, %	4.0	> 16	1.4	0.65



Uncoated samples

Temperature, °C	1000	1000
Exp. time, min	60	300
ECR_{CP}, %	11	23
ECR_{exp}, %	8	15
Hydrogen, ppm	120	12
$Ductility_{RCT135°C}$, %	> 16	3.8



- Evaluation:
  - Maximal load, ductility, …
- Embrittelment criteria ???
  - Coated samples:
  - Increase of copping time
  - ↓ ductility for the same ECR_{exp} (weight gain)

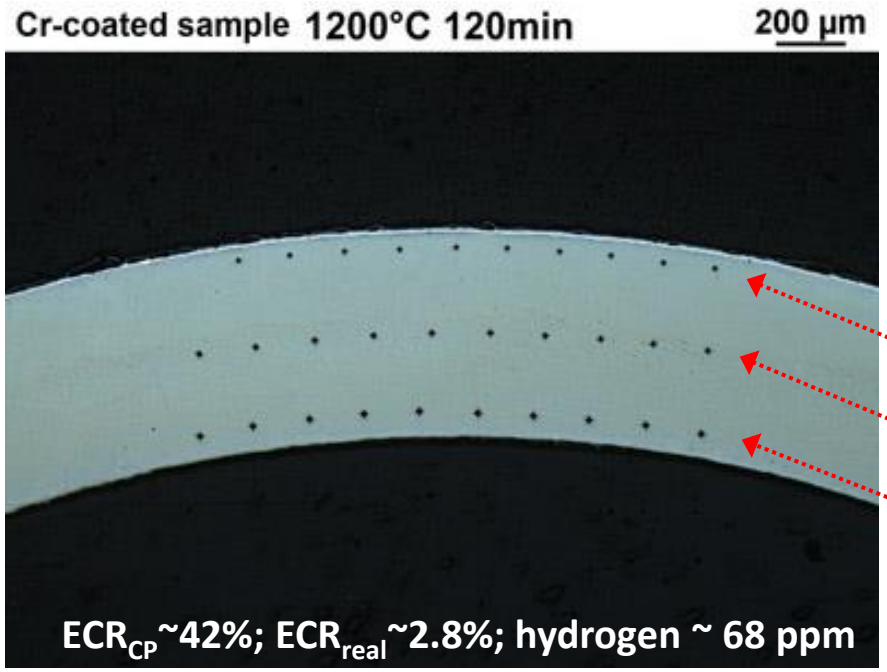
=> it is not possible to compare WG only without any mechanical testing

Microhardness

- Vickers indenter for measurements was used
- Applied load (100g) in 30 positions (3x 10)
- Microhardness: line average ± standart deviation
  - For 1100-1300°C observed HV gradient
  - Higher temperature, higher HV
  - For coated samples higher values of HV for lower ECR_{exp} (in „prior β-Zr“)
  - Microhardness ~ oxygen content => oxygen gradient (oxygen content analysis need to be performed)

Uncoated samples

Temperature, °C	1000	1100	1200	1300
ECR_{exp}, %	5.5	6.5	12	17
HV: line-3	220	200	250	300

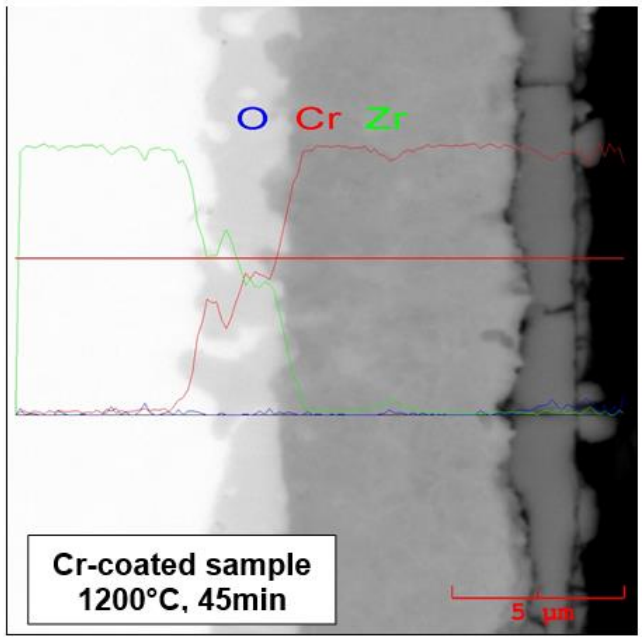


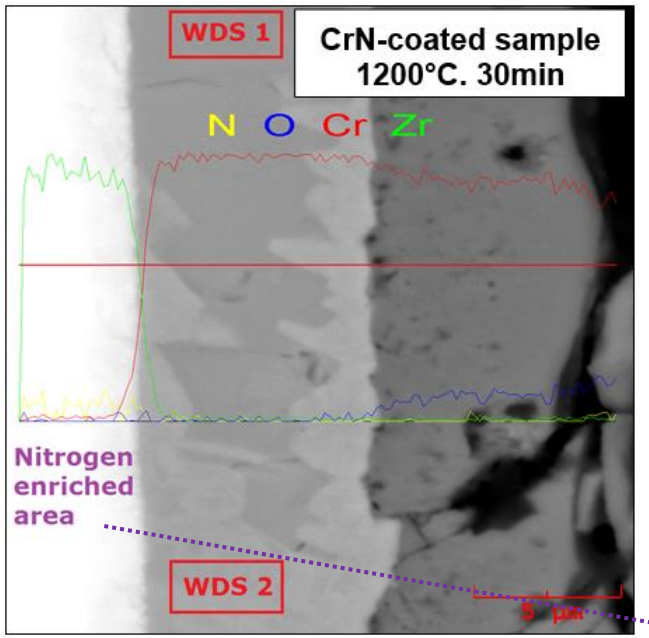
Cr-coated samples

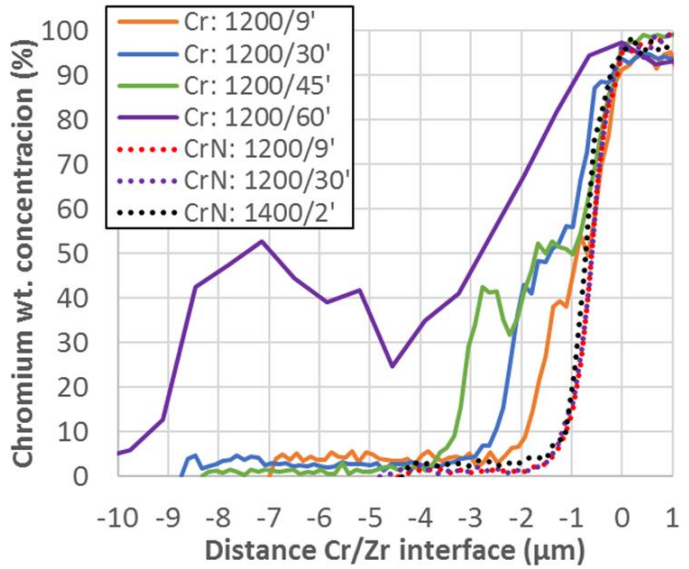
Temperature, °C	1000	1100	1200	1300
Exp. time, min	2880	180	120	30
ECR_{CP}, %	70	31	42	32
ECR_{exp}, %	2.2	1.7	2.8	3.2
HV: line-1	349 ± 8	502 ± 26	507 ± 9	514 ± 39
HV: line-2	351 ± 6	304 ± 2	376 ± 8	451 ± 11
HV: line-3	349 ± 3	234 ± 4	305 ± 5	347 ± 8

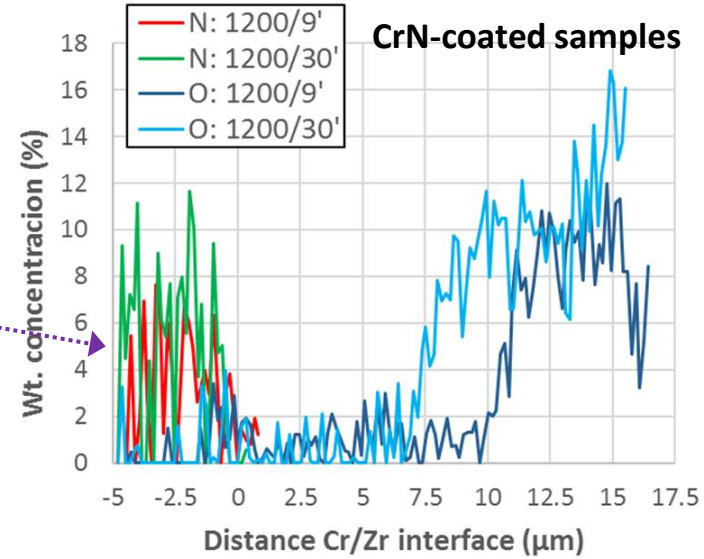
Coating/substrate interface

- EDS line-scan and WDS analysis: Zr, Cr, O, N content
- Cr: diffusion Cr to Zr-substrate => Cr-Zr mixed area
- CrN: in coating detected oxygen enriched areas, ZrN area







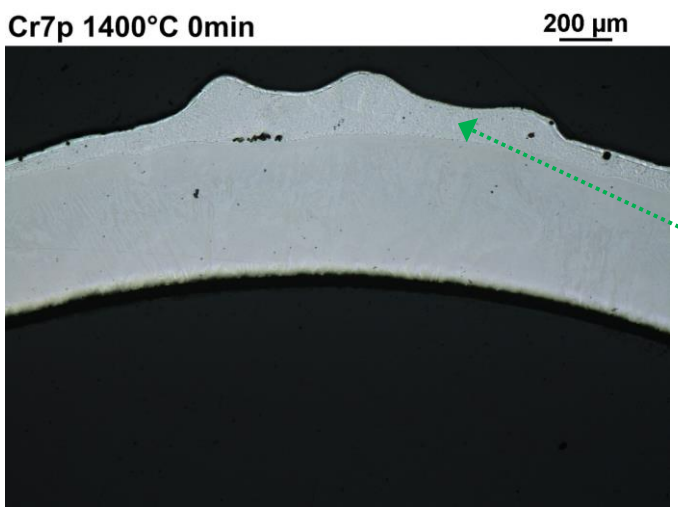


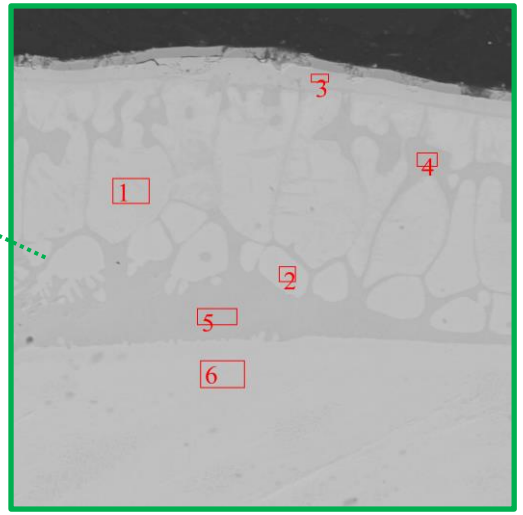
CrN-coated sample	N (wt.%)	Cr (wt.%)	O (wt.%)
As-coated state	20.7 ± 0.9	77.7 ± 0.8	-
1200°C/30min: "dark " area (WDS 1)	20.7 ± 0.7	78.4 ± 0.7	-
1200°C/30min: "light " area (WDS 2)	15.4 ± 0.6	71.5 ± 1.1	11.7 ± 0.9

Experiments beyond Cr-Zr eutectic point (1330°C)

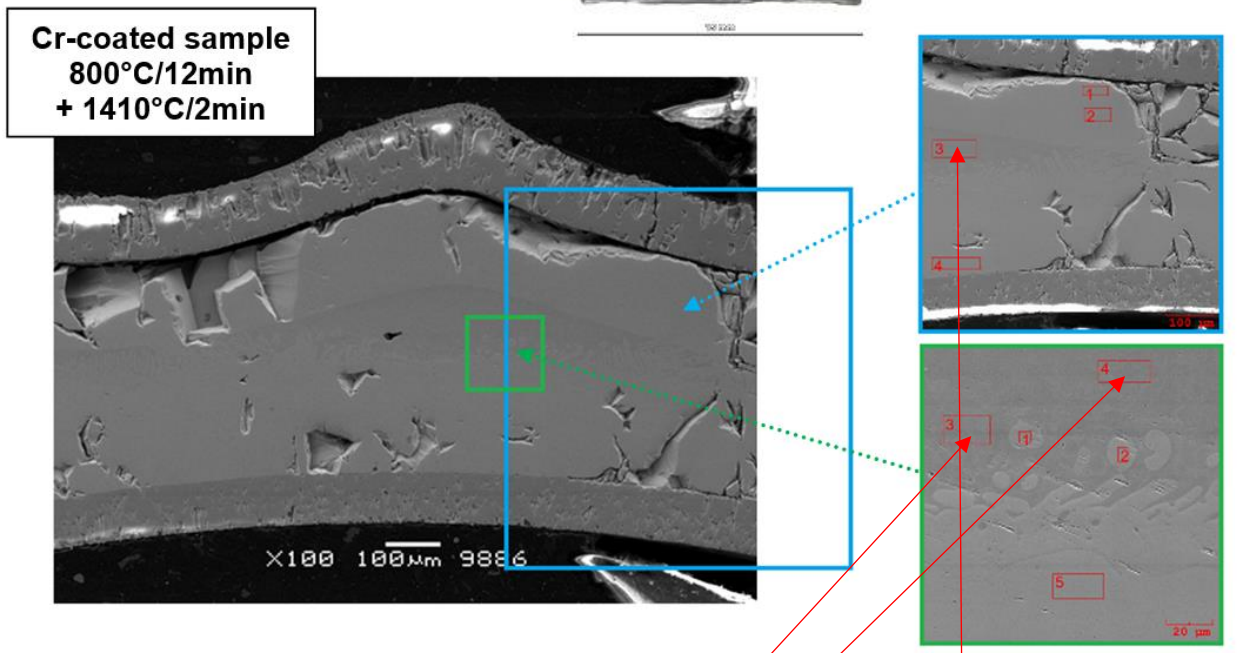
UJP PRAHA

- double-sided argon: heatin rate 50°C/min + 1410°C/0min + quench
  - Cr-coating 10mic (argon + quench):
  - Cr-coated surface melted (~150mic of wall thickness)









- double-sided steam oxidation: 1350-1420°C
  - Cr-coating 10mic (double-sided steam oxidation):
  - Cr-coated surface melted & deformed

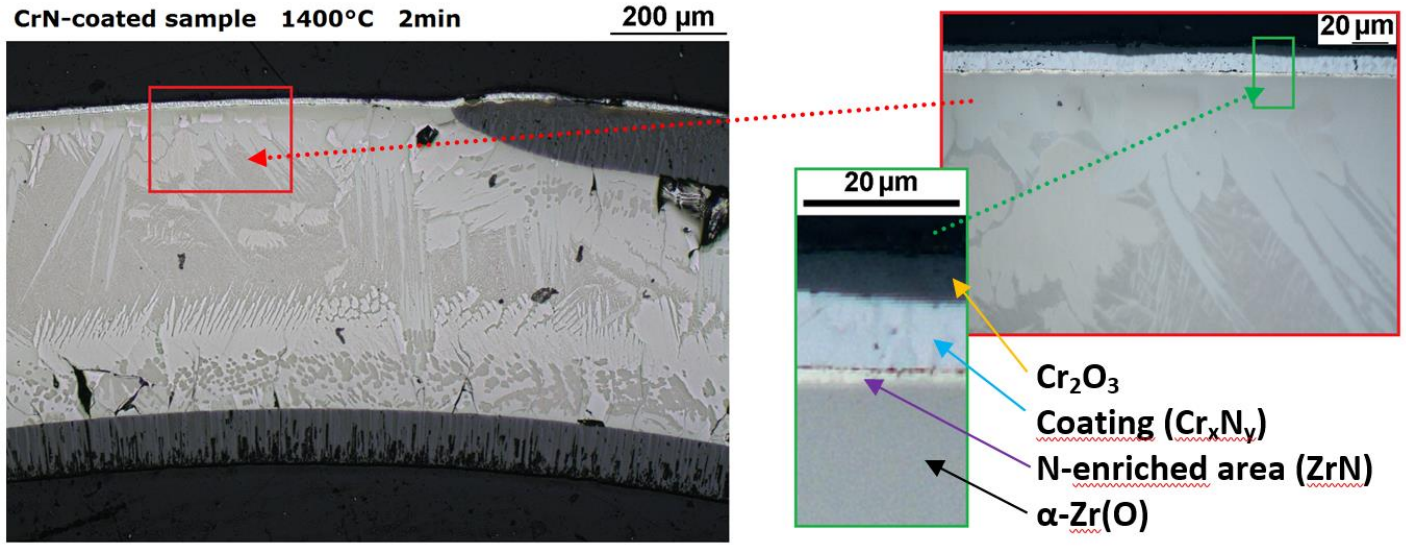
  ⇒ higher oxide-layer thickness
  - EDS: Cr detected in prior β-Zr only (in the middle of the wall)

  => <u>Cr diffusion from oxygen enriched area</u>

Location of analysis	"Green" area		"Blue" area	
	Cr (wt. %)	Zr (wt. %)	Cr (wt. %)	Zr (wt. %)
1	0.2	99.8	0.11	99.9
2	0.4	99.6	0.3	99.7
3	15.8	84.2	15.5	84.5
4	15.2	84.8	0.1	99.9
5	0.1	99.9	-	-

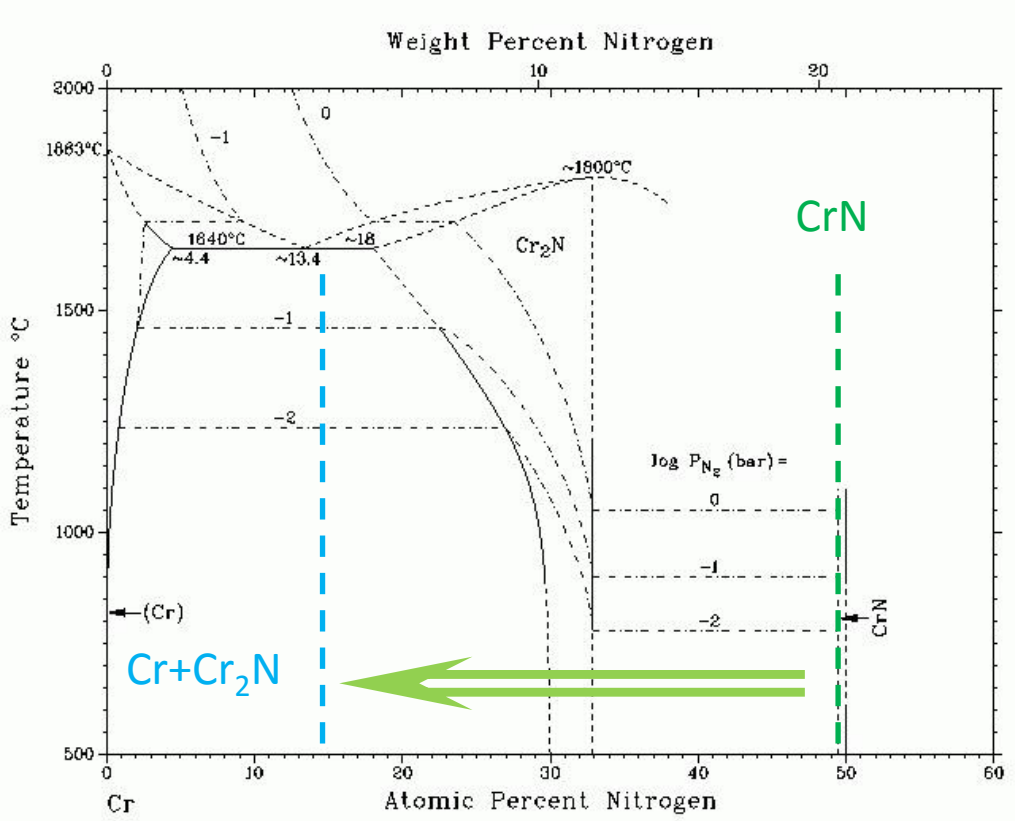
Experiments beyond eutectic point CrN

- Double-sided steam oxidation: 800°C/12min + 1400°C/2min
- CrN-coating 15mic:
  - Local coating failure (probably cracks)
  - ZrN area => no melting observed



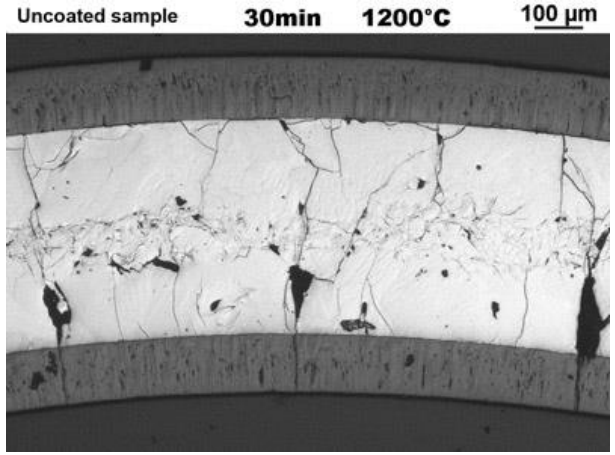
- Other concepts of coating:
  - Shift the eutectical point to higher temperatures
  - Without local failures (transformations CrN → Cr_2N)

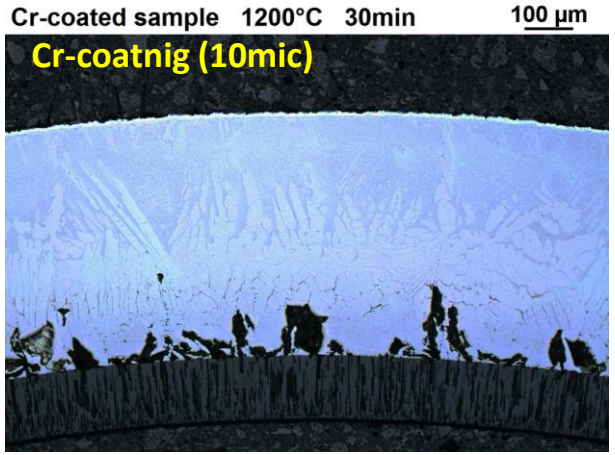
    ⇒ Substochiometric Cr_xN_y & multi-layer Cr+CrN

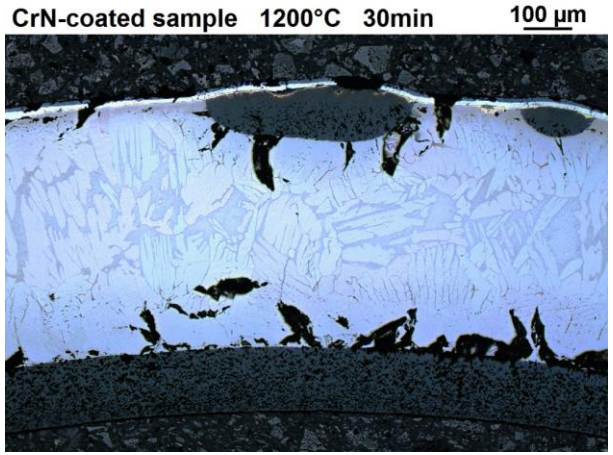


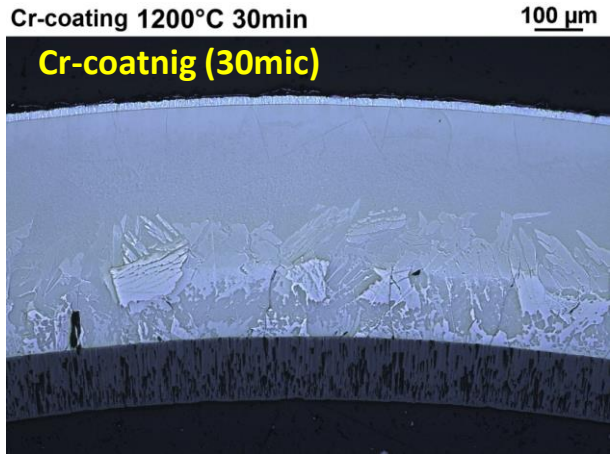
Microstructure after high temperature ox.

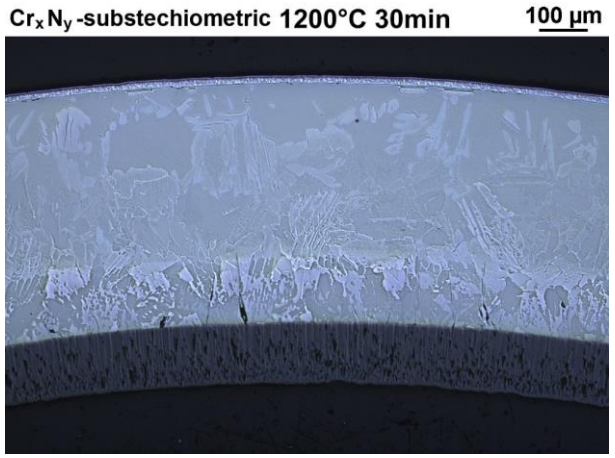
- Used variants (Cr_xN_y & Cr+CrN (~3mic)) did <u>not prevent melting</u> (=> thicker CrN)
- Double-sided oxidation: 1200°C/30min (one-sided oxidation: 21% ECR_{CP})
  - New variants without any local defect => similar to Cr-coating

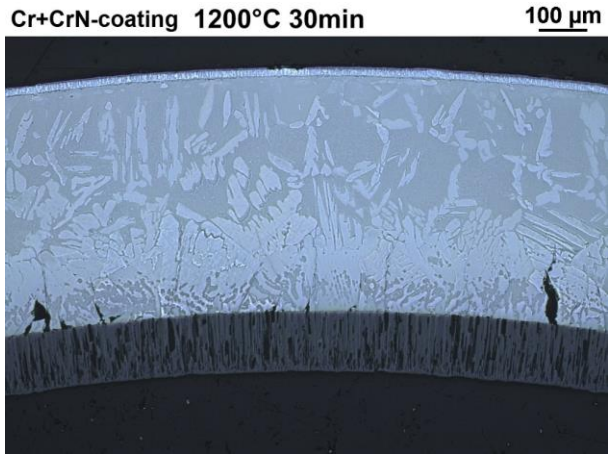












Summary

- Corrosion experiment (63d): 5x lower weight gain for Cr-coated samples in WWER conditions
- All variants of Cr-based coatings significantly lower mass gain in comparison with uncoated
- Cr-coating:
  - No local failures detected, coating very compact => Increasing of coping time
  - Different character of microstructure (HV & O-content) for coated samples => for the same weight gain ↓ ductility
  - On coating-substrate interface was identified Cr-Zr mixed area (Cr diffusion)
  - Experiments above melting point:
    - in argon: melted area found only on the coated surface
    - in steam: Cr diffusion from oxygen enriched areas through the sample wall
- CrN-coating:
  - Local coating failures at high temperatures
  - No melting observed at 1400°C
- Other Cr-based coatings: substochiometric Cr_xN_y & Cr+CrN
  - No local failures, oxidation behaviour close to Cr-coated samples
  - Melting observed for temperature 1400°C

Future work

- Evaluation of on-going corrosion experiment
- High temperature oxidation:
  - Single-sided oxidation 1000-1300°C
  - Multi-layer coating (Cr+CrN) development will be continued
- Performing burst-test experiment
- On-going cooperation:
  - X-ray diffraction of Cr & CrN coatings up to 1100°C
    (P. Šutta: New Research Technology Centre West University of Bohemia)
  - In-situ corrosion experiment using impedance spectroscopy: 280°C, 84bar, WWER chemistry
    (A. Krausová at al: University of Chemistry and Technology Prague)
  - Neutron irradiation of Cr & CrN coated samples (1dpa) + nanohardness measurements (from RT up to 750°C)
    (H. Namburi at al: Research Centre Řež)
- Participation on IAEA CRP T12030 project („Analysis of Options and Experimental Examination of Fuels for Water-Cooled Reactors with Increased Accident Tolerance“)

Thank you for your attention

Acknowledgements

The authors thanks to whole UJP-zirconium department (namely: V. Rozkošný, J. Šustr, D. Rada, P. Gajdoš, and V. Vrtílková) for sample preparation, running experiments, and post-experimental evaluation.

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Grant Agency of the Czech Technical University in Prague, grant No. SGS16/252/OHK4/3T/14 and IAEA CRP 21065,

Grants no. „TA02011025, program ALFA – TAČR“, „TH02020477, program EPSILON – TAČR“, CZ.2.16/3.1.00/21563.

High-temperature oxidation resistance and self-healing behavior of Cr_2AlC MAX phase coating on Zircaloy-4

Chongchong Tang*, Martin Steinbrueck, Mirco Grosse, Sven Ulrich, Michael Stueber, Hans Juergen Seifert

Karlsruhe Institute of Technology, IAM-AWP, POB 3640, 76021 Karlsruhe, Germany

23rd International QUENCH Workshop, October 17-19 2017, Karlsruhe, Germany

Zirconium-based alloys are currently utilized as fuel cladding and structural components in commercial light water reactors due to their low thermal neutron absorption cross section, good mechanical properties and reasonable corrosion resistance during operation conditions. One undesirable feature of zirconium-based alloy cladding is their extremely fast oxidation kinetics with high-temperature steam during loss of cooling accidents (LOCA). A considerable amount of heat and hydrogen gas is produced by the reaction of zirconium and steam. The claddings undergo severe degradation and hydrogen explosion can occur, followed by subsequent release of highly-radioactive fission products to the environment like during the nuclear accidents at the Fukushima Daiichi Nuclear Power Plant in 2011.

One strategy to improve the accident tolerance of the state-of-the-art zirconium-based alloy fuel claddings is to coat the outer surface with an oxidation resistant coating. This solution promises the elimination of corrosion degradation during normal operation, as well as significant reduced oxidation kinetics with steam during off-normal conditions. $M_{n+1}AX_n$(MAX) phases represent a family of ternary layered carbides or nitrides which possess a unique combination of the merits of both metals and ceramics. Alumina-forming MAX phase materials, like Ti_2AlC and Cr_2AlC, are being considered as protective coatings with respect to their excellent oxidation resistance up to 1400°C.

In this study, Cr_2AlC coatings have been deposited on Zircaloy-4 substrates by magnetron sputtering using elemental nano-multilayer thin films, and subsequent thermal annealing in argon. The total thickness of the coatings is around 6.5 µm and both coatings have a 500 nm Cr layer as bonding layer and diffusion barrier. One design of coatings also deposited a 1.5µm thick Cr capping layer to migrate potential fast dissolve of Al during normal operation. Crystallization of Cr_2AlC MAX phase starts from 480°C by annealing in Ar and formation of phase-pure Cr_2AlC MAX phase but with surface microcracks at 550°C is confirmed. Both coatings demonstrated high adherence, excellent oxidation resistance up to at least 1200°C and self-healing capability with growth of protective Al_2O_3 scale or of protective Al_2O_3 scale beneath Cr_2O_3 during high-temperature oxidation.


2

Karlsruhe Institute of Technology

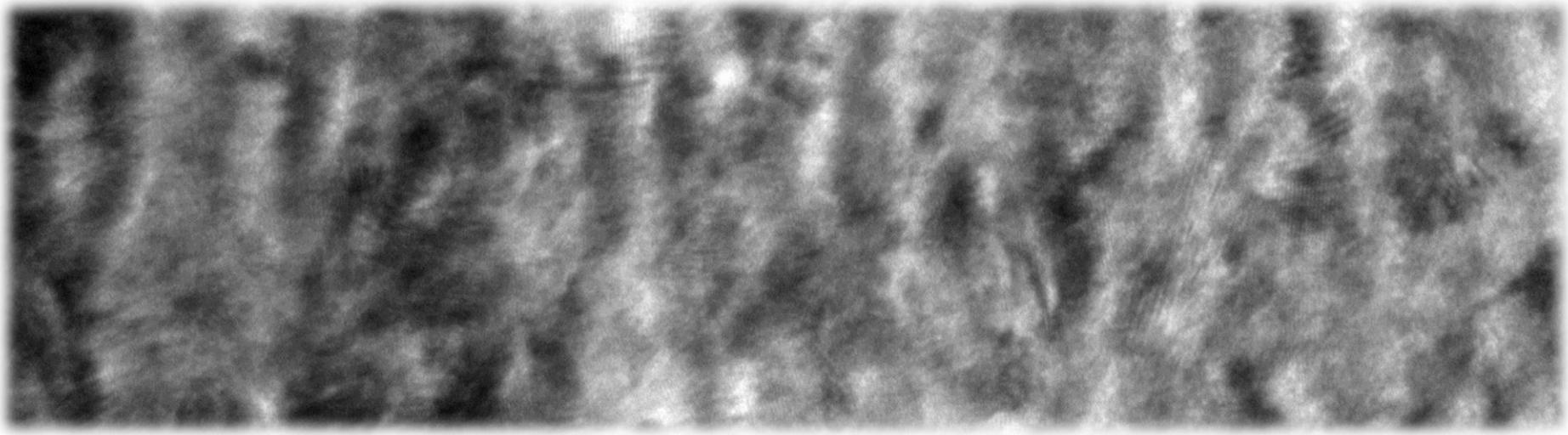
Institute for Applied Materials

High-temperature oxidation resistance and self-healing behavior of Cr_2AlC MAX phase coating on Zircaloy-4

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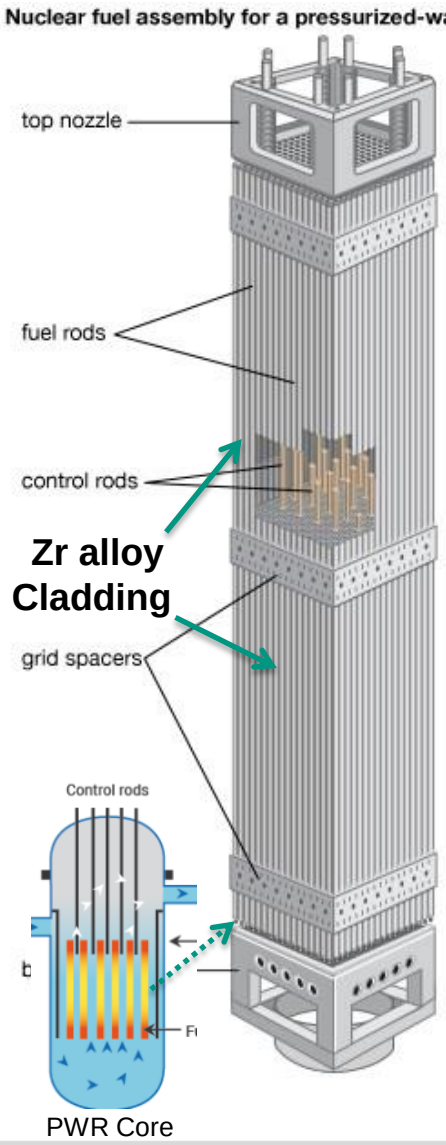
Institute for Applied Materials IAM-AWP



Outline

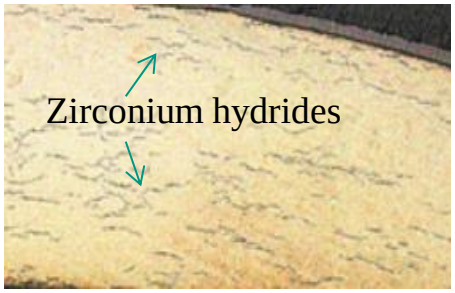
- Introduction and Motivation
- Experimental setup
- Results and discussions
  - Cr_2AlC coatings on alumina substrates
  - Cr_2AlC-based coatings on Zircaloy-4 substrates
  - High-temperature oxidation of Cr_2AlC coated Zircaloy-4
  - Thermal-shock/Quench behavior of Cr_2AlC coated Zircaloy-4
- Conclusions and outlook

Introduction



PWR Core

- Two major limitations
  - Hydride embrittlement during normal operation.
  - Degradation and fast oxidation kinetics with HT steam during accident scenarios. Strong release of hydrogen and heat!



(Allen,2012)





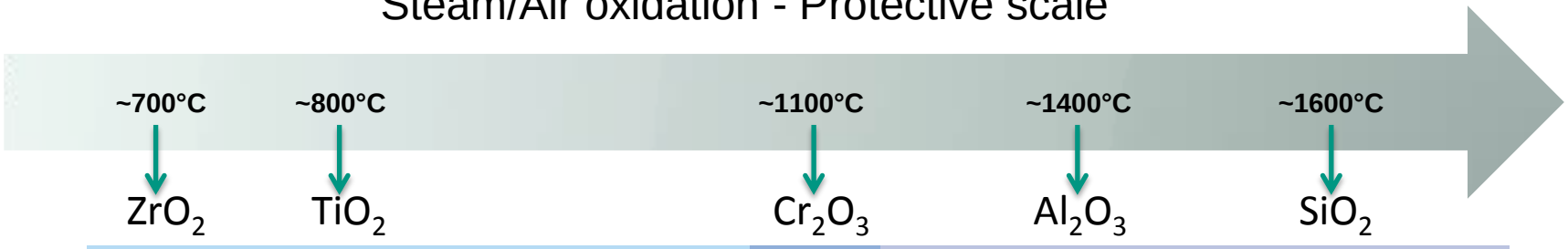
Accident Tolerant Fuels (ATF)

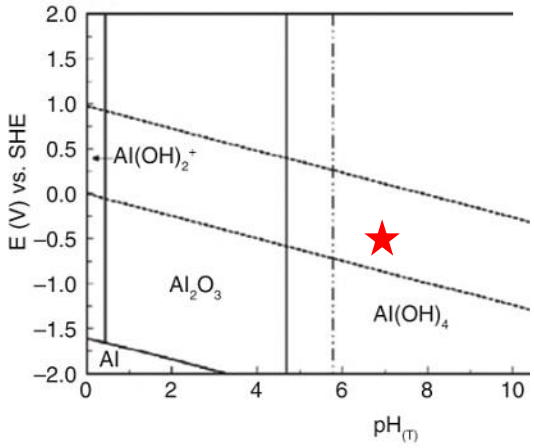
- Coated Zr-alloy cladding
- Advanced alternative claddings

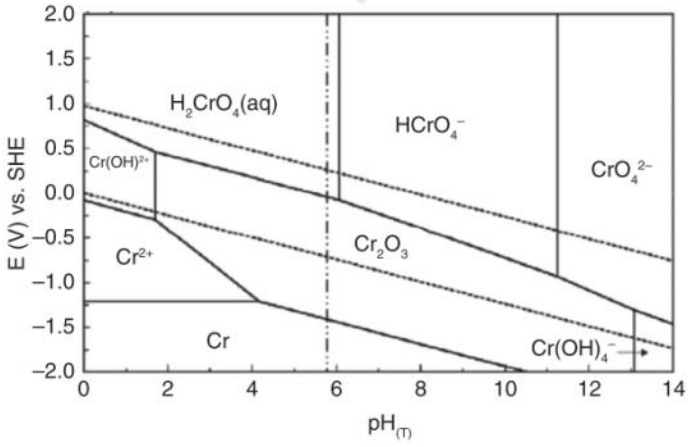
[1]T.R. Allen, R.J.M. Konings, A.T. Motta. Compr. Nucl. Mater. 5 (2012) 49–68.

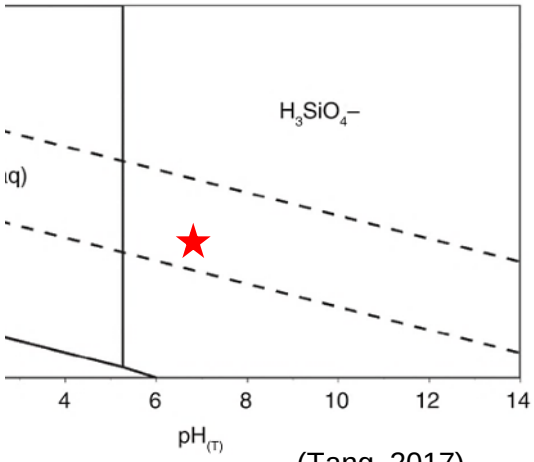
Motivation

Steam/Air oxidation - Protective scale









(Tang, 2017)

★ PWR normal operation

350°C, 25 Mpa,10^{-6} mol/kg

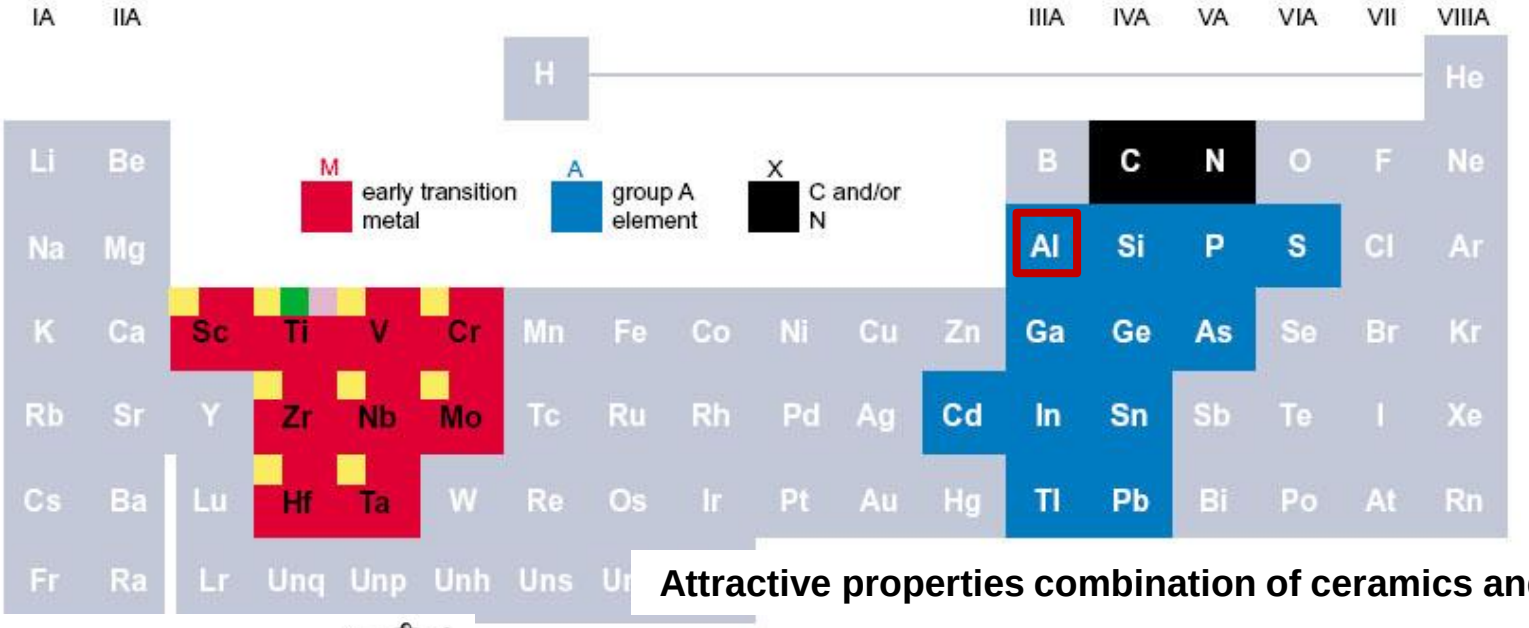
Non-stable: Al_2O_3 and SiO_2

Ternary carbide coatings (Zr or Ti or Cr - C - Al)

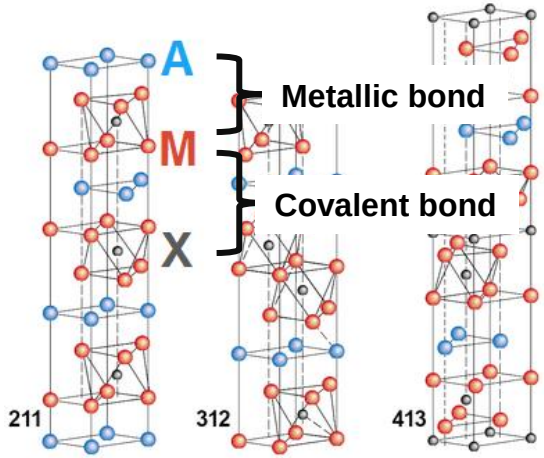
[2] C. Tang, M. Stueber, H. J. Seifert, M. Steinbrueck. *Corros. Rev.* 374–379 (2017).

Motivation

Why ternary carbides → $M_{n+1}AX_n$ phase



Attractive properties combination of ceramics and metals



- Low density
- Good thermal conductivity
- High melting/decomposition temperature
- Relative high fracture toughness
- Good radiation tolerance

Ti_2AlC, Cr_2AlC
Alumina-forming
during HT oxidation

+

Zr_2AlC

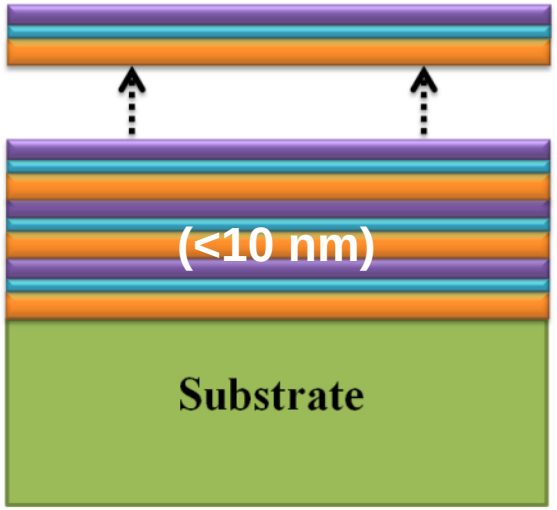
[3] M.W. Barsoum, MAX phases: properties of machinable ternary carbides and nitrides, John Wiley & Sons, 2013.

Experimental setup

Film total thickness:

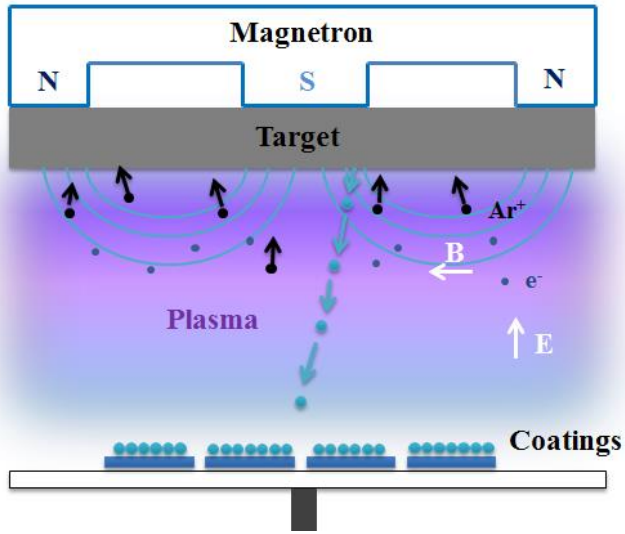
~ 3 μm on SiO_2/Si, Al_2O_3

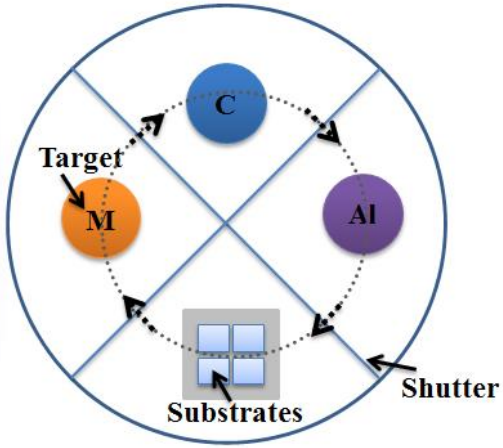
5.5-6.5 μm on Zircaloy-4



Square: 10 × 10 mm

Film design





(Leybold Z 550 coater)

Deposition-Magnetron Sputtering

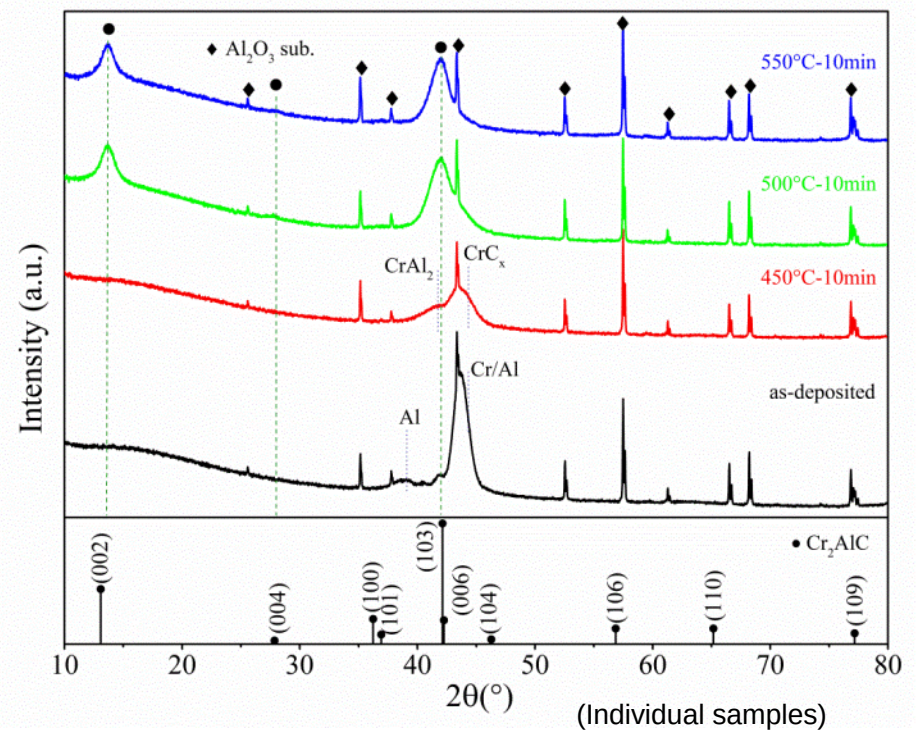
Post annealing

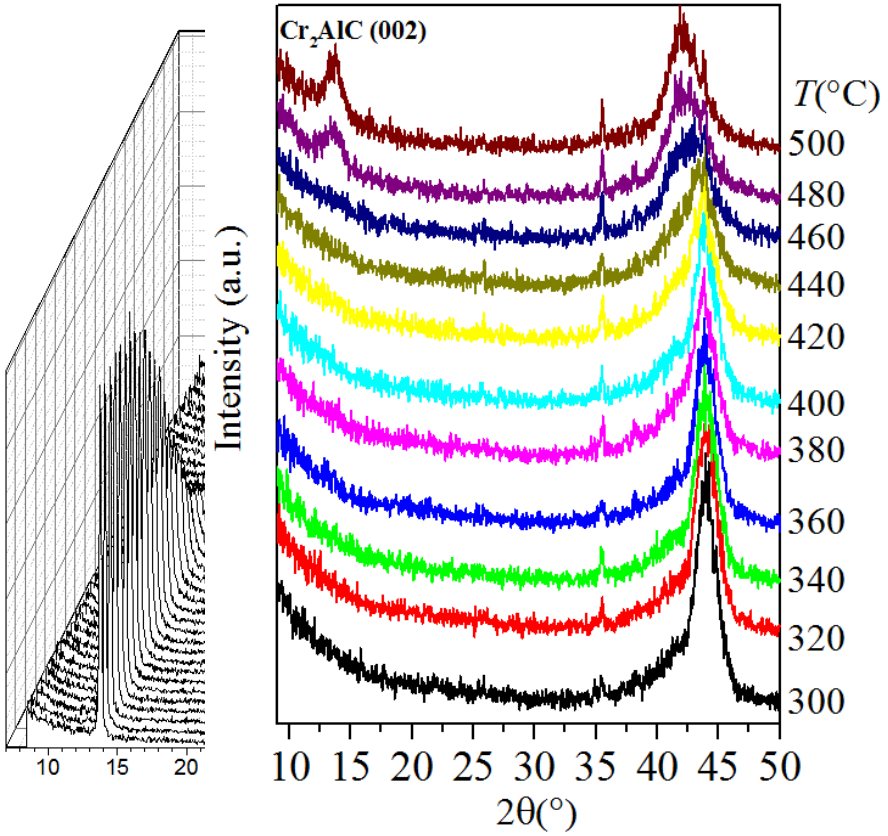
- Atmosphere: pure Ar
- Annealing temperature: 300°C-1000°C
- Holding time: 10 min

- Dense and single-phase Ti_2AlC and Cr_2AlC; formation of ZrC rather than Zr_2AlC
- Non-protective effect of Ti- and Zr- based coatings once oxidation temperatures reaching 1000°C

Cr_2AlC coatings on alumina substrates

- XRD
- *In-situ* HTXRD



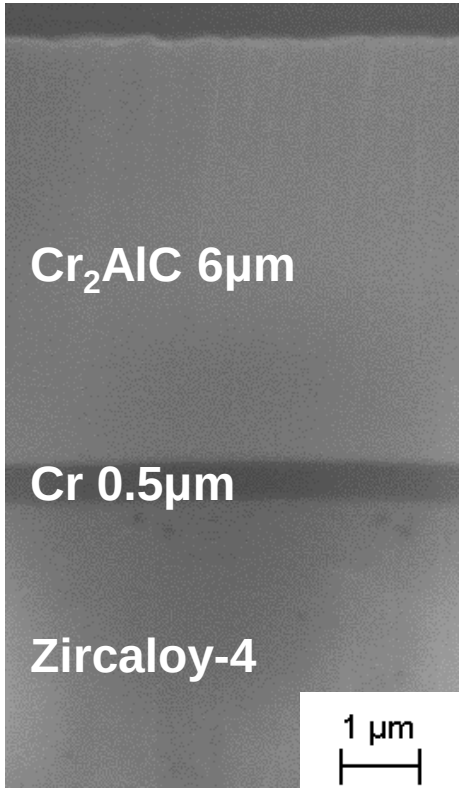


- Nanoscale elemental multilayers remain stable up to around 420°C
- Interdiffuison leads to formation of Cr-C and Cr-Al starting from 440°C
- Crystallization of Cr_2AlC MAX phase from 480°C

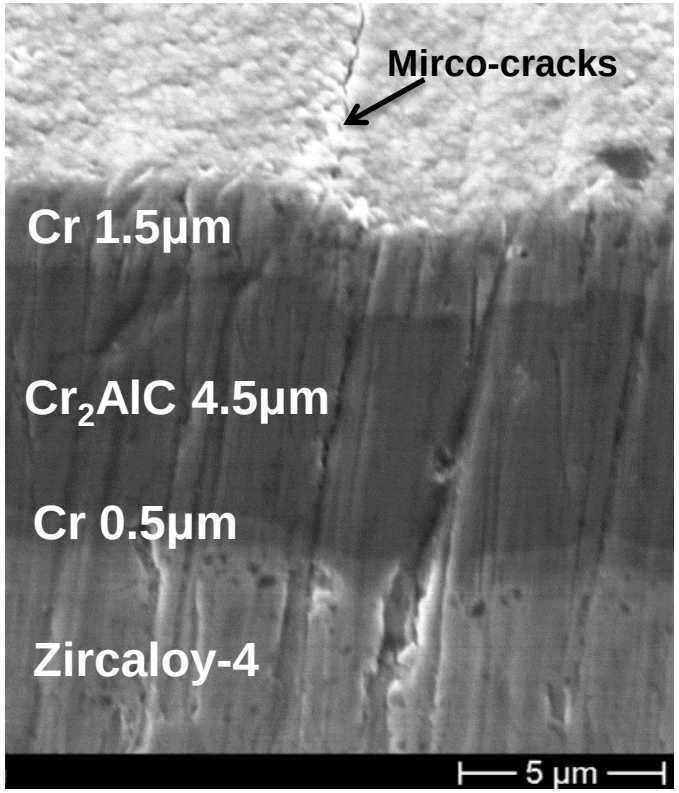
Cr_2AlC-based coatings on Zircaloy-4 substrates

- Annealed coatings- 550°C 10min

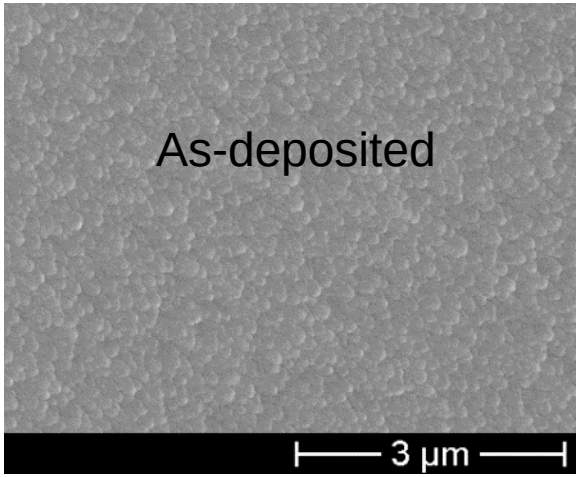
Two designs

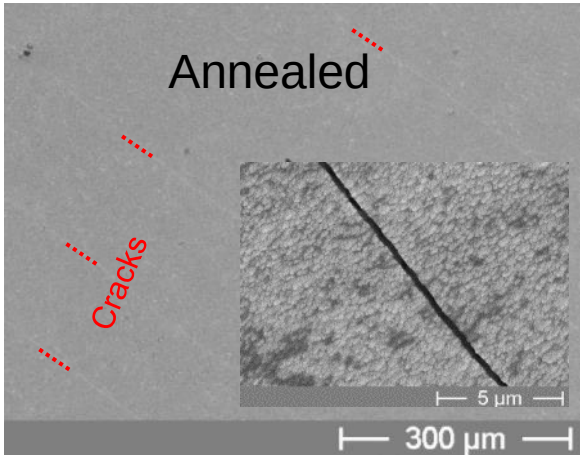


(FIB)



(Cutting)

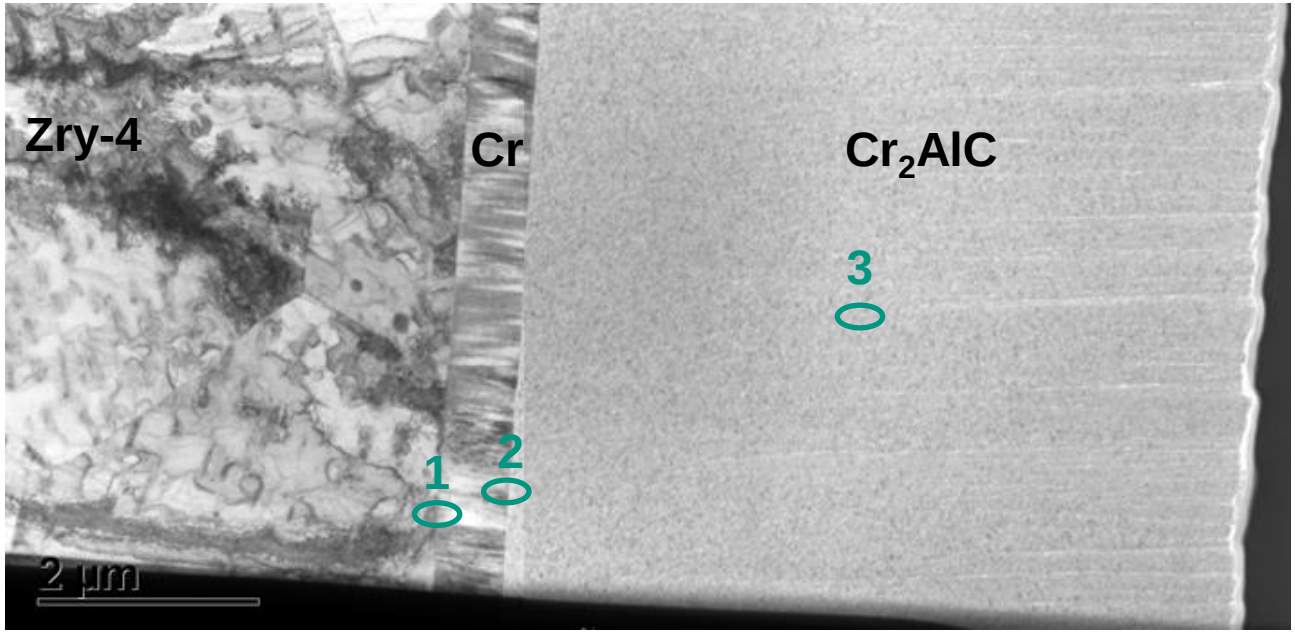


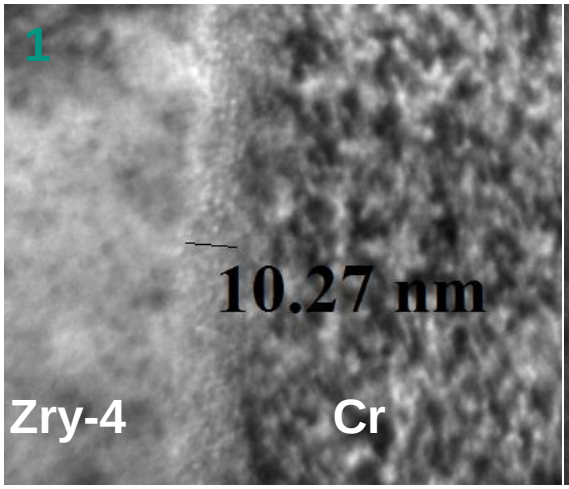


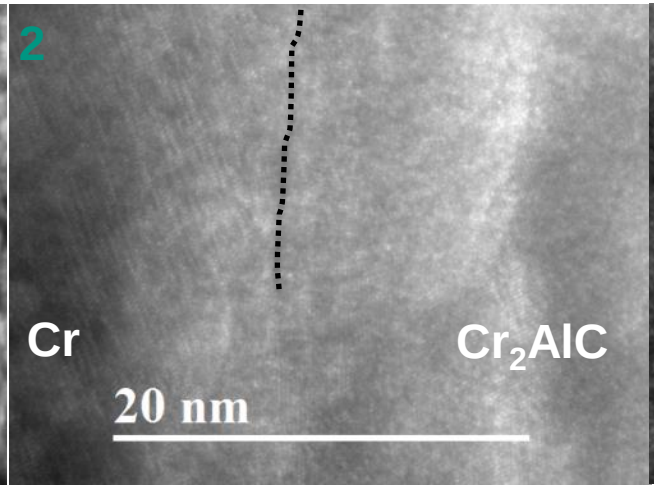
- Micro-cracks were observed on both coatings after annealing

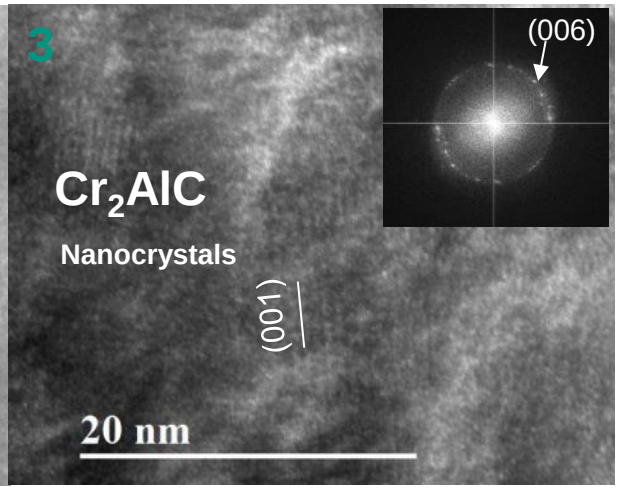
Cr_2AlC-based coatings on Zircaloy-4 substrates

- Annealed coatings- 550°C 10min







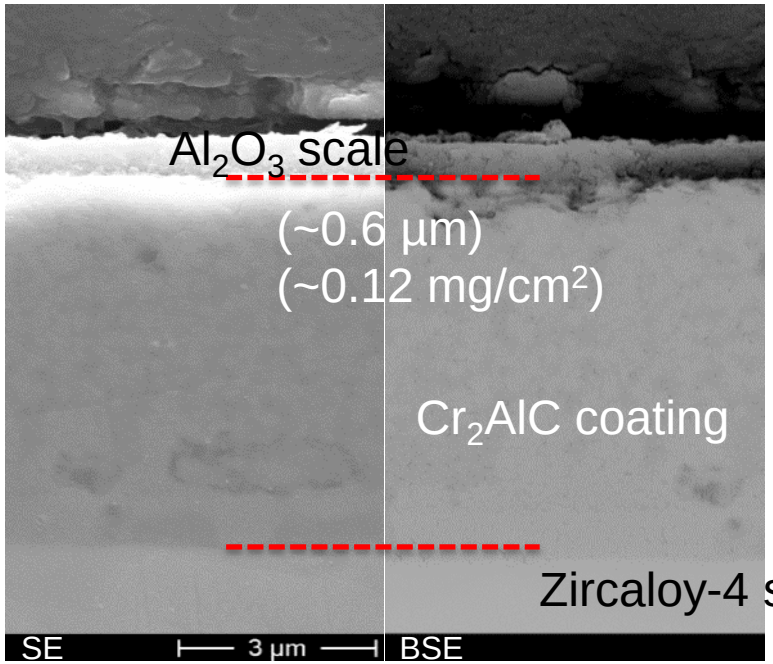


High-temperature oxidation of Cr_2AlC coated Zircaloy-4

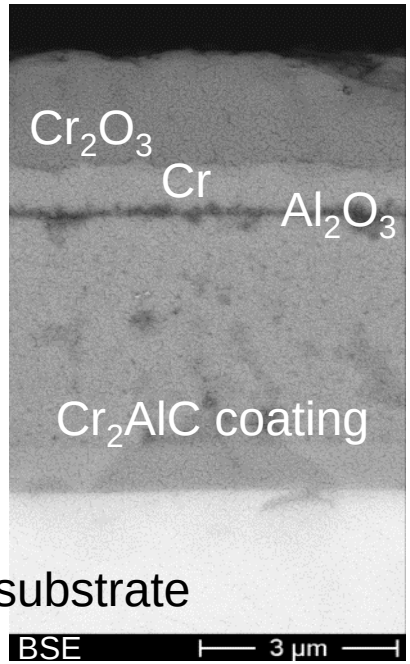
- **Isothermal Oxidation**

550°C Annealed coatings-Isothermal 1000°C 60min Steam

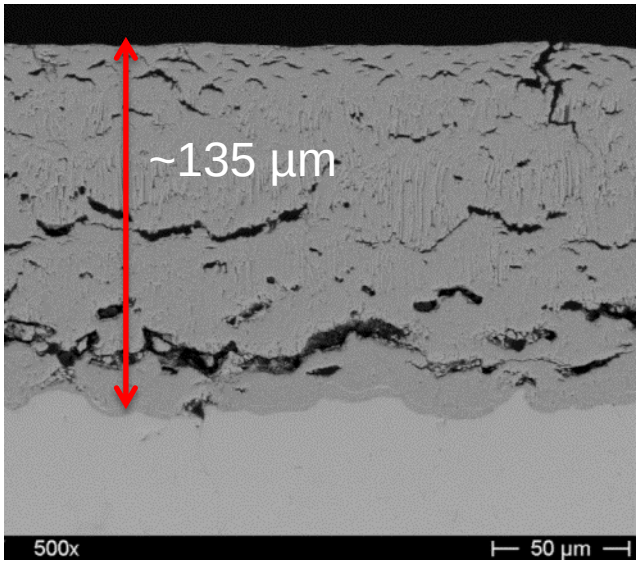
Final mass gain: Cr_2AlC /Cr coated Zircaloy-4: 3.29 mg/cm^2
Cr/ Cr_2AlC /Cr coated Zircaloy-4: 3.77 mg/cm^2
Uncoated Zircaloy-4: 25.06 mg/cm^2



Cr_2AlC /Cr coating



Cr/ Cr_2AlC /Cr coating

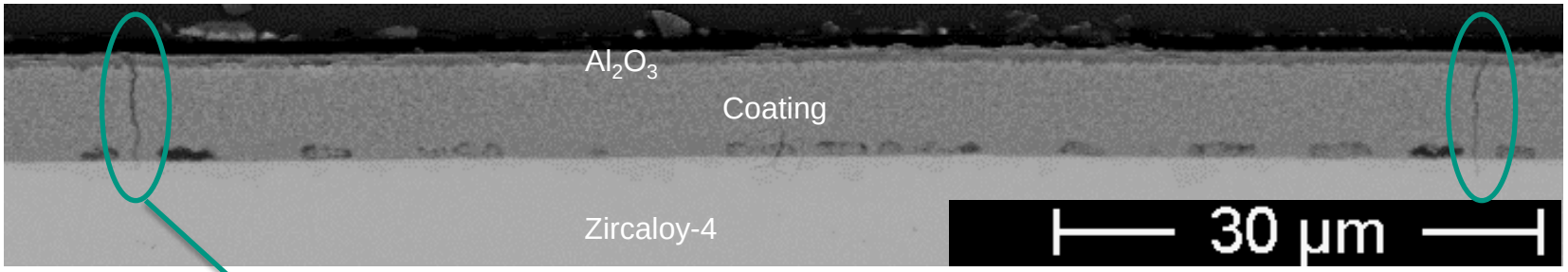


Uncoated Zircaloy-4

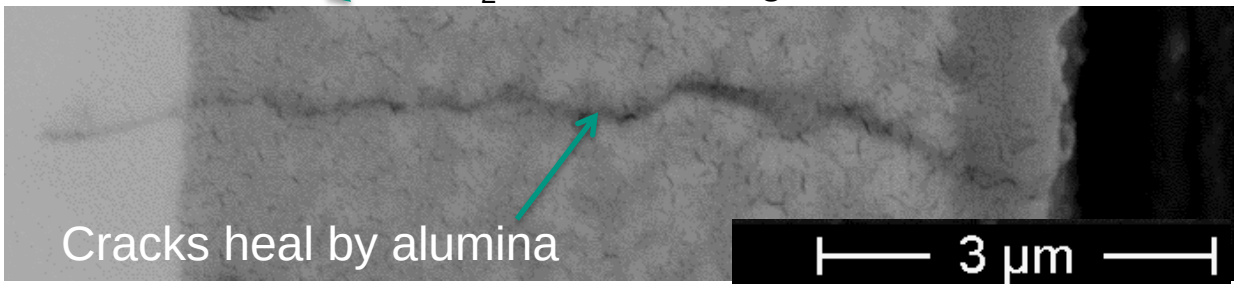
High-temperature oxidation of Cr_2AlC coated Zircaloy-4

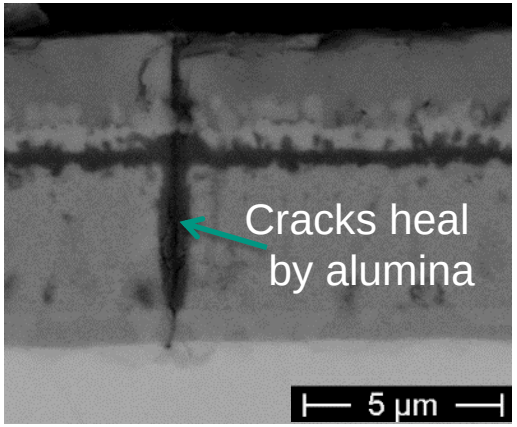
- **Isothermal Oxidation**

  550°C Annealed coatings-Isothermal 1000°C 60min Steam



Cr_2AlC /Cr coating





Cr/ Cr_2AlC /Cr coating

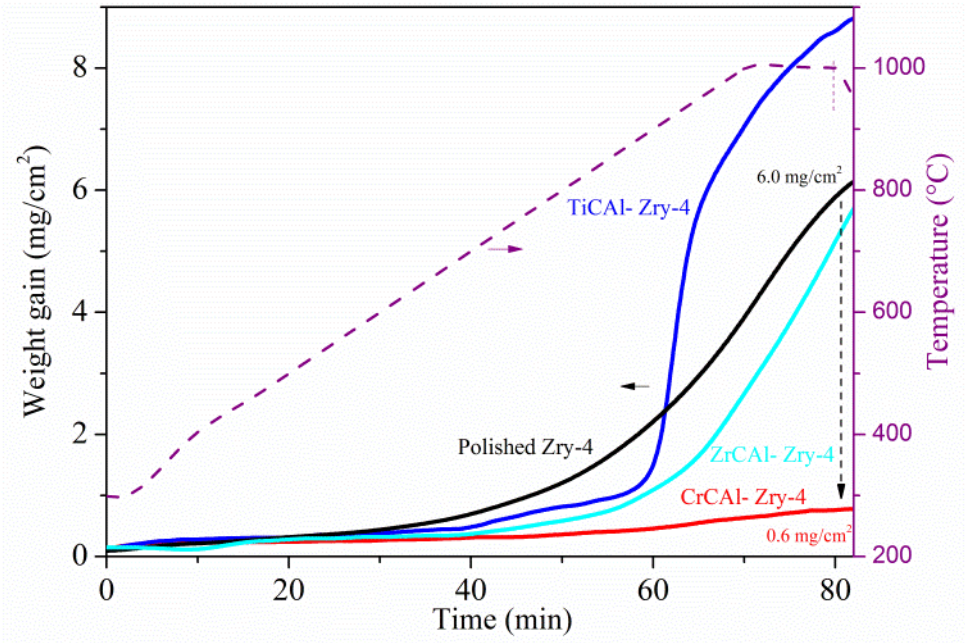
- High adherence of the coatings without delamination or spallation
- Voids observed at the coating/substrate interface after oxidation

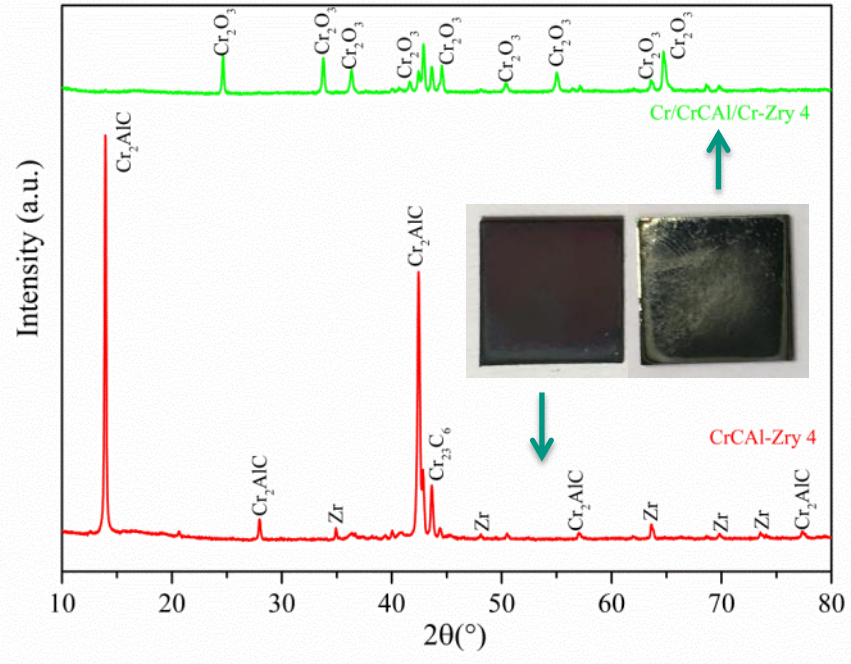
High-temperature oxidation of Cr_2AlC coated Zircaloy-4

- **Transient Oxidation**

As-deposited samples without additional annealing

300-1000°C 10K/min + 1000°C 10min, Steam





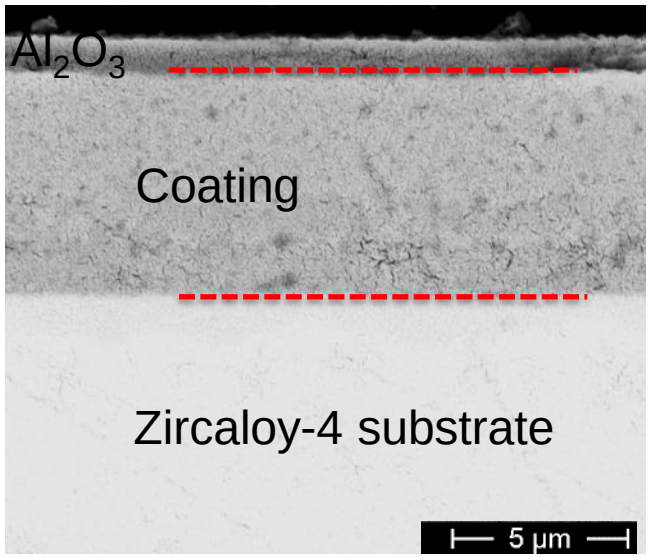
- Elemental mutilayers transform to Cr_2AlC MAX phase before manifest oxidation during ramp up heated

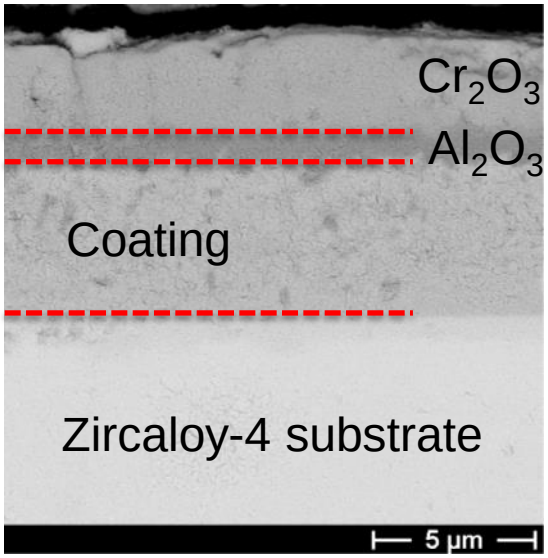
High-temperature oxidation of Cr_2AlC coated Zircaloy-4

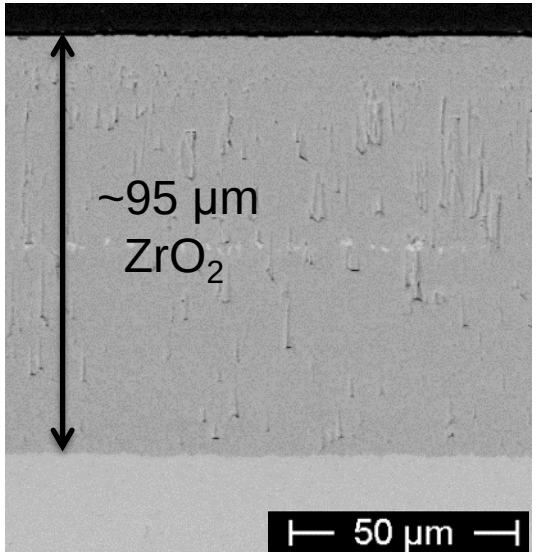
- **Transient Oxidation**

As-deposited samples without additional annealing

300-1200°C 10K/min + 1200°C 10min, Steam



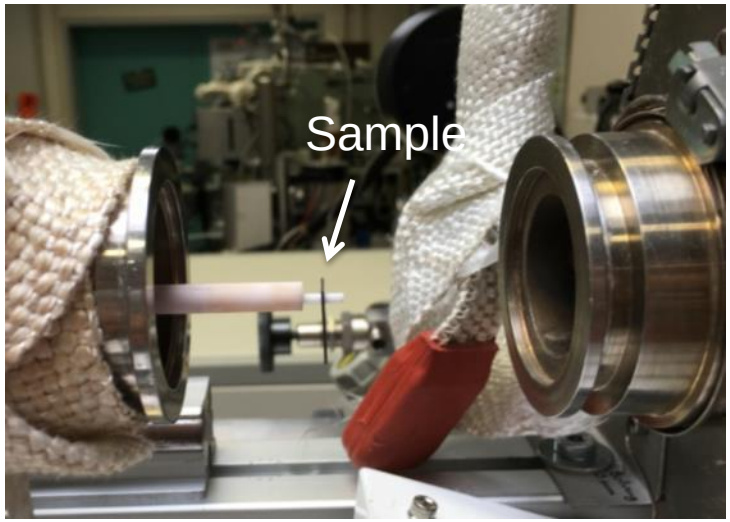




CrCAl/Cr coating | Cr/CrCAl/Cr coating | Uncoated Zircaloy-4

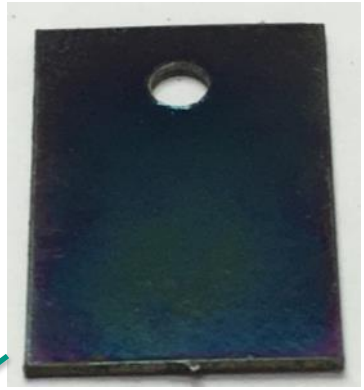
- No cracks oberved on the surface after oxidation
- Growth of dense Al_2O_3 scale on CrCAl coatings during HT oxidation
- Growth of dense, protective Al_2O_3 scale beneath Cr_2O_3 on Cr/CrCAl coatings

Thermal-shock/Quench behavior of Cr_2AlC coated Zircaloy-4



(15×10 mm)

Oxidation at 1000°C for 30 min in steam
→ Quickly pull out from the furnace
→ Drop into ambient water

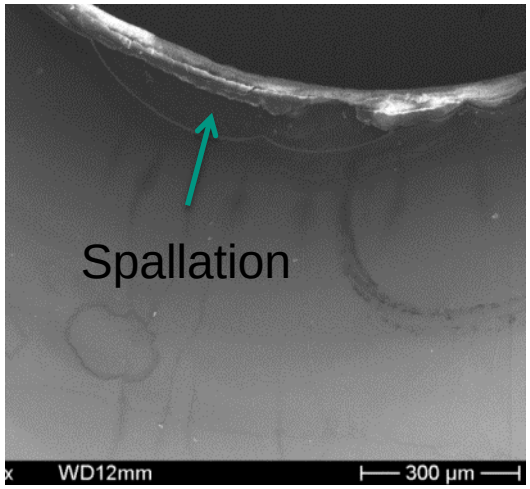


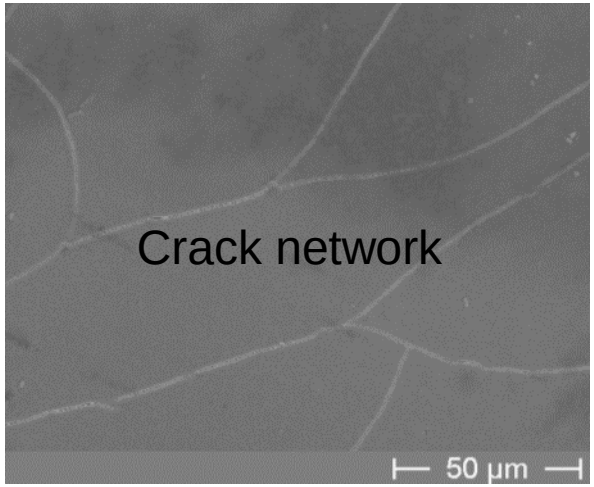


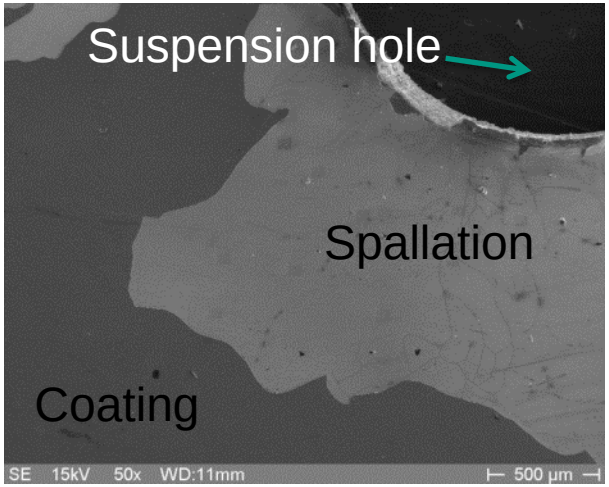
High adherence

CrCAl/Cr coating

Cr/CrCAl/Cr coating





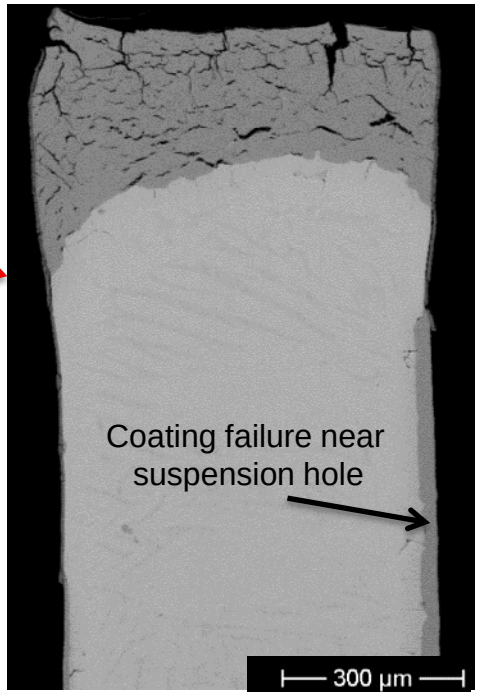


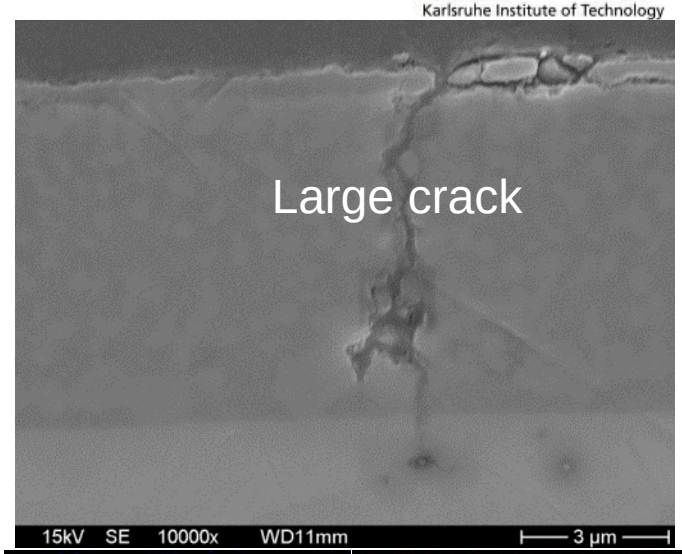
Thermal-shock/Quench behavior of Cr_2AlC coated Zircaloy-4

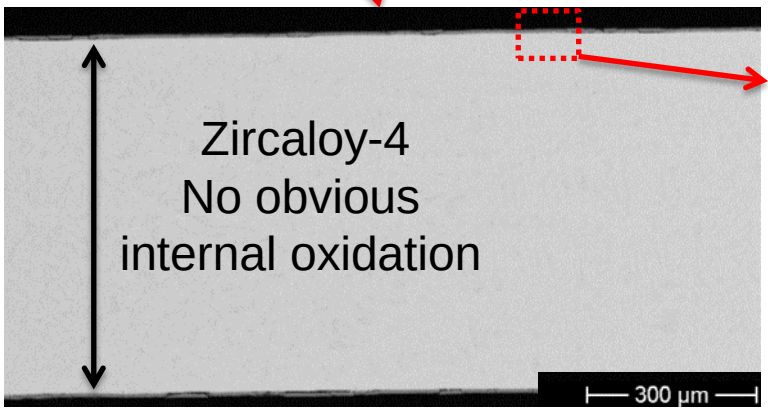
CrCAl/Cr coated sample

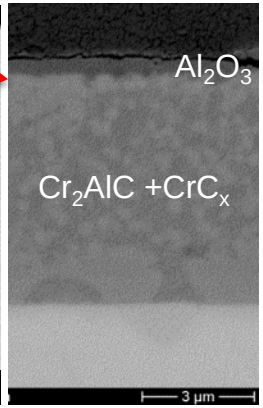
Quench +
1000°C 60min Steam

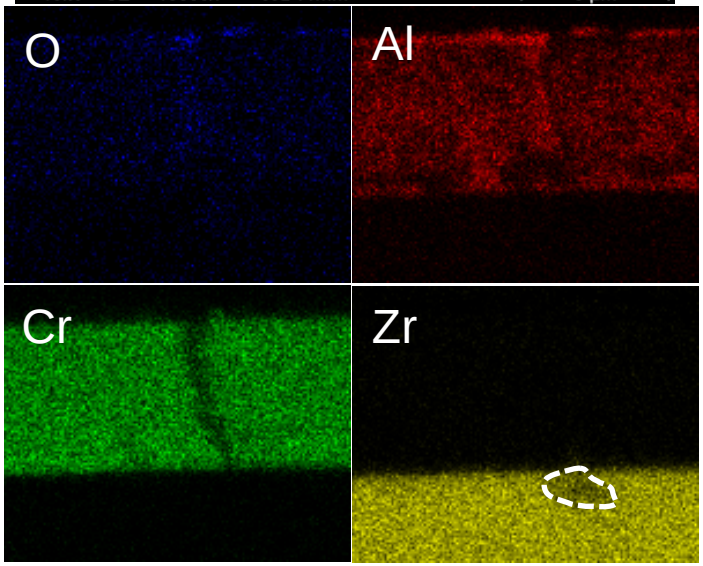












Conclusions and outlook

- Crystallization of Cr_2AlC MAX phase starts from 480°C by annealing of magnetron sputtered nanoscale elemental multilayer stacks in Ar.

- Multilayer Cr/C/Al coatings without or with a Cr capping layer were synthesized on Zircaloy-4 substrate by normal temperature deposition. Annealing of the coatings at 550°C resulted in formation of Cr_2AlC MAX phase but with surface microcracks.

- Both coatings demonstrated high adherence, excellent oxidation resisitance up to at least 1200°C and self-healing capability with growth of protective Al_2O_3 scale or of protective Al_2O_3 scale beneath Cr_2O_3 during HT oxidation.

- Utilization of as-deposited multilayer coatings without subsequent thermal annealing provides one solution to avoid cracks while maintains the excellent oxidation resisitance.

  - Autoclave corrosion (PWR normal operation) in cooperation with Westinghouse, USA
  - High-temperature, long exposure oxidation test in steam

Many thanks to:

Mrs. U. Stegmaier, Mrs. P. Severloh, Mr. J. Glaser
Dr. H. Leiste, Mr. S. Zils, Dr. H. Geßwein
Dr. M. Klimenkov, Mrs. U. Jäntsch, Mr. T. Weingärtner, other colleagues
Mrs. X. Yu
China Scholarship Council (CSC)

Thanks for your attention !

A New Approach to Enhance the Oxidation Resistance of a Zirconium Alloy: High-Temperature Pre-Oxidation

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[a]*Ulsan National Institute of Science and Technology, 50 UNIST-gil, Eonyang-eup, Ulju-gun, Ulsan, 689-798 Korea*
[*]*Corresponding author: dssohn@unist.ac.kr*

Zirconium alloys have been used as fuel cladding in light water reactors owing to their low thermal neutron absorption cross section, good mechanical strength, and acceptable corrosion resistance. However, after the Fukushima accident, concerns arose over their integrity during and after the accident, because the zirconium alloys are known to have poor oxidation resistance during a loss-of-coolant accident condition. To solve this problem, many types of research have been performed, and these efforts have been incorporated into the development of the accident tolerant fuels (ATF). One of the possible candidates of ATF is the high-temperature pre-oxidized zirconium alloy. So far, most of the pre-oxidation studies have focused on the pre-oxide formed at the normal operating condition, and slight enhancement of the oxidation resistance have been reported. During our previous study, it was found that a zirconium alloy pre-oxidized at the high temperature (1200 ^{o}C) has the much more significant effect to enhance the oxidation resistance. However, the physical explanation of this phenomenon is still not enough, and it seems that more experiments and analysis need to be performed. Recently, more pre-oxidation conditions were tested, additional microstructure analysis using TEM were conducted, and these recent progresses will be presented in this study.



A New Method to Enhance the Oxidation Resistance of a Zirconium Alloy Cladding: High-Temperature Pre-Oxidation

ULSAN NATIONAL INSTITUTE OF SCIENCE AND TECHNOLOGY

Department of Nuclear Engineering
Cheol Min Lee, Dong-Seong Sohn*

23rd International QUENCH Workshop

17-19 October 2017, KIT, Karlsruhe, Germany

Outlines

01. Introduction

02. Experimental Methods

03. Results

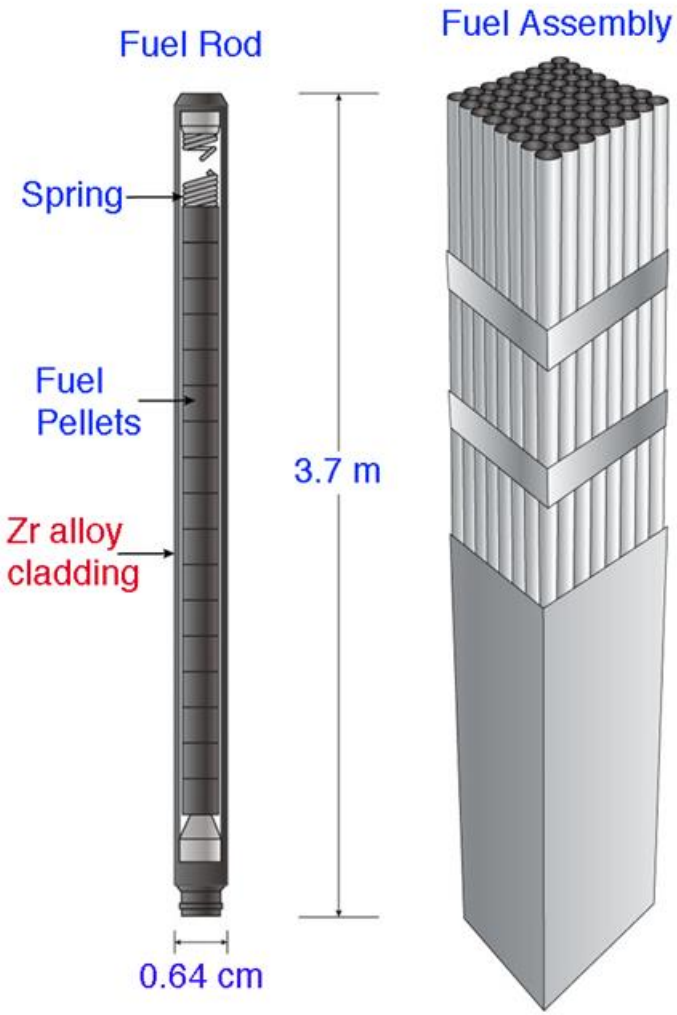
04. Summary

1. Introduction

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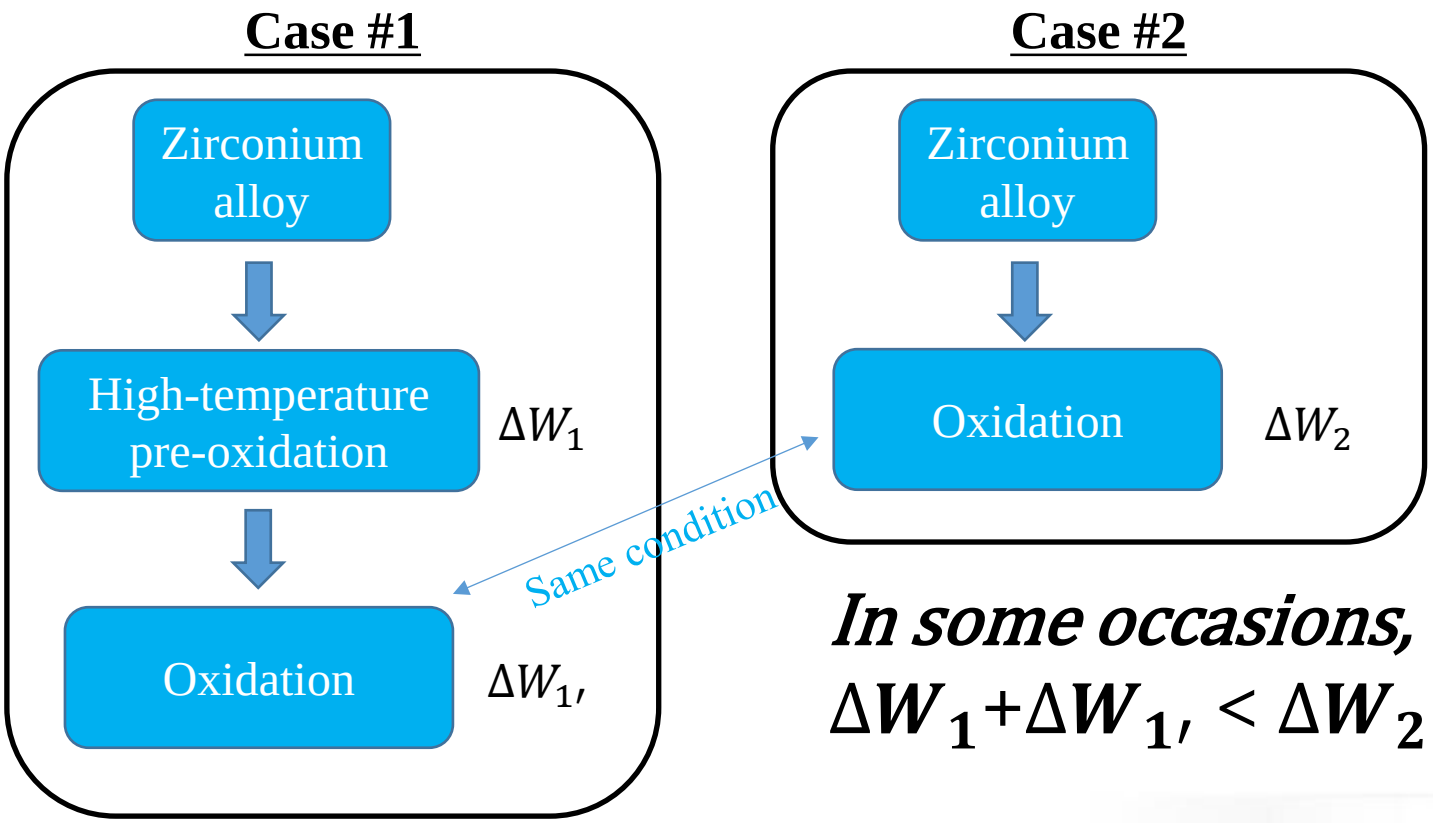
- **Zirconium Alloy**

  - After the Fukushima accident, the accident resistance of a zirconium alloy has been arising as a problem.
  - The zirconium alloys show relatively high oxidation rate at high temperatures above 1000 ^{o}C.
  - One of the solutions is to develop accident tolerant fuels (ATF).
  - Zirconium alloy with high-temperature pre-oxide can also be applied as the ATF.



High-Temperature Pre-Oxidation

- During our research, it was found that the oxidation resistance of a zirconium alloy can be enhanced by the **high-temperature pre-oxide**.



In some occasions,

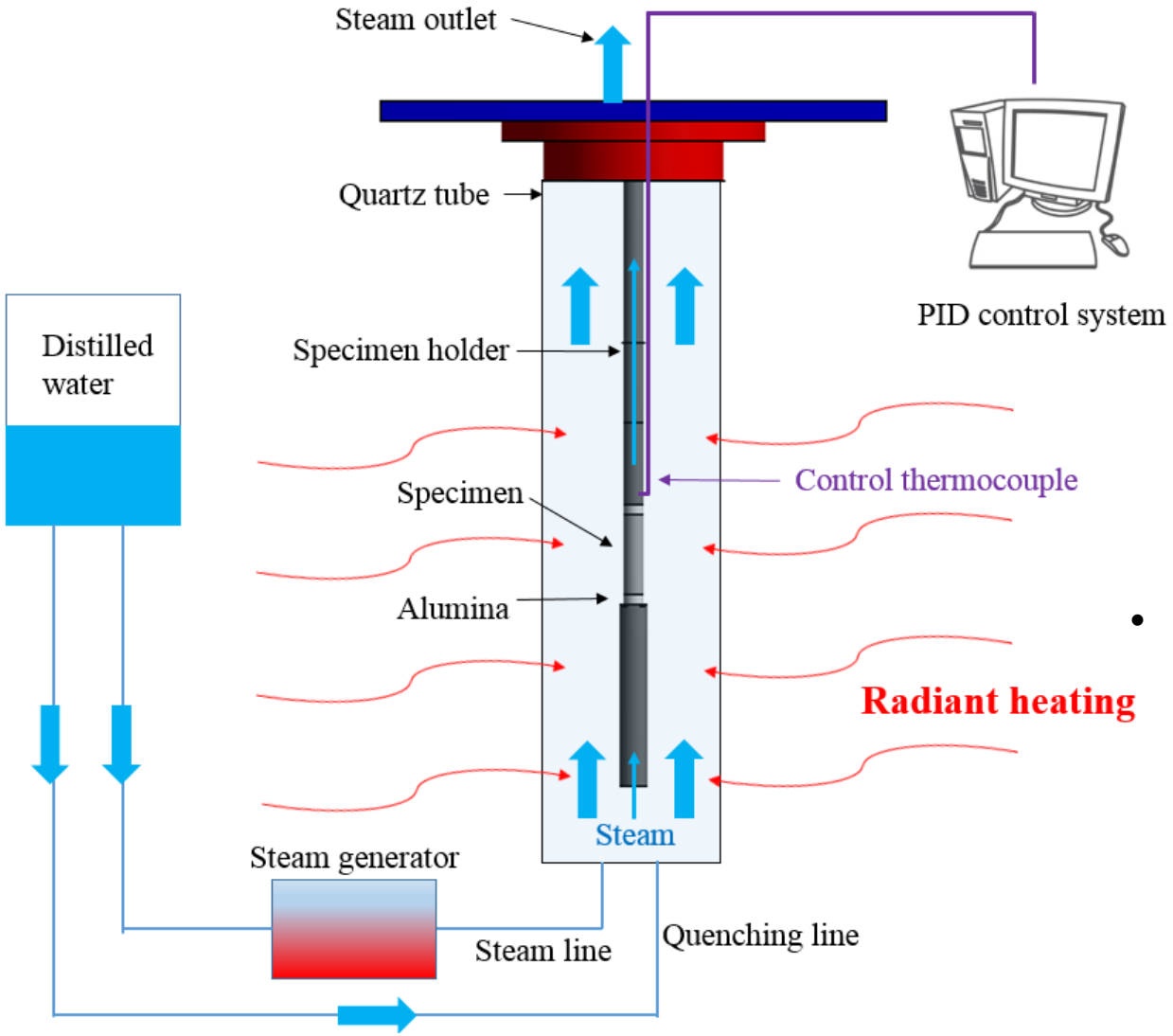
$$\Delta W_1 + \Delta W_{1'} < \Delta W_2$$

2. Experimental Methods

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2. Experimental Methods

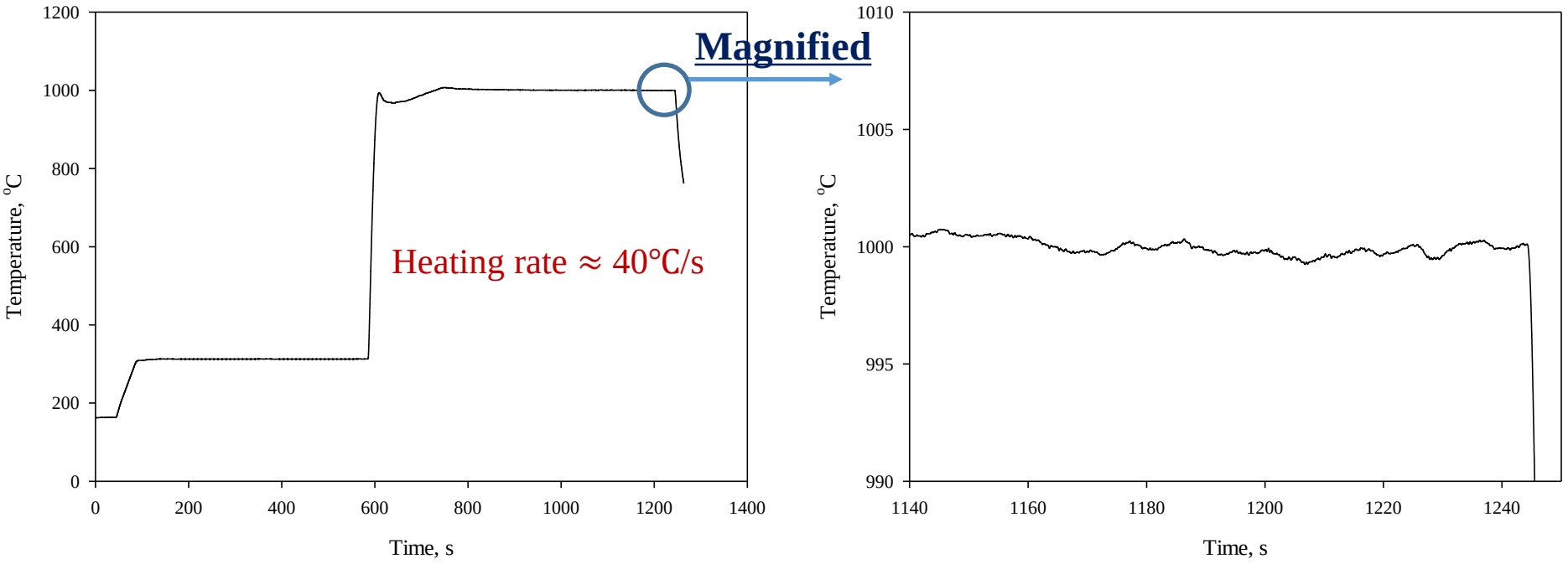
▪ Experimental System



- **Our system satisfied all the recommended conditions suggested by NRC.**
  *Ref : US NRC, Regulatory Guide 1.222

→Temperature distribution, temperature control, steam flow rate, and so on.

- **Temperature Control**



- The temperature was measured on the surface of the specimen.
- The temperature of the specimen during the isothermal oxidation was maintained within ±1℃.

2. Experimental Methods

- **Experiment**
  - **Material**
    : Zr-1Nb-1Sn-0.1Fe cladding tube
  - **Test matrix**

1. Pre-oxidation		**2. Cooling**	**3. Oxidation**	
Temperature, ^{o}C	Time, s	Air cooling to the room temperature	Temperature, ^{o}C	Time, s
1100	265		1000	5130
1200	98		1100	600, 1060, 1650, 2380, 3250
			1200	220, 392, 615

3. Results

3-1. Pre-Oxidation

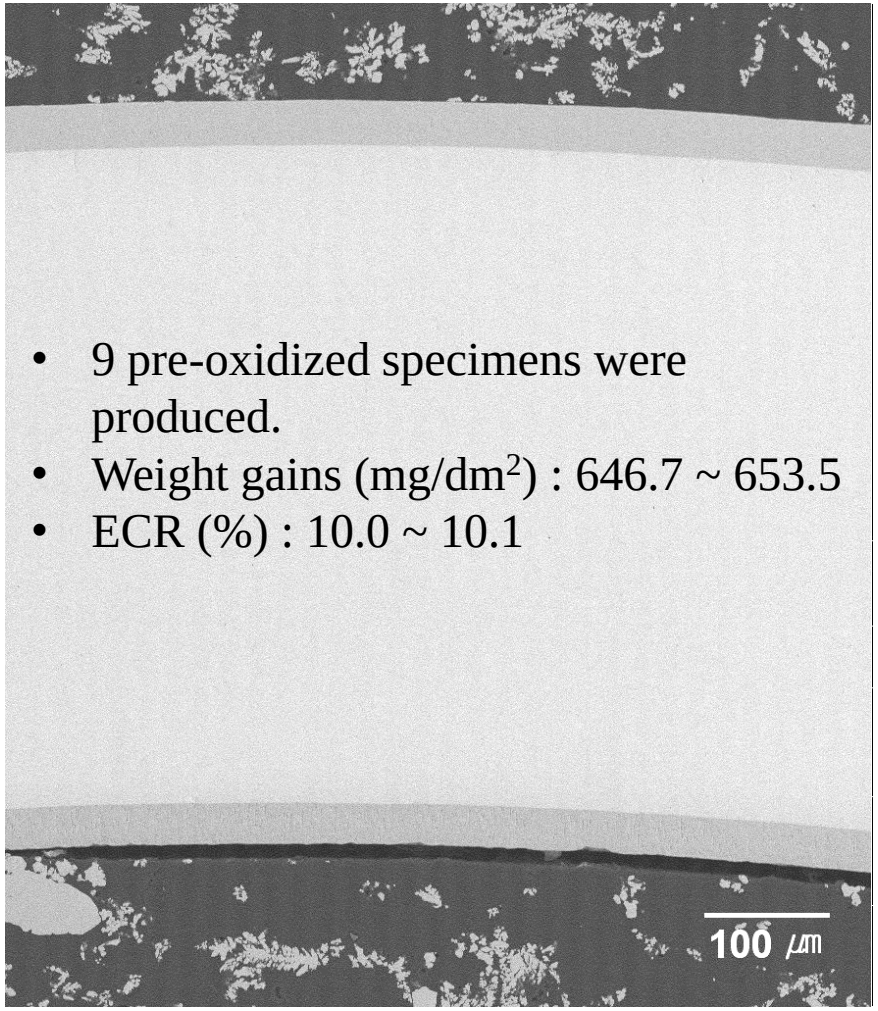
3-2. Subsequent Oxidation

3-3. TEM Analysis

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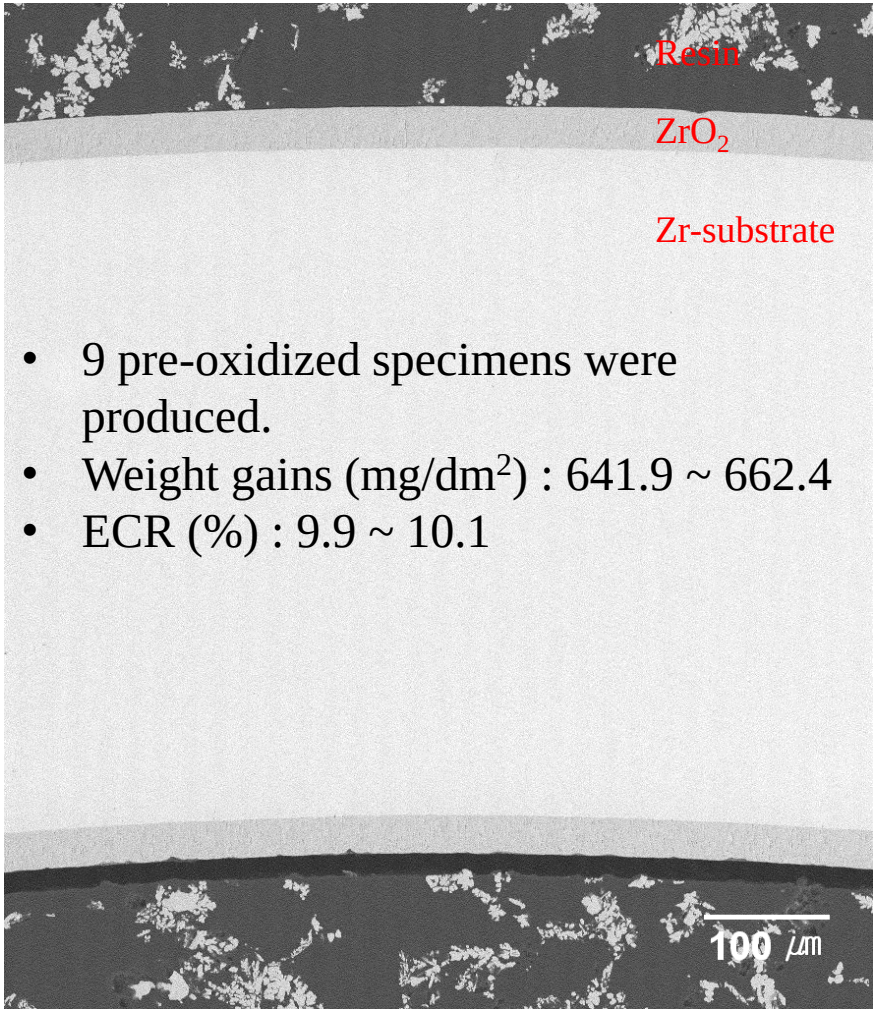
3-1. Pre-Oxidation

- **SEM BSE Image (Pre-oxidation at 1100 °C 265 s)**



- 9 pre-oxidized specimens were produced.
- Weight gains (mg/dm^2) : 646.7 ~ 653.5
- ECR (%) : 10.0 ~ 10.1

- **SEM BSE Image (Pre-oxidation at 1200 °C 98 s)**



- 9 pre-oxidized specimens were produced.
- Weight gains (mg/dm^2) : 641.9 ~ 662.4
- ECR (%) : 9.9 ~ 10.1

→Uniform pre-oxides with no cracks were formed

*BSE : Back scattered electron

3-2. Subsequent Oxidation at 1000 °C

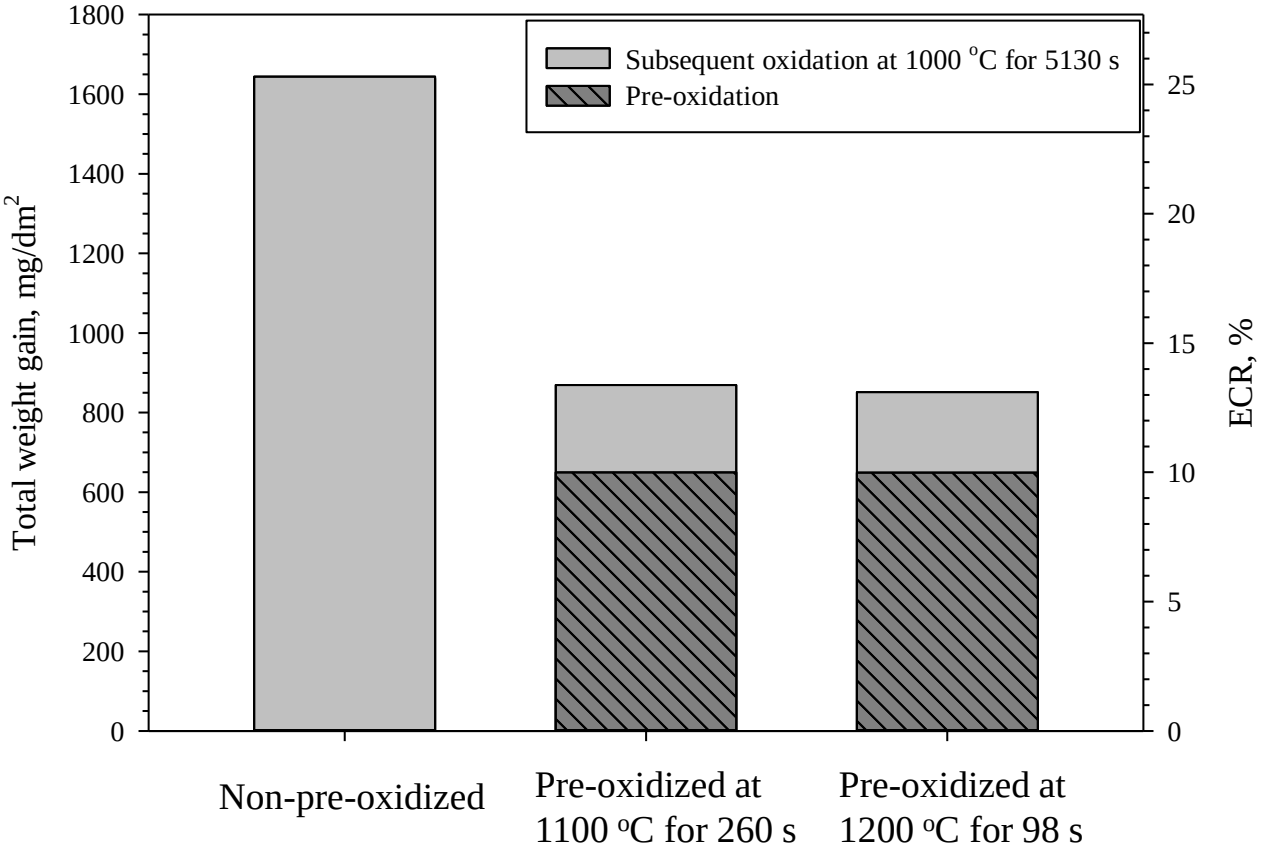
▪ Breakaway Oxidation

Pre-oxidation	no			1100 °C 260 s	1200 °C 98 s
Subsequent oxidation	1000 °C 1630 s	1000 °C 2630 s	1000 °C 5130 s	1000 °C 5130 s	1000 °C 5130 s
Surface appearance					

→Breakaway oxidation causes a zirconium alloy to become rapidly brittle.
→Severe breakaway oxidation occurred on the non-pre-oxidized specimen.
→The pre-oxidized specimens show better breakaway oxidation resistance.

3-2. Subsequent Oxidation at 1000 °C

- **Weight Gain**

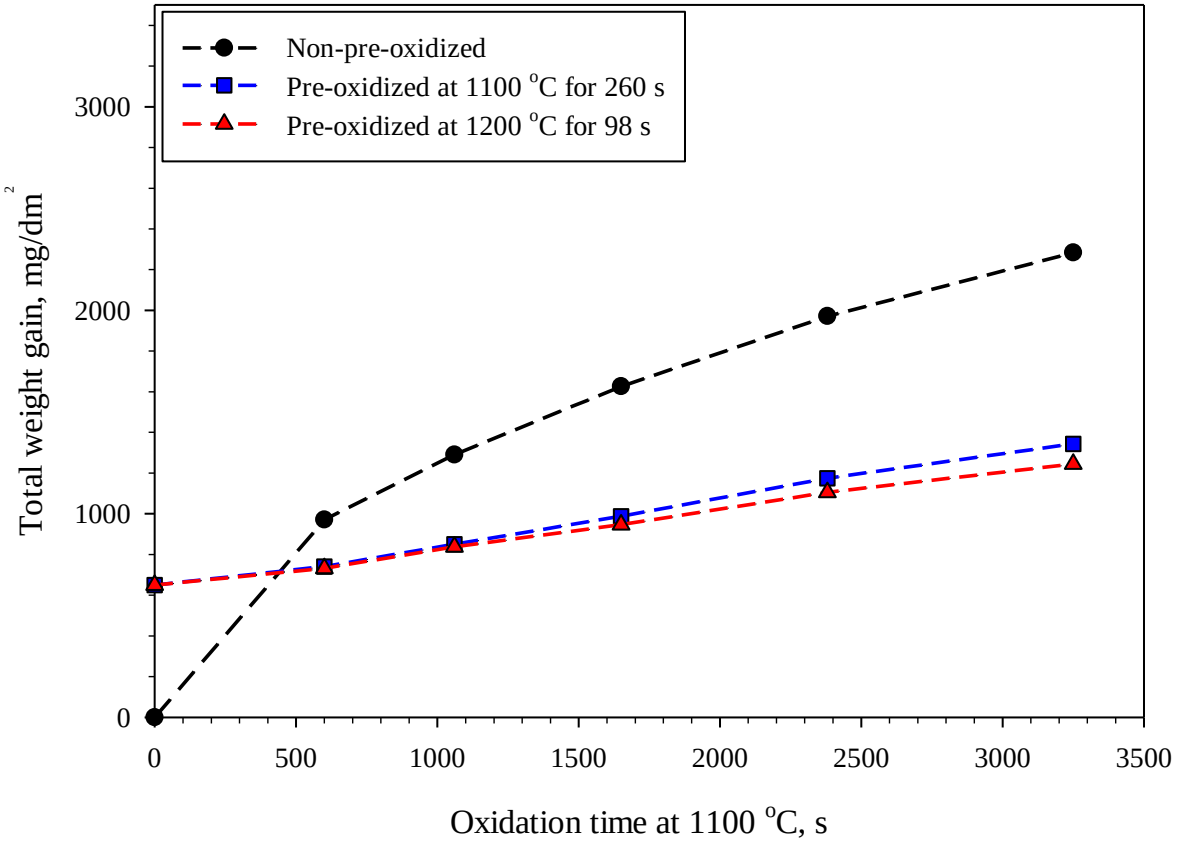


→Total weight gain = $\Delta W_{pre-oxidation} + \Delta W_{subsequent\ oxidation}$

→Weight gains of the pre-oxidized specimens are lower than the non-pre-oxidized specimens at 1000 °C.

3-2. Subsequent Oxidation at 1100 °C

- **Weight Gain**

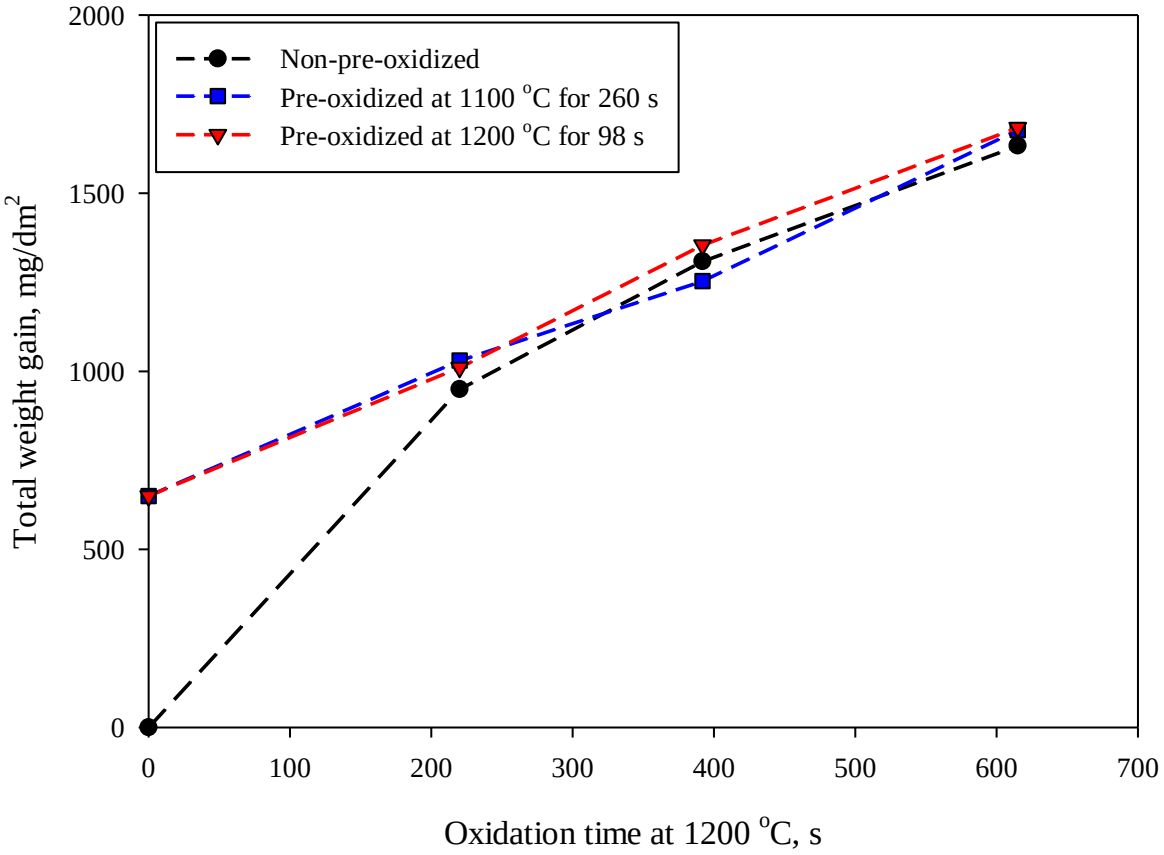


→Total weight gain = $\Delta W_{pre-oxidation} + \Delta W_{subsequent\ oxidation}$

→Weight gains of the pre-oxidized specimens are lower than the non-pre-oxidized specimens at 1100 °C.

3-2. Subsequent Oxidation at 1200 °C

- **Weight Gain**

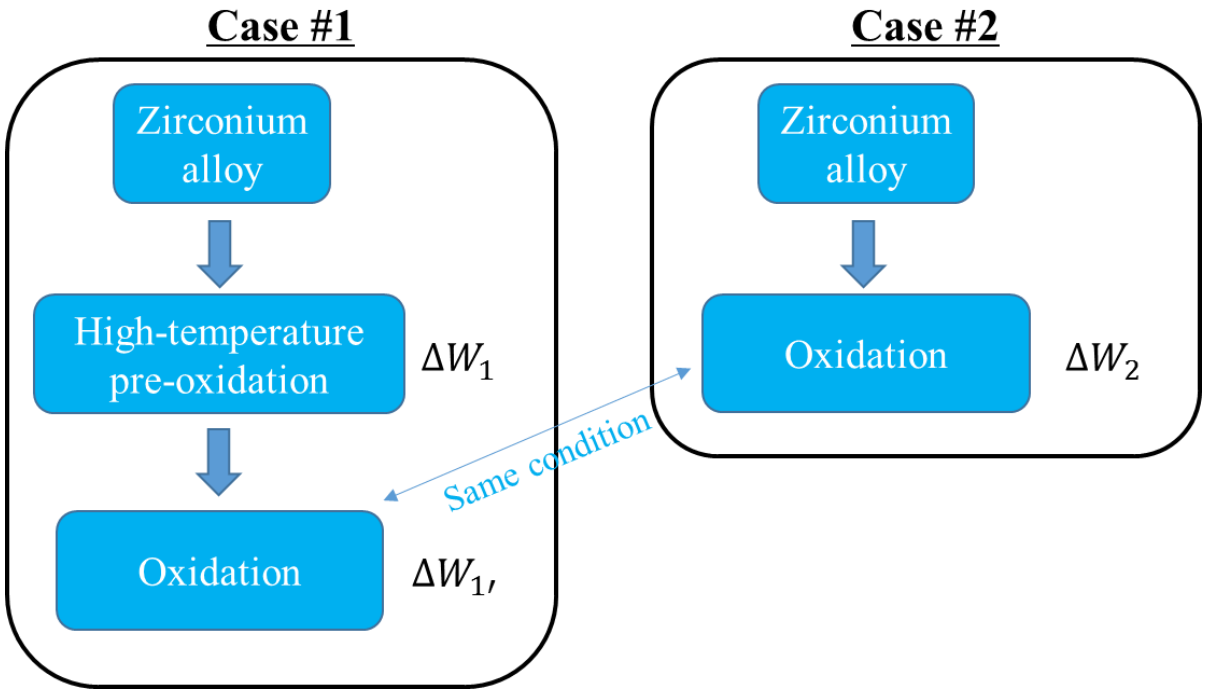


→Total weight gain = $\Delta W_{pre-oxidation} + \Delta W_{subsequent\ oxidation}$

→Weight gains of the pre-oxidized specimens are comparable to those of the non-pre-oxidized specimens at 1200 °C.

3-2. Subsequent Oxidation

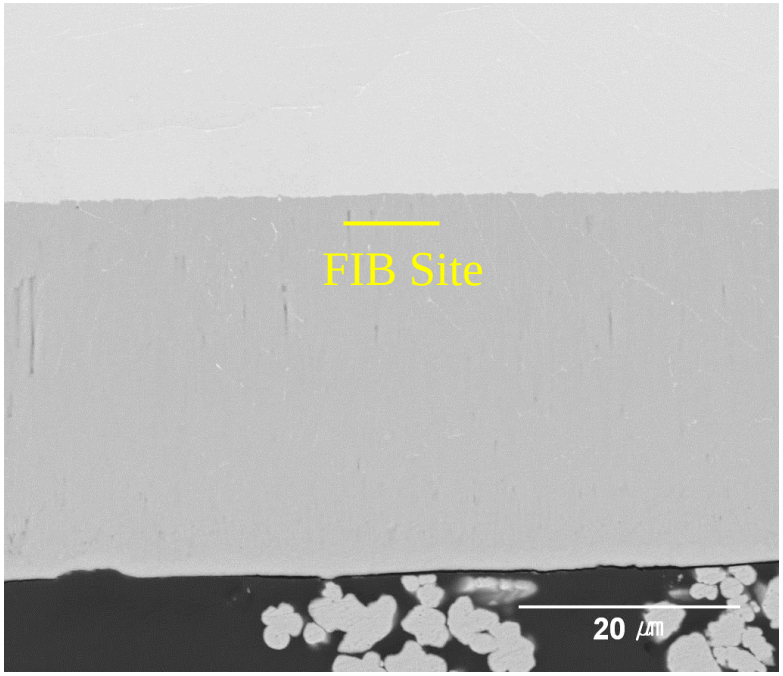
▪ Effects of the Pre-Oxides

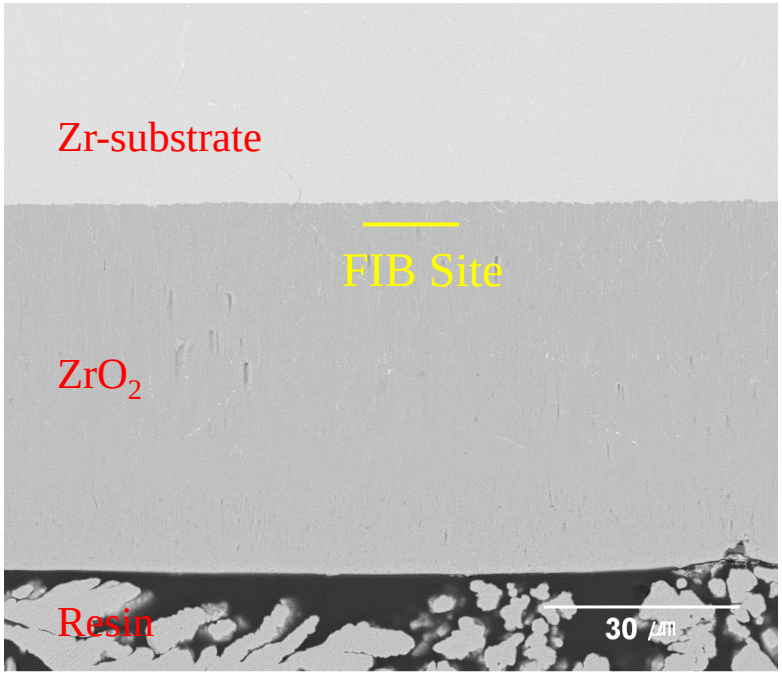


- $\Delta W_{1,} << \Delta W_2$
  : Heat and hydrogen generated from the oxidation of claddings will be significantly reduced during an accident.
- In some occasions, $\Delta W_1 + \Delta W_{1,} < \Delta W_2$
  : The pre-oxides are effective to enhance the oxidation resistance.
  : Possible reason for this was analyzed using TEM.

3-3. TEM Analysis

- **FIB Sites**





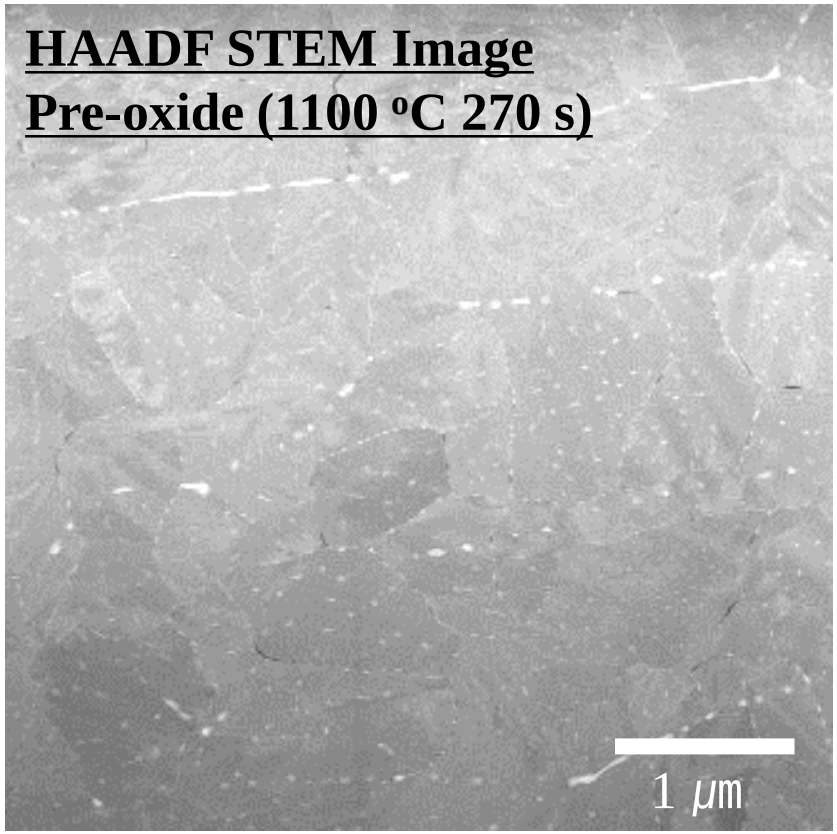
- **SEM BSE Image**
  Pre-oxidized at 1100 °C for 270 s
  No subsequent oxidation

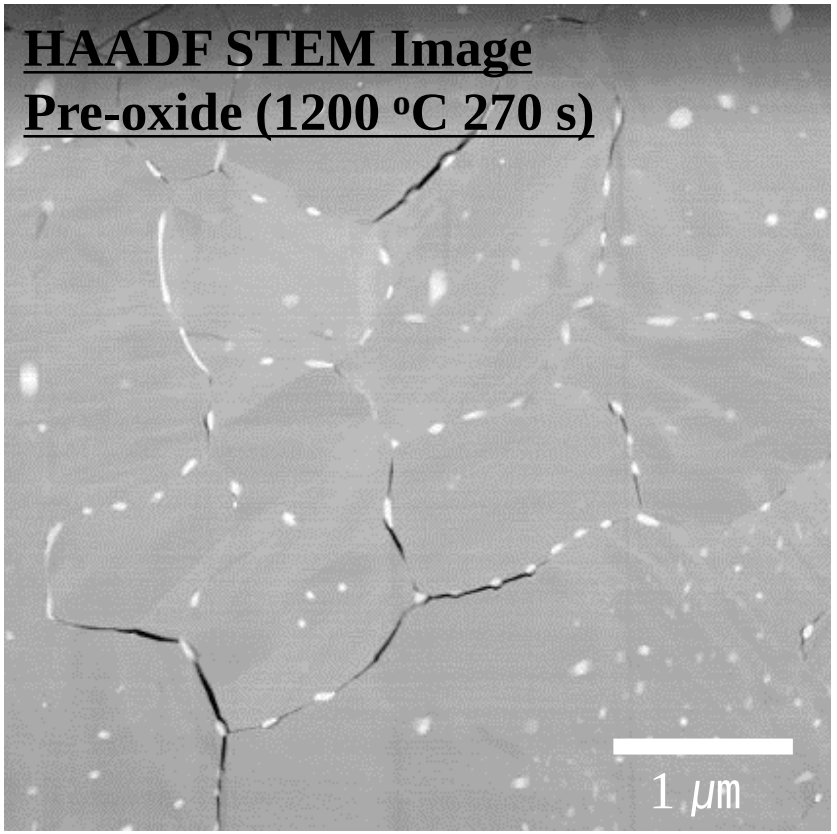
- **SEM BSE Image**
  Pre-oxidized at 1200 °C for 270 s
  No subsequent oxidation

*FIB : Focused ion beam

3-3. TEM Analysis

- **HAADF STEM Images**



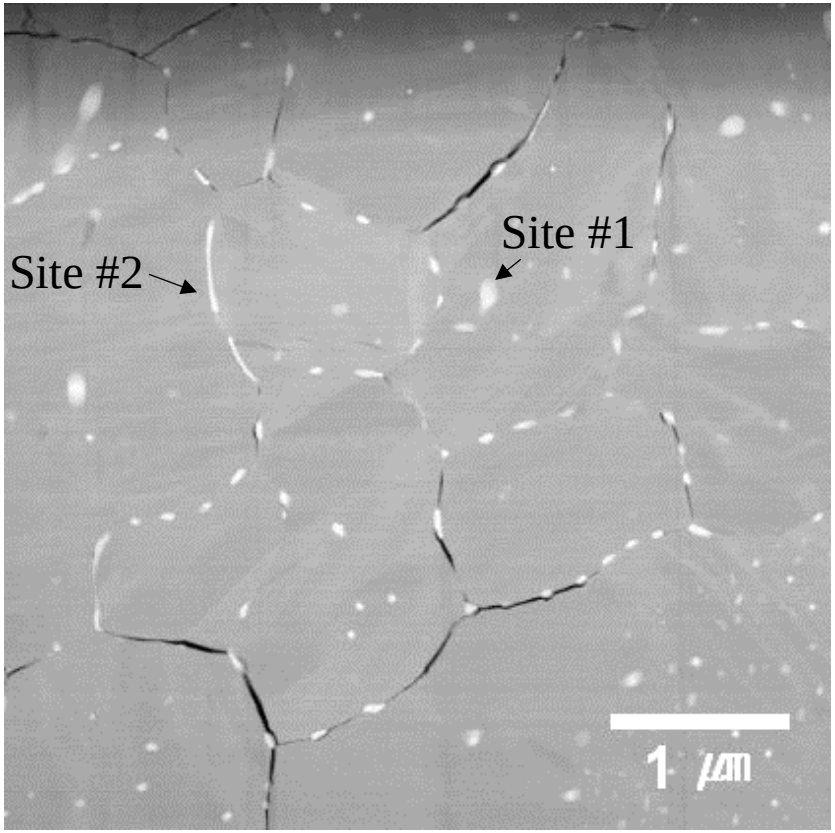


→The white particles are the precipitates containing Sn, Nb, or Zr.

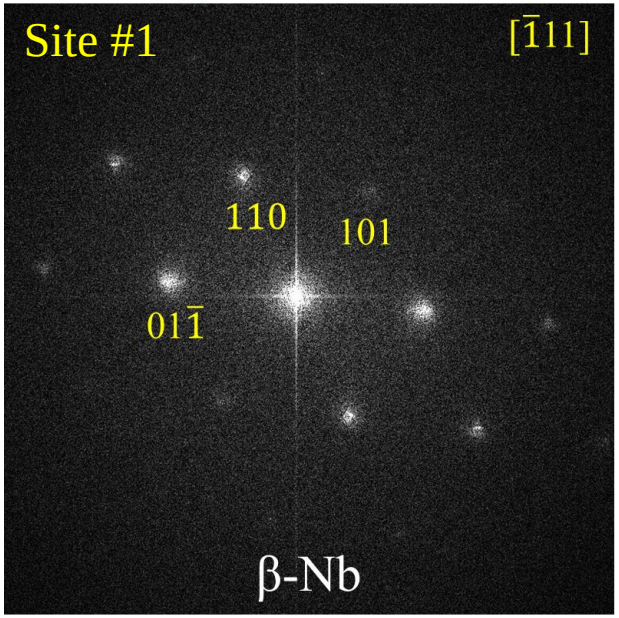
*HAADF STEM : High angle annular dark field scanning transmission electron microscopy

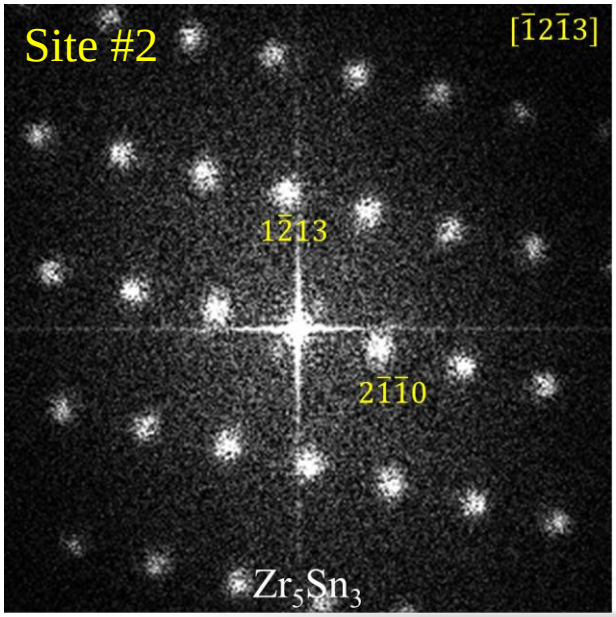
3-3. TEM Analysis

- **Diffraction Patterns**



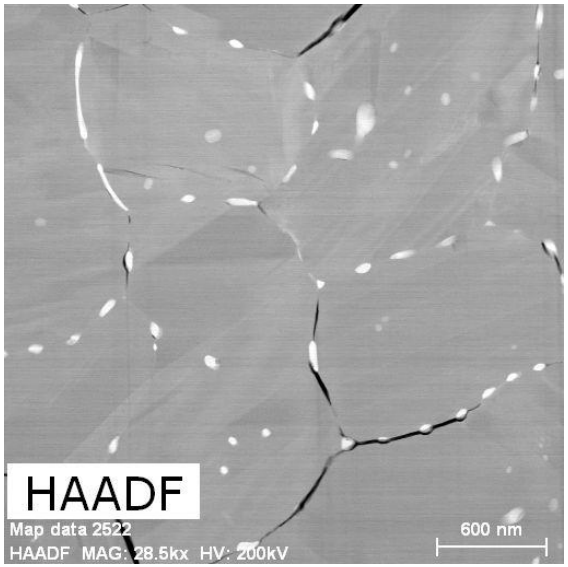
HAADF STEM Image
Pre-oxide (1200 °C 270 s)

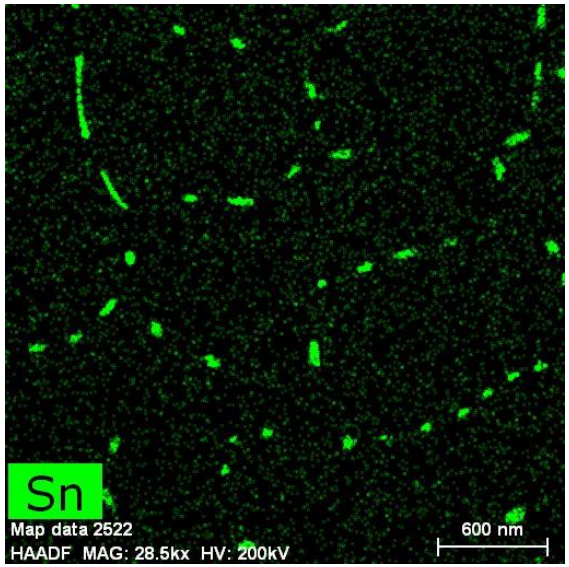


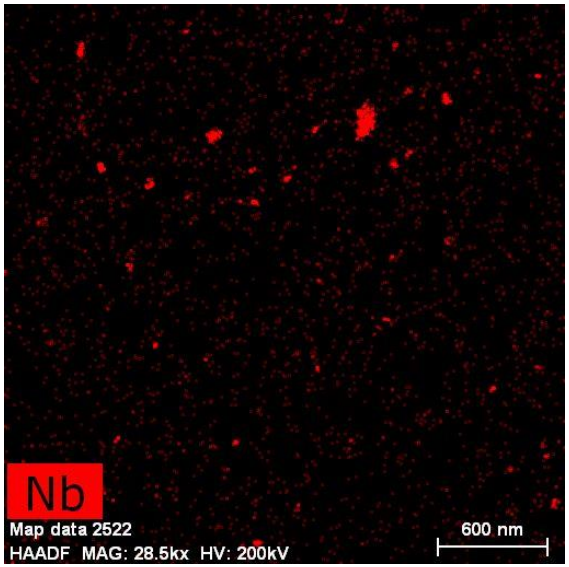


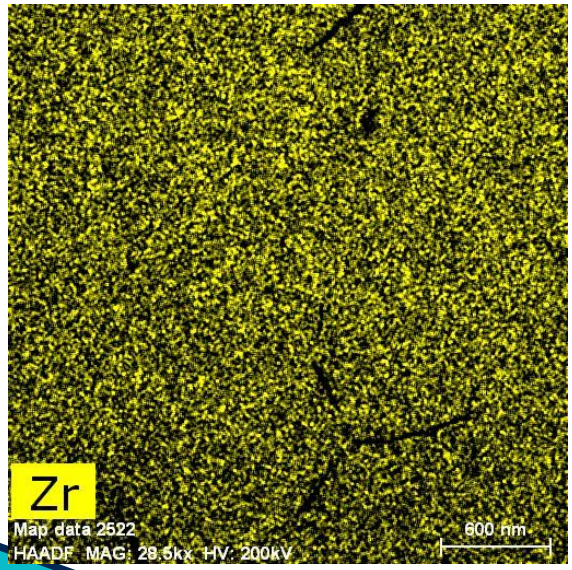
3-3. TEM Analysis

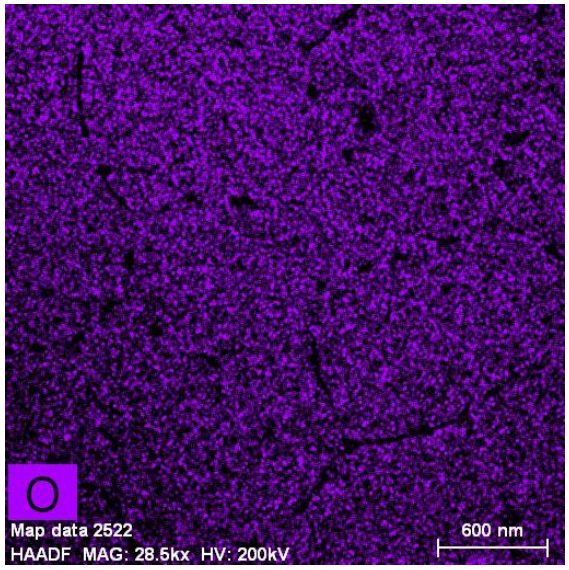
- **EDS Mapping**







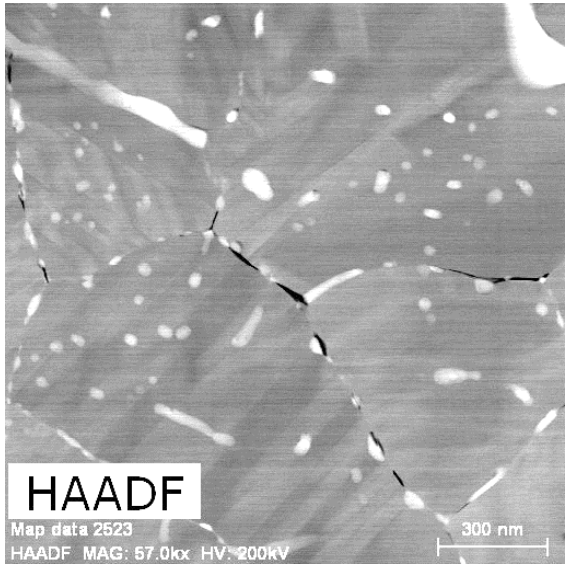


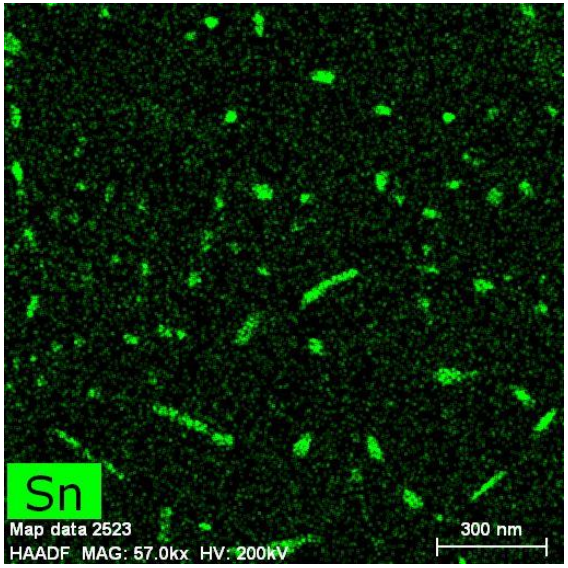


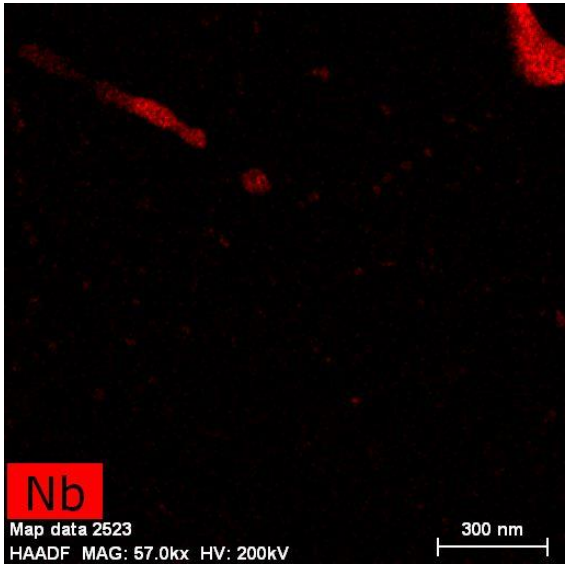
- Pre-oxide (1200 ^{o}C 270 s)
- Precipitates
  : Zr-Sn type
  : β-Nb

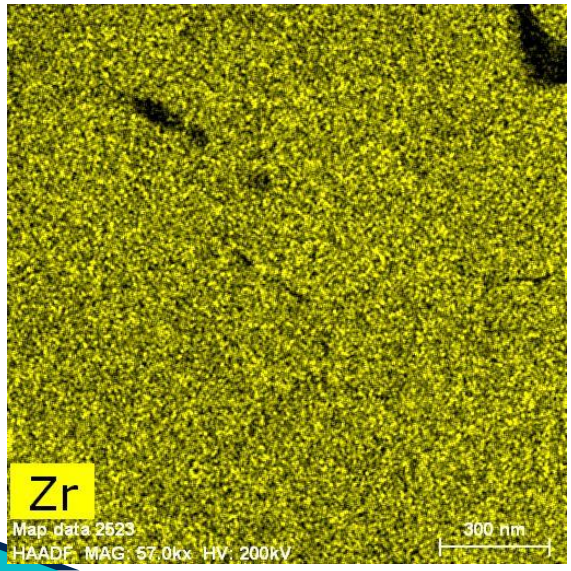
3-3. TEM Analysis

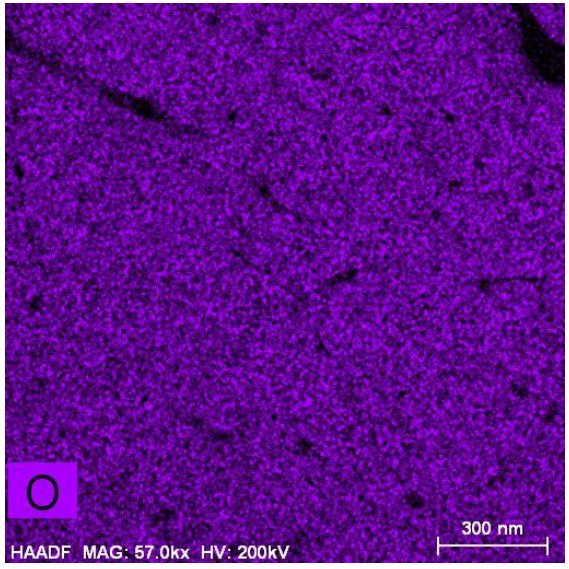
- **EDS Mapping**











- Pre-oxide (1100 °C 270 s)
- Precipitates
  : Zr-Sn type
  : β-Nb

3-3. TEM Analysis

Precipitates in Pre-Oxide

- Precipitates in the pre-oxide : **β-Nb, Zr-Sn type**

- β-Nb precipitates are present before the pre-oxidation.
  *Concentration of Nb in Zr > Solubility limit of Nb in Zr
  **Reference: J. P. Abriata et al., Bulletin of Alloy Phase Diagrams Vol. 3 No. 1 1982.*

- Zr-Sn type precipitates are not present before the pre-oxidation.
  *Concentration of Sn in Zr < Solubility limit of Sn in Zr
  **Reference: J. P. Abriata et al., Bulletin of Alloy Phase Diagrams Vol. 4 No. 2 1983.*

▪ Zr-Sn Type Precipitates

- Formation of Zr-Sn type precipitates during the pre-oxidation is unlikely considering the standard Gibbs free energies of formation at 1200 °C.

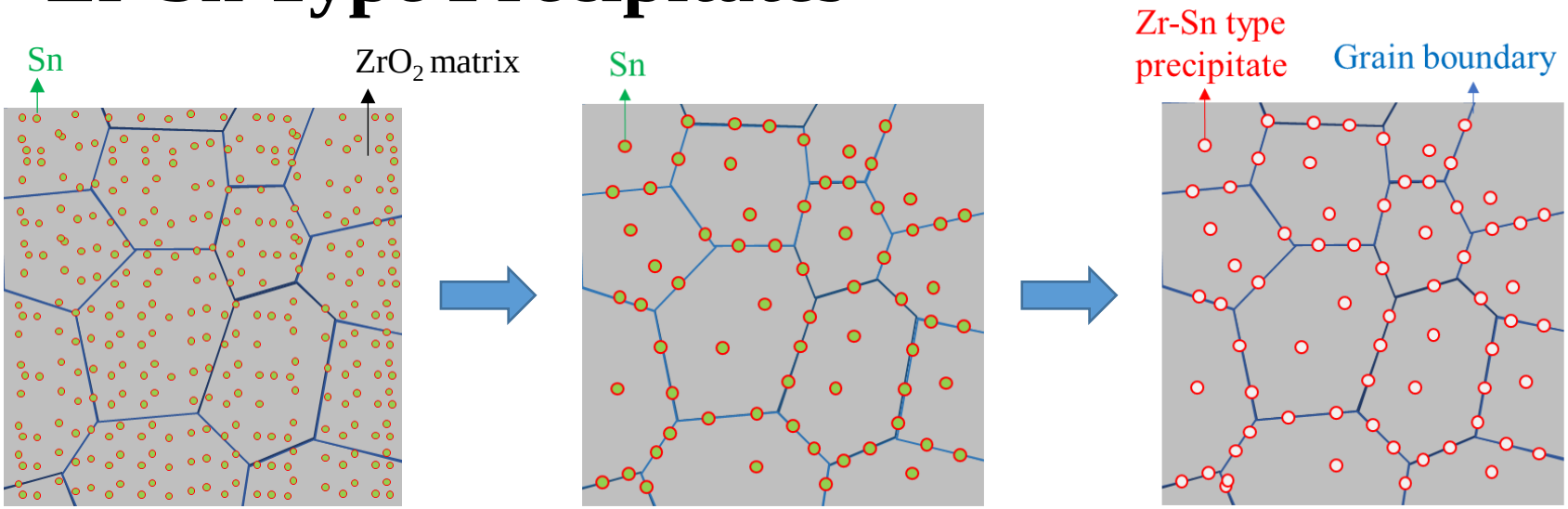
	ZrO_2	Zr_4Sn	Zr_5Sn_3	$ZrSn_2$
Standard Gibbs free energy of formation at 1200℃, kJ/(mol of Zirconium)	-856	-63	-140	-202

**Reference : NIST-JANAF Thermochemical Tables*
**Reference : Korotkova, N.V, Izvestiya Akademii Nauk SSSR, Metally, 1990*

- **The Zr-Sn type precipitates may be formed during the cooling.**
(β-ZrO_{2-x}(tetragonal) → α-ZrO_2(monoclinic) + α-Zr)
**Reference: V.F.Urbanic et al., Journal of Nuclear Materials 7.5 (1978) 251-261*

3-3. TEM Analysis

▪ Zr-Sn Type Precipitates



Pre-oxide formation **Pre-oxidation** **After cooling**

- The precipitates formed at the grain boundaries of oxides are known to enhance the oxidation resistance.

  **Reference: E. Polman, T. Fransen, P. Gellings, Journal of Physics: Condensed Matter, 1 (1989) 4497.*
  **Reference: C.M. Cotell, G.J. Yurek, R.J. Hussey, Oxid Met, 34 (1990) 173-200.*

- **The oxidation resistance enhancement presented in this study may be related to the Zr-Sn type precipitates.**

4. Summary



4. Summary

- Zr-1Nb-1Sn-0.1Fe alloy with the high-temperature pre-oxide show the significantly enhanced oxidation resistance during the subsequent oxidation at 1000 and 1100 ^{o}C.

- This oxidation resistance enhancement may be related to the Zr-Sn type precipitates formed at the grain boundaries of the pre-oxide.

Acknowledgement

We wish to thank Dr. Steinbrück in KIT, and Dr. Große in KIT for their useful and considerate comments.

FIRST IN CHANGE

Thank you!



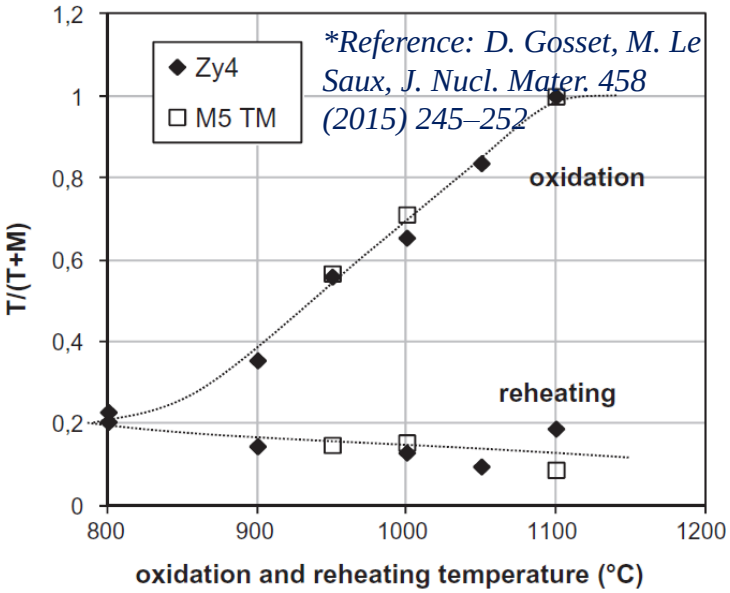
ULSAN NATIONAL INSTITUTE OF SCIENCE AND TECHNOLOGY

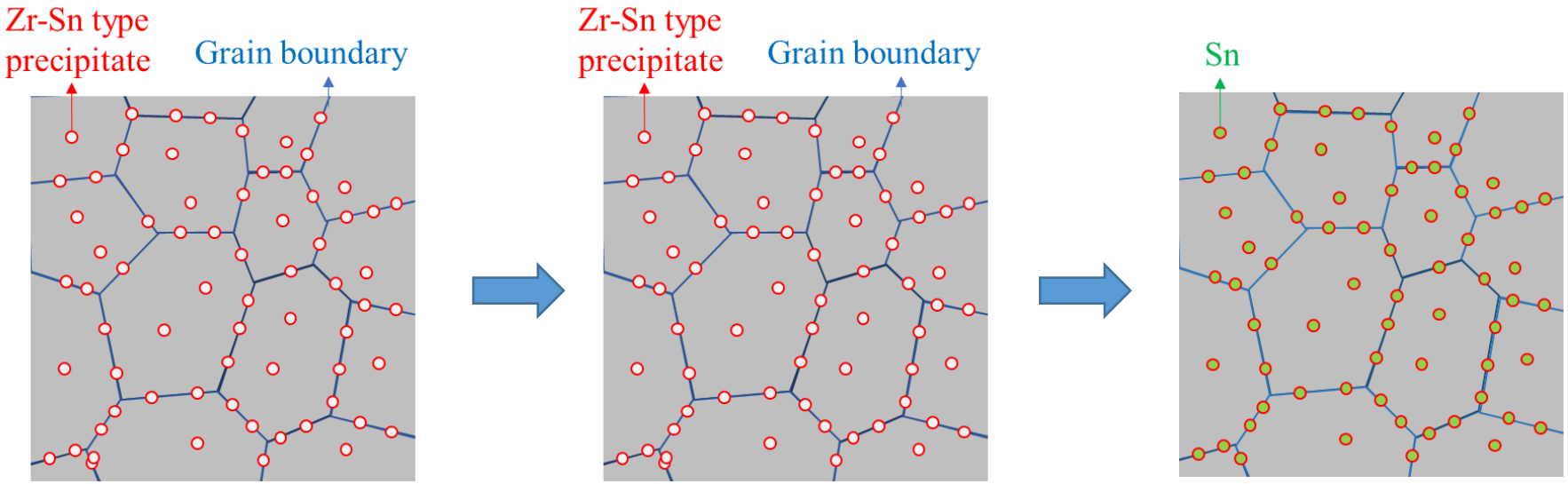
3-3. TEM Analysis

▪ Zr-Sn Type Precipitates

- The pre-oxide transforms to the tetragonal phase above 1200 °C.
  **Reference: D. Gosset, M. Le Saux, J. Nucl. Mater. 458 (2015) 245–252*
- At 1200 °C, Zr-Sn type precipitates may be decomposed, which may cause the oxidation resistance enhancement to disappear.
  (α-ZrO_2(monoclinic) + α-Zr → β-ZrO_{2-x}(tetragonal))

ZrO_2 tetragonal phase fraction





Summary Report of 23rd QUENCH Workshop

Xiaojun HE，Qian XU, Songtao JI

Nuclear Safety and Fuel Performance Research Lab.
China Institute of Atomic Energy

For study the new zircaloy cladding behavior under LOCA condition, the high temperature steam oxidation test facilities have been established in China Institute of Atomic Energy. By using these test facilities, several tests concerning with high temperature oxidation are lunched. The study of breakaway oxidation of Zr-4 and M5 cladding are especially introduced during the 23rd QUENCH Workshop, the contents include: the study methods, the experimental data, the models for predict the onset of breakaway oxidation. The results reveal that the breakaway oxidation performance of Zr-4 and M5 cladding is meet the requirement of current safety criteria under LOCA condition.

The progress of the PWR annular fuel R&D project in CIAE is also introduced. For increasing the economic and safety of nuclear energy, annular fuel assembly for PWR has been developing in CIAE since 2010. Five tasks, which are reactor core design, fuel assembly design experimental V&V， manufacture technology, safety study and in-pile tests, have been lunching to get the license of LTAs' irradiation test in current PWRs. These kinds of research work are supposed to be finished by 2020.



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New Zircaloy Cladding Behavior Study In LOCA condition and Annular Fuel R&D introduction in China

He Xiao-jun, Xu Qian, Ji Song-tao

Nuclear Safety and Fuel Performance Research Lab.
China Institute of Atomic Energy

Nuclear Energy In China

- China is making more effects to develop new energy to ensure the country's energy security and boost economic growth.
- Nuclear, Solar, Wind and Water Electricity have been fast developing to meet the request of air quality, climate change and fossil fuel shortages;
- To raise the percentage of China's electricity produced by nuclear power from the current 2% to 4% by 2020 (now 3%);

Nuclear energy fast development requires high performance fuel assembly :

- Independent Intellectual Property
- More Safe and More Economic
- Used at Current and Next Generation NPPs



NPPs in China mainland, (NNSA, China)

In Operation 37 units (Sep 17, 2017)

Under Construction 19 units

26 Sites is planning to Construct NPP

China National Nuclear Corporation (1955-)

CF3/4 Fuel Assembly (Nuclear Power Institute of China)

N18 / N36 / N45 Cladding

Annular Fuel Assembly (China Institute of Atomic Energy)

China General Nuclear Power Group(1994-)

Step12 Fuel Assembly Fuel Assembly Based AFA-3G

CZ Cladding

National Power Investment Corp. (2015-)

China Power Investment Corp.

State Nuclear Power Technology Corporation (2007-)

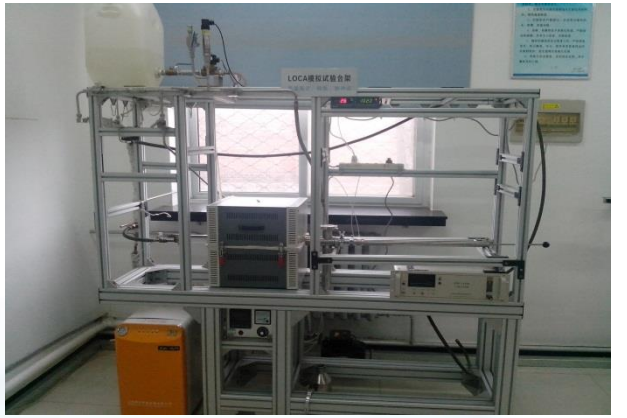
AP1400 Fuel Assembly (SNERDI, Shanghai)

SAZ Cladding

Overview of the LOCA study in CIAE

Research Tasks

- *Establish the test facilities for LOCA study*
- *High-temperature oxidation*
- *Transient analysis code development and modification*
- Brittleness behavior： **breakaway oxidation** and toughness after quench
- Ballooning and burst
- Anti thermal shock ability with axial-loading
- High temperature Creep behavior



High-temperature oxidation test loop

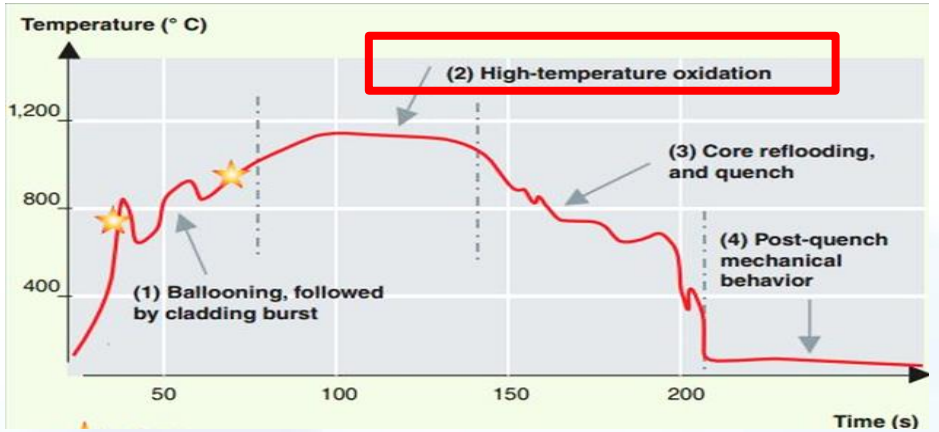
Samples

- Zero-hydrogen , un-irradiated cladding
- M5, ZIRLO, Zr-4, SAZ-4, SAZ-6…..



LOCA test loop

Breakaway Oxidation Study



LOCA Criteria (Brittle criteria)

17%ECR

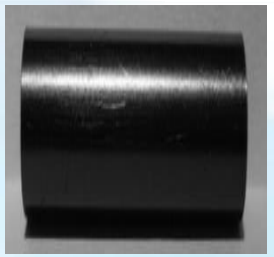
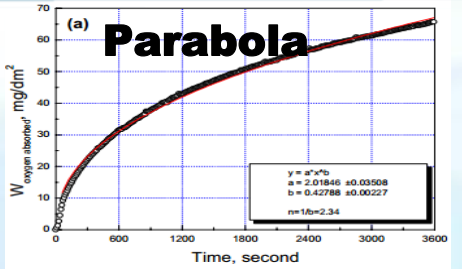
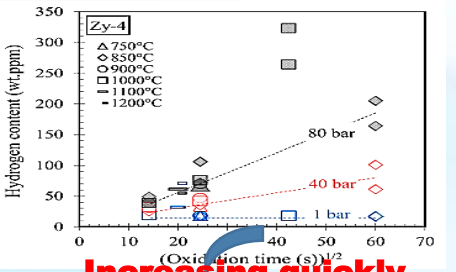
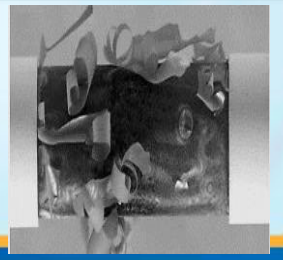
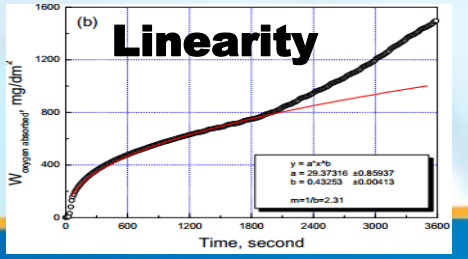
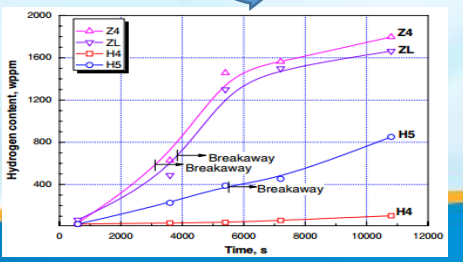
B-J formula

$2545 \cdot \exp(-11550/T_K) \cdot \sqrt{\tau} \leq 17$

Maximum oxidation time at certain temperature

safe — Lower — 17% — Beyond — brittle — Unsafe

Breakaway oxidation

	Oxidation film	Oxidation weight	Hydride
Normal Oxidation			
Breakaway Oxidation			

Increasing quickly

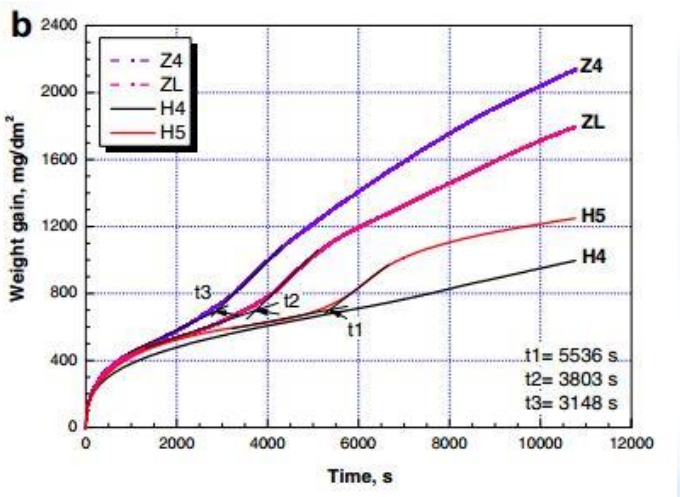
ANL 2008: set *200μg/g* as the brittle criteria for the breakaway oxidation

- **For certain cladding material, get the onset time of breakaway oxidation, *used as a reference for current safety criteria*;**
- **Evaluate the response time of ECCS**

Y.Yan,'High-temperature steam-oxidation behavior of Zr-1Nb cladding alloy E110', ANL,2009

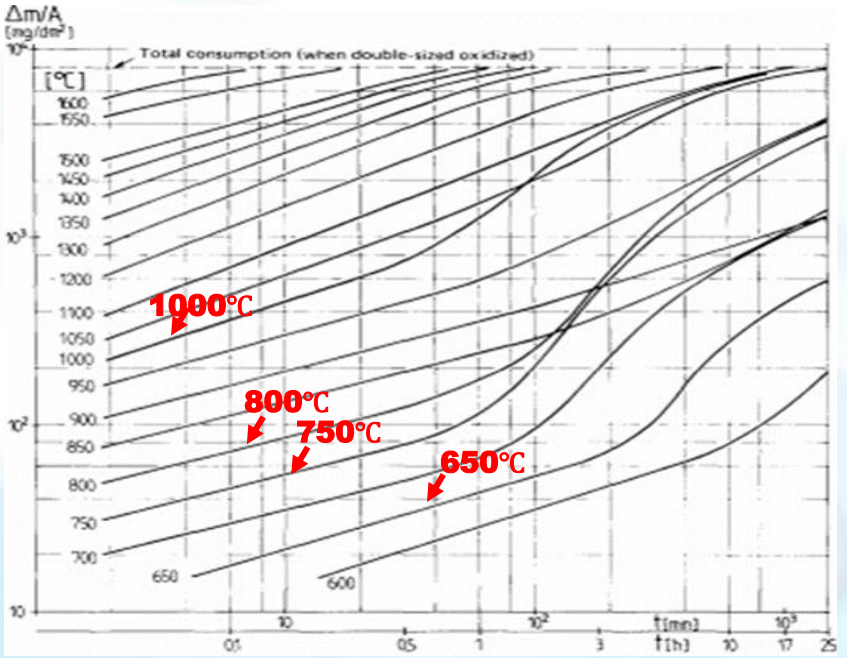
Jong Hyuk Baek,'Breakaway phenomenon of Zr-based allooys during a high-temperature oxidation',2008

- Lots of factor could affect the process of the breakaway oxidation :
  - *Materials* : composition, heat treatment…..
  - *Environments*: temperature, time, oxidation medium, pressure and flux of the steam



Different materials@*1000℃ steam*, KAERI

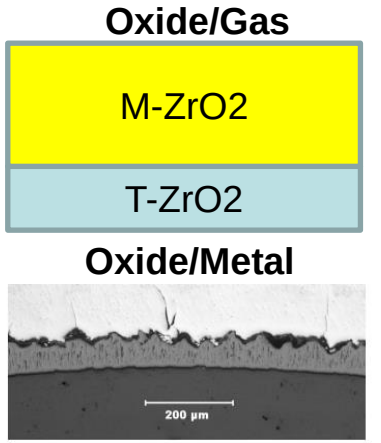
Jong Hyuk Baek, etc.'Breakaway phenomenon of Zr-based alloys during a high-temperature oxidation',KAERI,2008.



S.Leistikow, etc.'The oxidation behavior of Zircaloy-4 in steam between 600 and 1600℃'.

Different Temperature Zr-4@600~1600℃ steam

- Zr-4: No breakaway oxidation appear 850-950℃, >1100 ℃



The phase transition theory

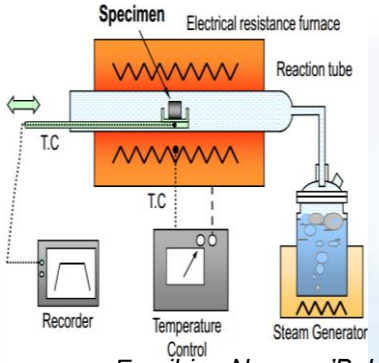
•During the normal oxidation , compressive stress let the T-ZrO2 stably exist;

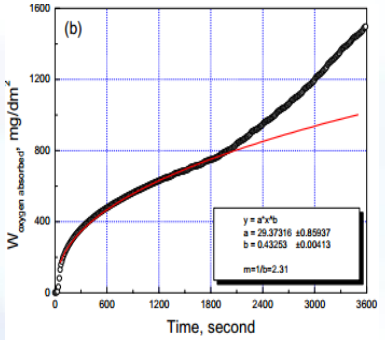
•During the breakaway oxidation, the compressive stress will be loosen due to O/M wave-style interface, T-ZrO2→ M-ZrO2 , the oxidation rate will speedup through the cracks of M-ZrO2

Samples

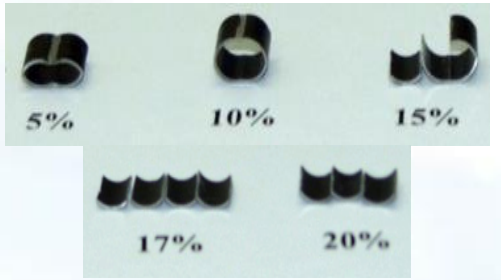
- M5, Zr-4 (made in china)

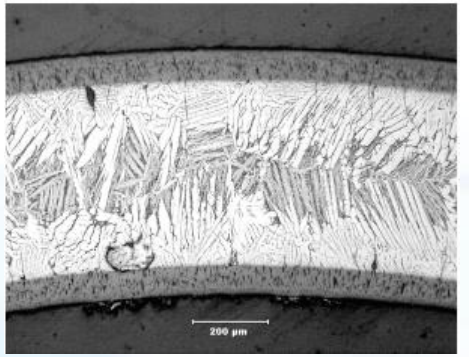
Methods





Fumihisa Nagase,'Behavior of high burnup fuel during LOCA-Key observations and test plan at JAEA



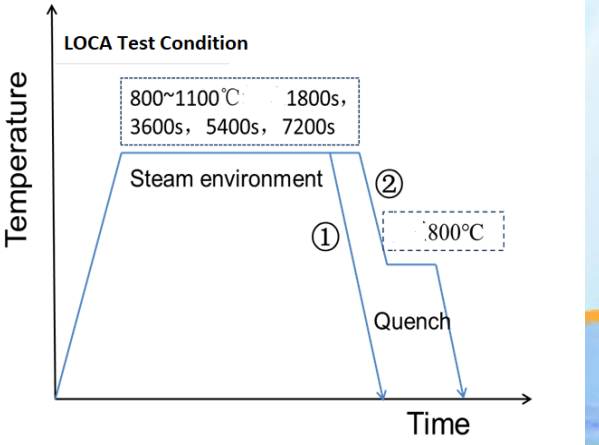


'Cladding embrittlement during postulated LOCA-of-Coolant Accidents', NUREG/CR-6967

H-T oxidation → **Oxidation weighting** → **Ring-compression** → **Microstructure analysis**

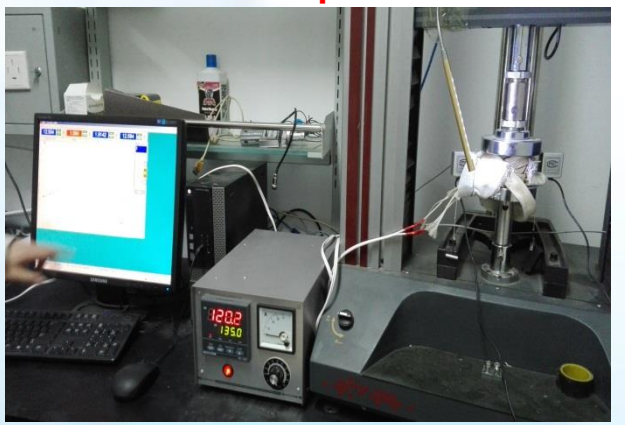
Test condition

- *800-1100 ℃, Steam, steam + air (for oxidation)*
- *135 ℃, compression rate: 2mm/min (for compression- ring test)*





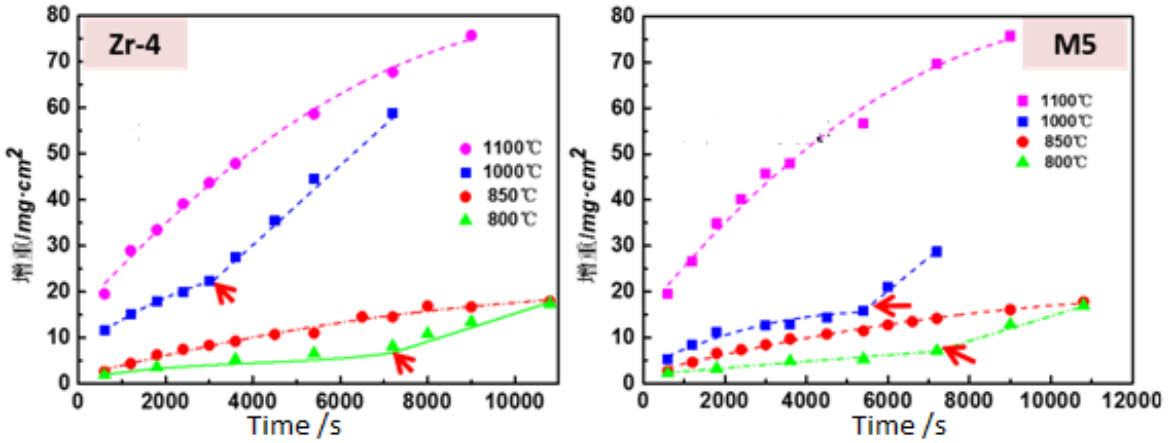
breakaway oxidation test loop



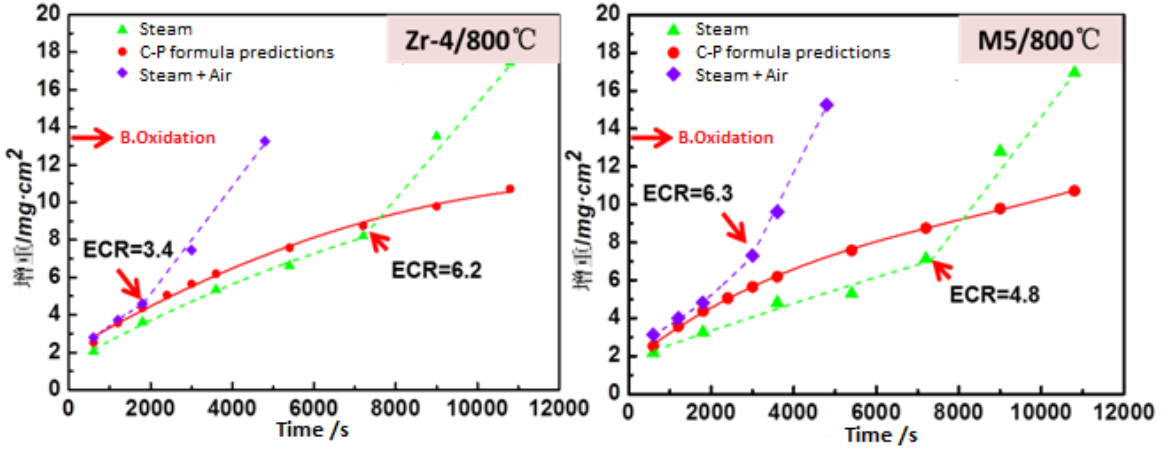
Ring-compression Installation

Experimental data for oxidation weight

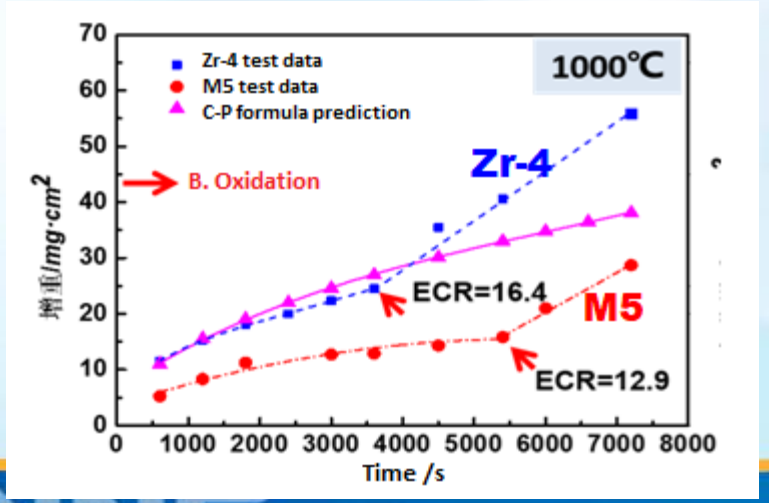
- Different temperature



- Different environment (steam, steam + air)



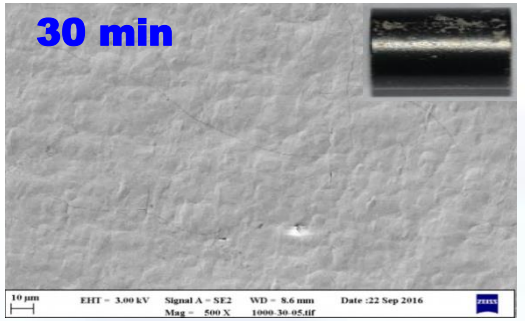
- Different materials (1000℃，steam)

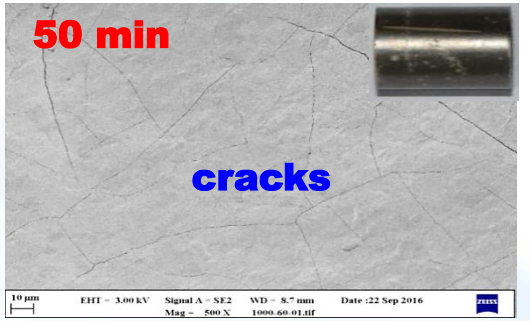


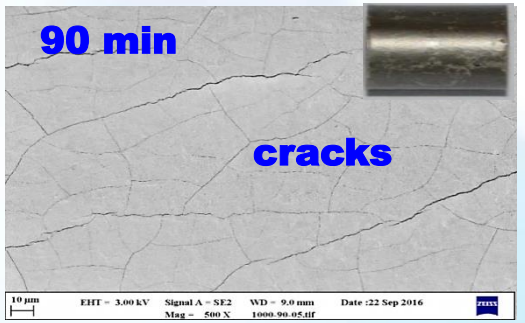
- Breakaway oxidations are found in 800℃ and 1000 ℃, (850 ℃ or 1100 ℃, no oxidation)； the higher T get the faster oxidation rate； it's earlier for mixture gas to get breakaway oxidation compare with pure steam.
- Oxidation weight accords good with C-P formula, while breakaway oxidation weight is not.

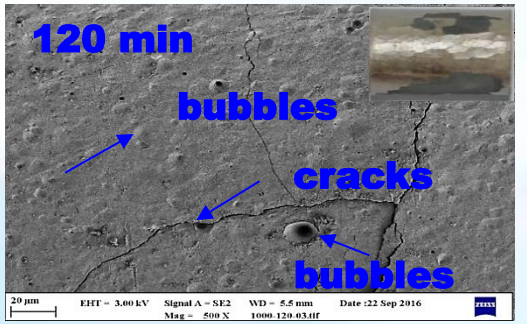
Microstructure of oxide film surface (SEM)

- Zr-4 (1000℃, steam)

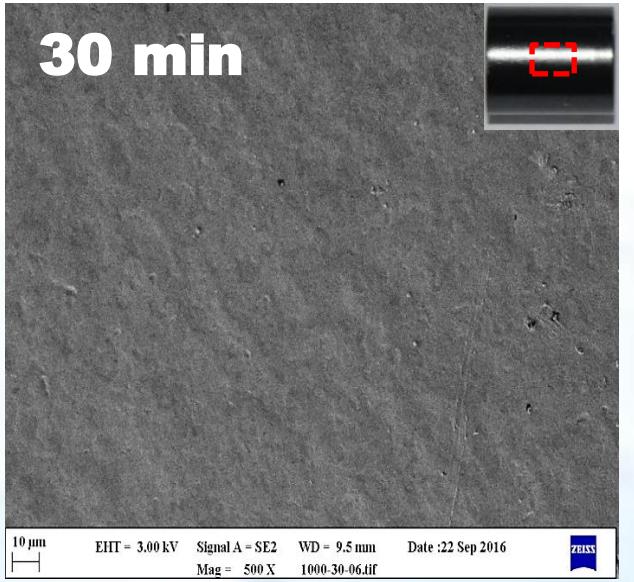


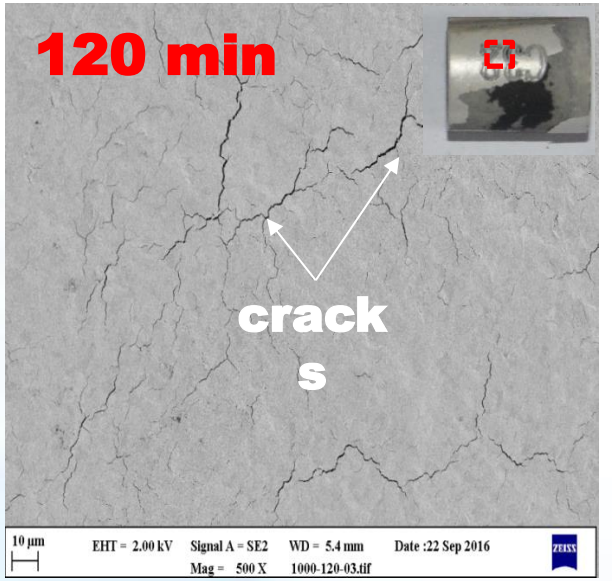






- M5 (1000℃, steam)



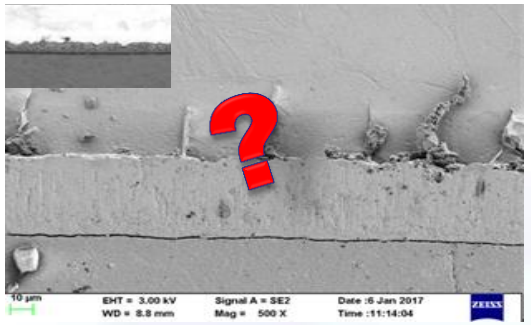


- No film bubbles are found in M5 cladding

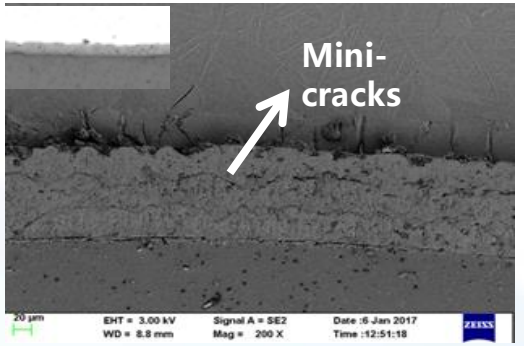
30min- no cracks
50min- breakaway oxidation，cracks
90min- breakaway oxidation，more cracks
120min- breakaway oxidation，deep cracks, film bubbles

Microstructure of oxide film cross-section

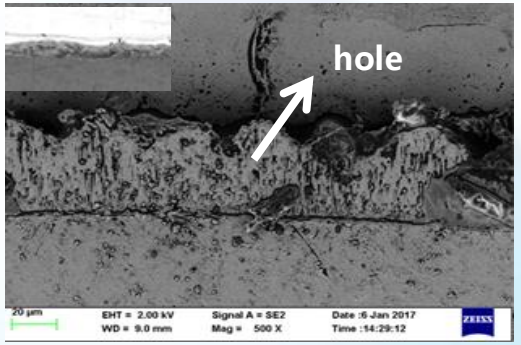
- Zr-4 (1000℃, steam , 500X)



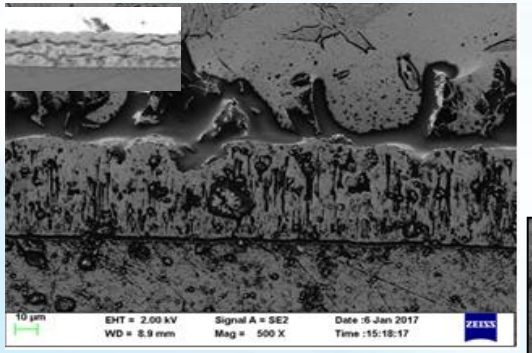
20 min



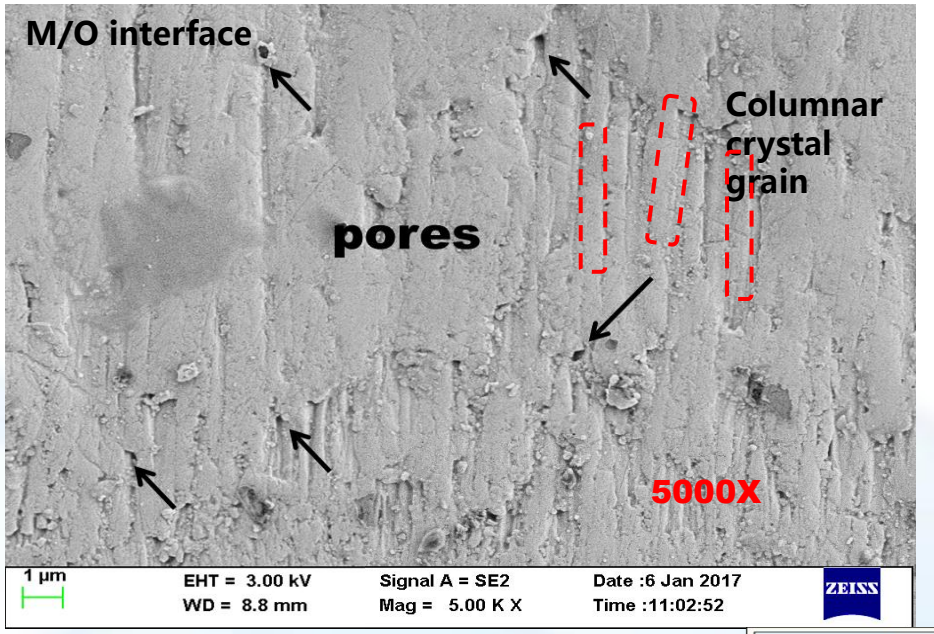
40 min



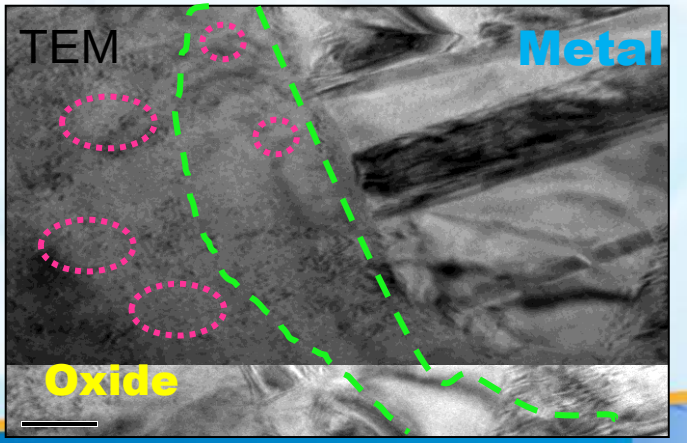
50 min

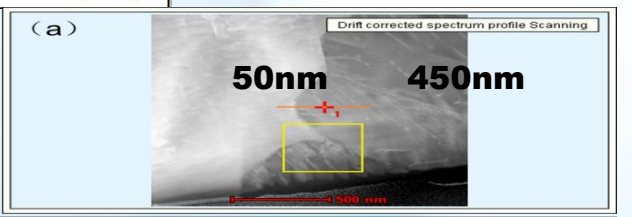


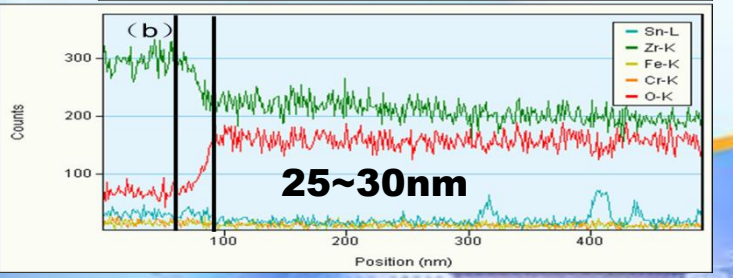
90 min



- *Columnar crystal in O/M interface region;*
- *Nano-holes existed*
- *Two layers exist in 25-30 nm near O/M interface, Zr_3O , small size nano-hole ;over this region, ZrO_2 , bigger size nano-holes*



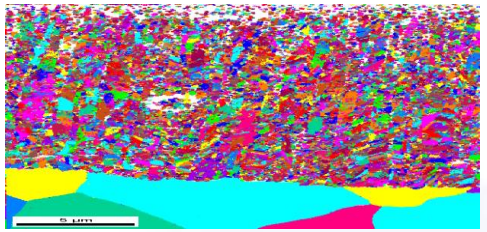




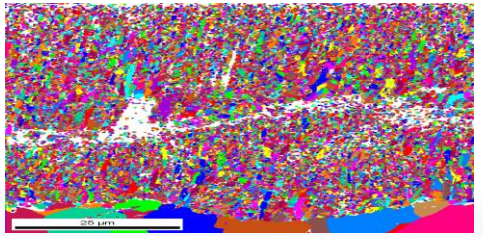
- *At start period, The oxide film looks like complete and dense*
- **With the oxidation, holes and mini-cracks appears**
- **After breakaway oxidation, bigger holes and deep cracks**

Grain distribution in oxide film (steam)

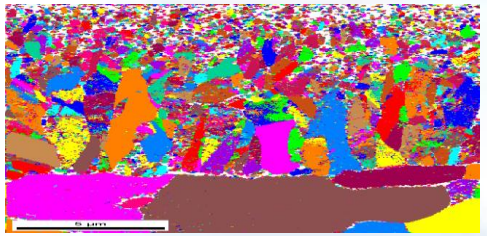
Zr-4 1000℃ 10min



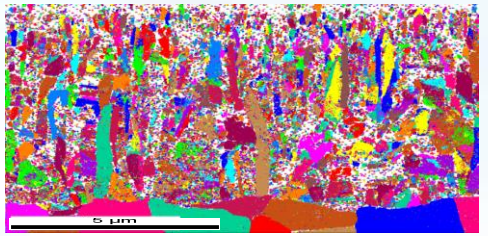
Zr-4 1000℃ 50min



M5 1000℃ 30min



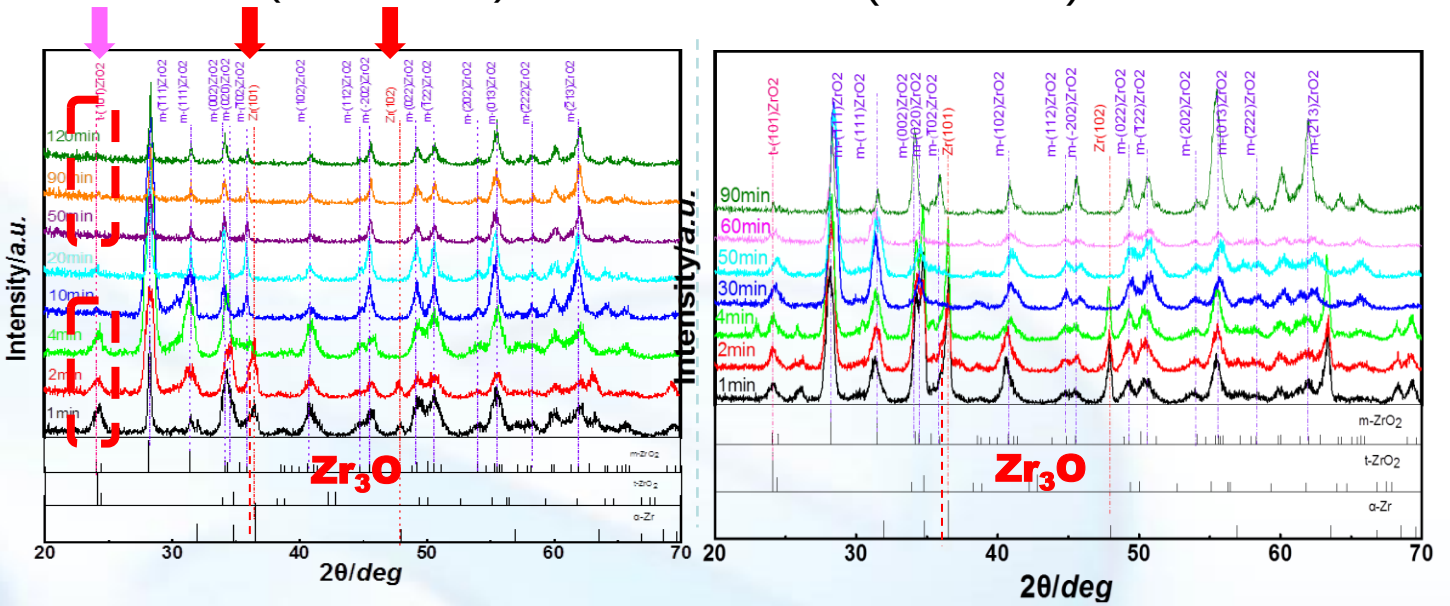
M5 1000℃ 90min



***EBSD results*: Big columnar grain → small equiax grain → Higher grain density → diffusion rate speed up**

Oxide film structure (1000℃, steam)

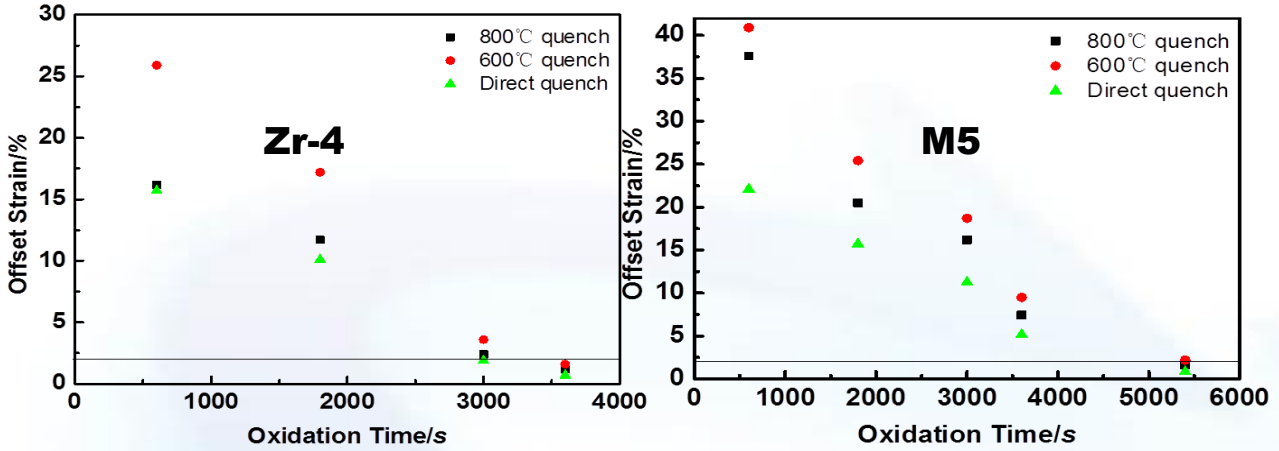
- Zr-4 (1-120 min)
- M5 (1-90 min)



XRD results:

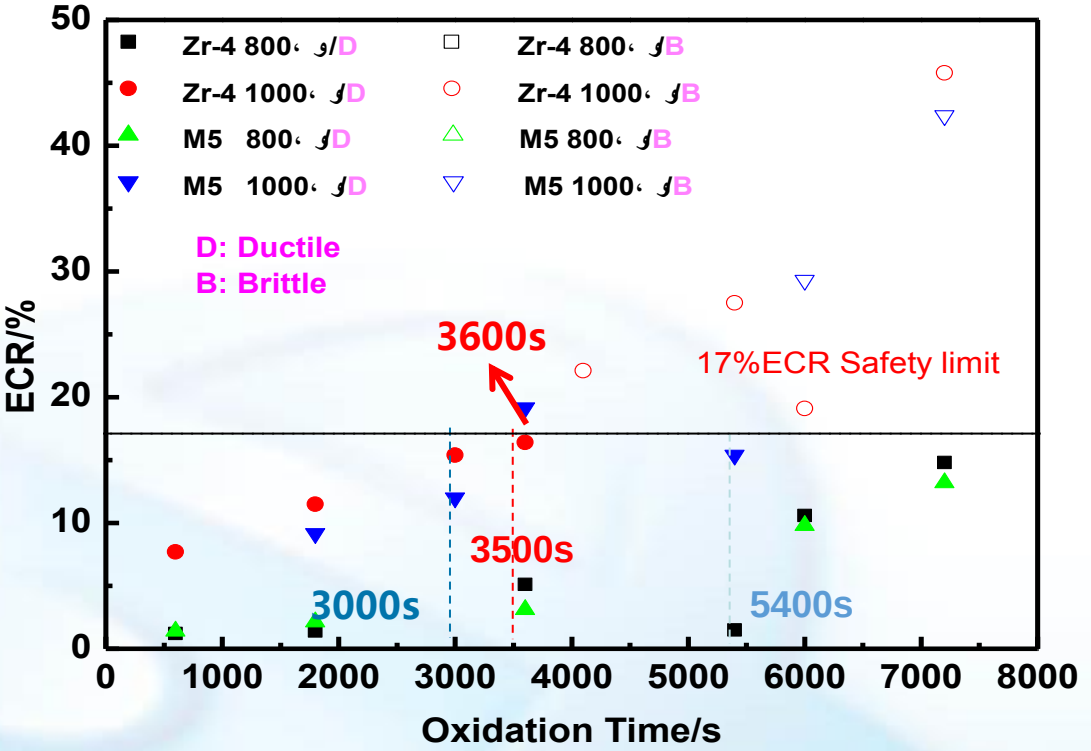
- *t-ZrO_2, m-ZrO_2, Zr martix, and Zr_3O existed;*
- *m-ZrO_2 is the main structure for oxide film, Zr_3O is found in M/O interface region;*
- *Higher t-ZrO_2 is found in M/O interface region, after breakaway oxidation, almost disappear*
- *Three layers: Zr martix+Zr_3O ; t-ZrO_2 + m-ZrO_2 ; m-ZrO_2*

Quench temperature's effects



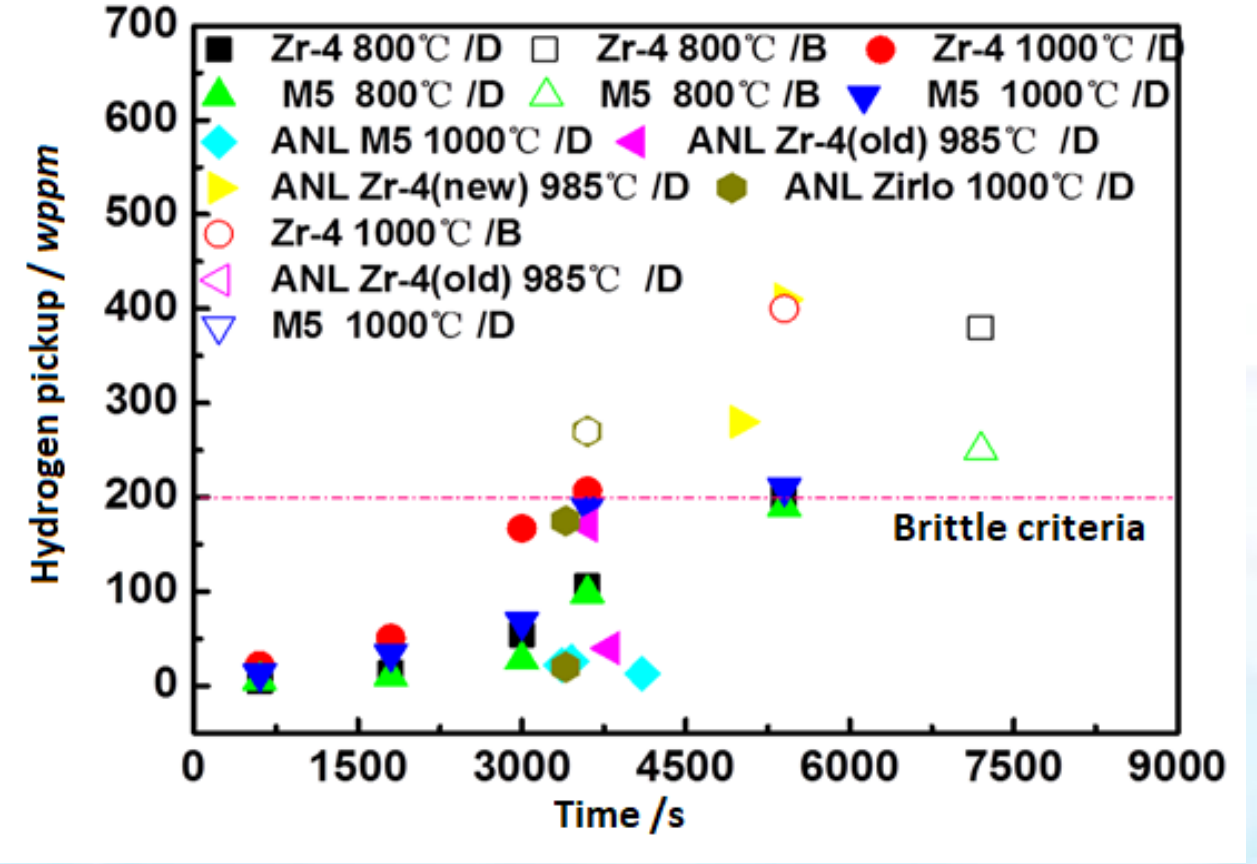
- *Quenching at 600℃case , Ductility of samples are bigger;*
- *Direct quenching at 1100℃case is more conservative than real LOCA situation*
- *Under CIAE's test condition, M5 has a better performance*

Ductile and Brittle in different ECR before and after breakaway oxidation



- *M5:B.O appears after the ECR-17% criteria;*
- *Zr-4: B.O appears before ECR-17% criteria, but still meet the requirement(ductile).*

- ANL: 200ug/g hydride criteria for breakaway oxidation (brittle criteria)



200ug/g hydride criteria is suit for Zr-4 and M5 materials' breakaway oxidation

'Cladding embrittlement during postulated LOCA-of-Coolant Accidents', NUREG/CR-6967

Conclusion

- Establish facilities and get the experimental data concerning with breakaway oxidation (M5/Zr-4);
- Develop models for predicting onset of breakaway oxidation;
- For Zr-4 material, more brittleness after breakaway oxidation, but still meet the current 17%-ECR criteria;
- 200ug/g hydride criteria is suit for Zr-4 and M5 materials' breakaway oxidation

Future work

- More temperature range
- More cladding materials (SAZ-6 and SAZ-4)
- Modeling and code development
- ……..

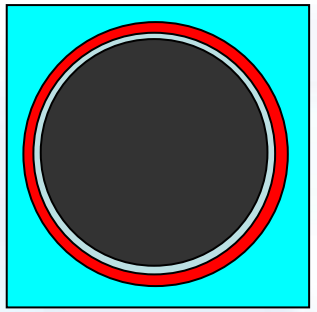


Thermal Gravimetric Analyzer

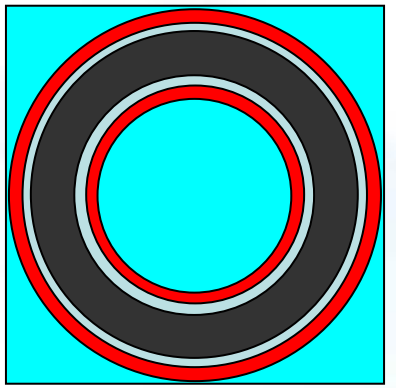
More details : Mrs. Xu Qian E-mail: **magic-xu@hotmail.com**

Annular Fuel R&D Introduction in China

- **What's Annular fuel?**



Solid Fuel Rod



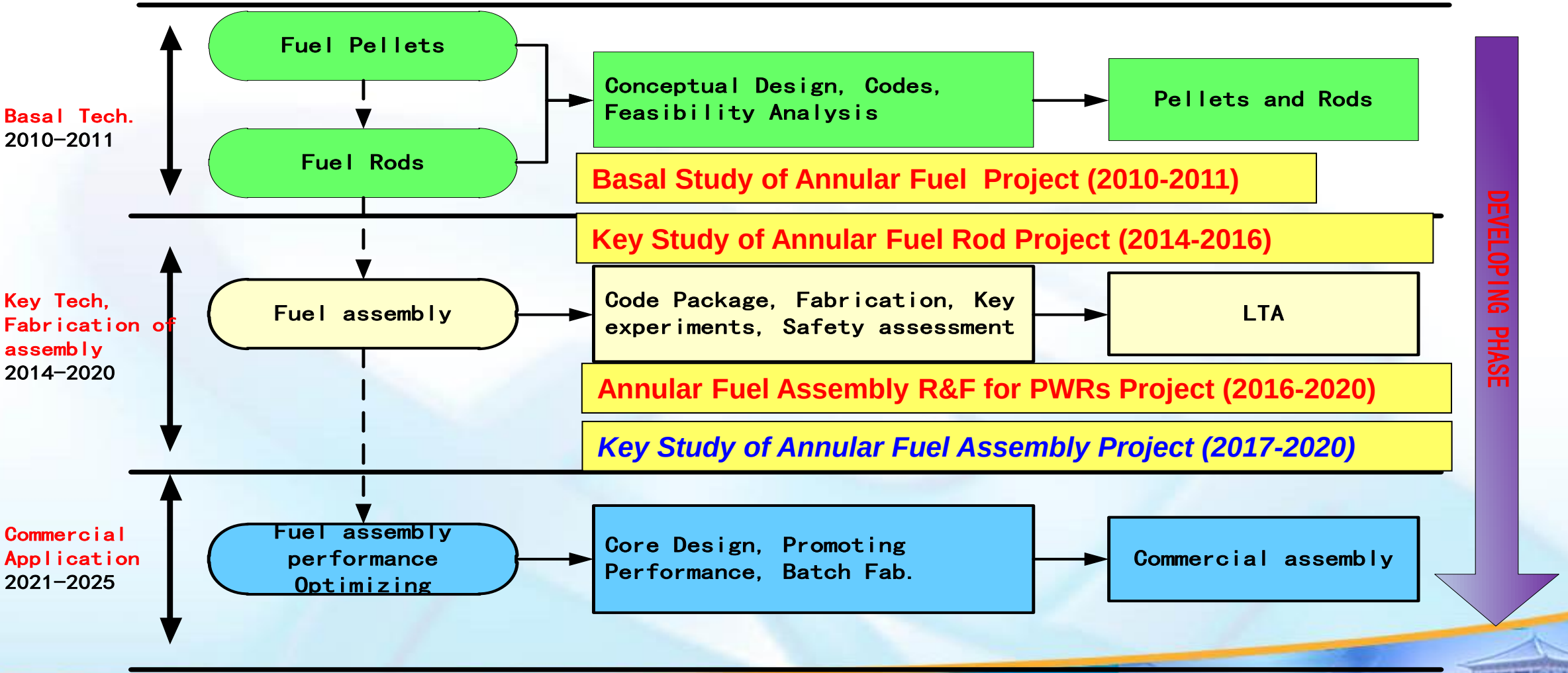
Annular Fuel Rod

- Double sides coolant - *lower temperature*
- Thinner pellet - *lower store energy*
- Two claddings – *safer in transient condition*
- *……*

- ***Annular fuel could be used to clearly increase the power density, while keeping or improving the safety of NPP.***

  - ***NERI***, 2001-2005, U.S, for building new NPPs
  - ***NEPO,*** 2006-2013, U.S, for upgrading current NPPs
  - ***Dual-Cooled Fuel R&D Project,*** 2007-2015, Republic of Korea, for upgrading current OPR-1000 NPPs;
  - Other organizations

Roadmap for Developing Annular Fuel



Motivation

- **Increase the Economic and Safety of Nuclear Energy;**
- ***Develop a new type nuclear fuel assembly with China Brand***
  - Listed in the 12th and 13rd Five-Year Plan (2010-2020)
  - Listed in the " Energy Technology Revolution Innovation Action Plan (2016-2030)" , as a main content of nuclear fuel innovation.

Objective

- ***2014-2020, Key design and manufacture technologies, complete the out-pile validation experiments and short rod in-pile irradiation test, get the license for LTAs' irradiation in current commercial PWRs;***
- 2021-2025, Engineering and technological research on application of annular fuel in PWR, 2025, annular fuel are expected to be used in current commercial PWRs

- **To get the license, 5 tasks are defined to develop technologies for design, manufacture and in-pile & out-pile performance study**

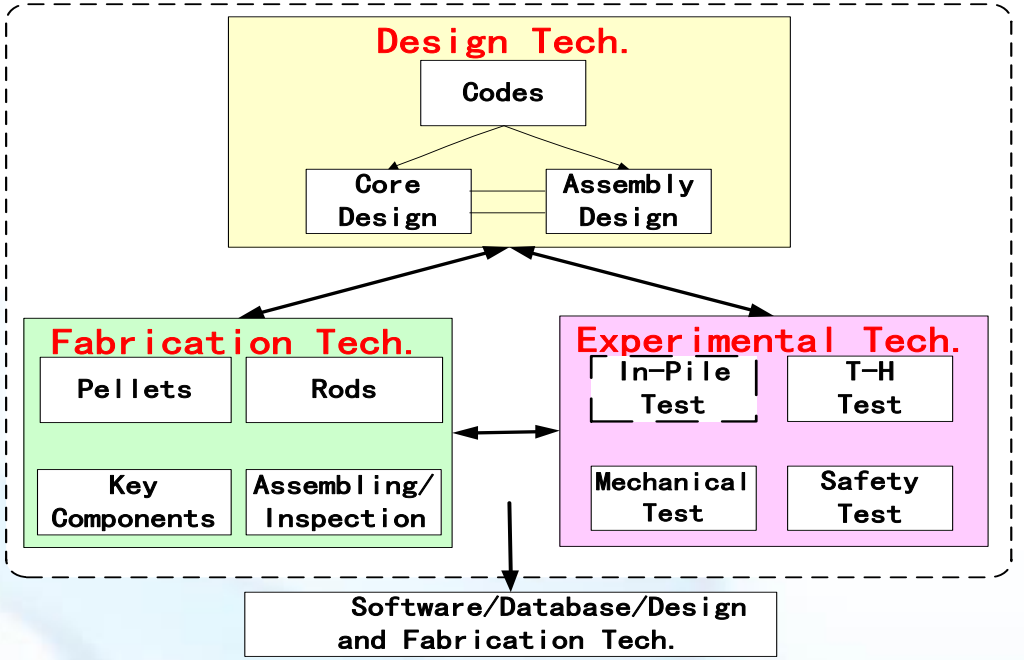
Task 1：Design of reactor core with annular fuel assembly

Task 2：Design of annular fuel assembly and related experimental V&V

Task 3：Manufacture of annular fuel assembly

Task 4：Safety performance of annular fuel assembly

Task 5：In-pile performance of annular fuel assembly

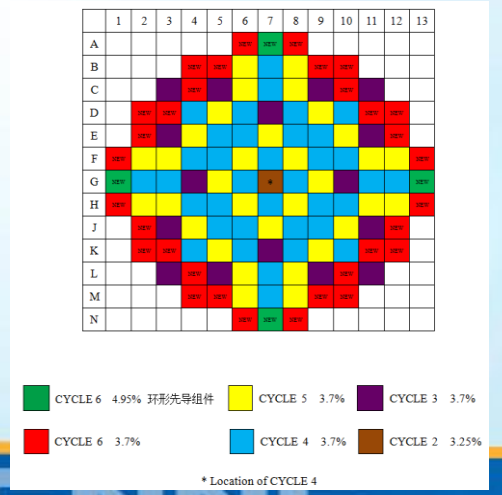


Proposed Roadmap for Key technologies /Fabrication of FA

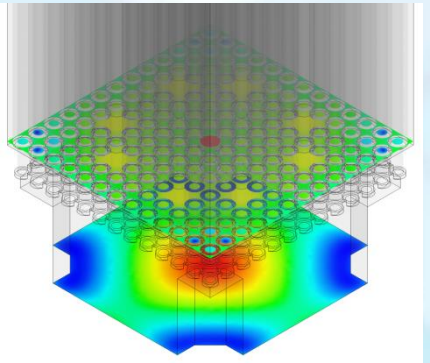
Task1：Design of reactor core with annular fuel assembly

- **Development and V&V of computer codes for annular fuel design and application**
  - Physics；T/H；Fuel performance；safety analysis；Mechanics
- **Reactor design for application of annular fuel in Qinshan-2 NPP**
  - Power promotion up to 15%（full AF/mixed core of AF and SF）：Nuclear design，T/H design，Safety analysis；
- **Mixed core design related to irradiation of LTA in commercial NPP**
  - Mixed core： Nuclear design，T/H design，Safety analysis ；

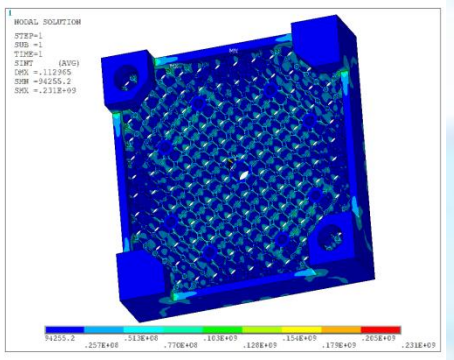
Applied field	Code name	Main function
Reactor physics	MVP-MVPBURN	Monte Carlo burnup calculation
	CASMO5-CMSLINK5-SIMULATE5	Reactor physics and fuel management
	MCNP	Reactor physics
thermal-hydraulics	SAAF	Full annular FA core T-H analysis
	MARC	Annular-Solid FA mix-core T-H analysis
	ANASYS-CFX	Flow field analysis
Safety analysis	RELAP5-MOD3	DBAs analysis
	MELCOR/ASTEC	Severe accident analysis
	ICARE/CATHE	Severe accident analysis
Fuel performance	AFPAC	In-core performance analysis
	ANASYS	Mechanical performance analysis



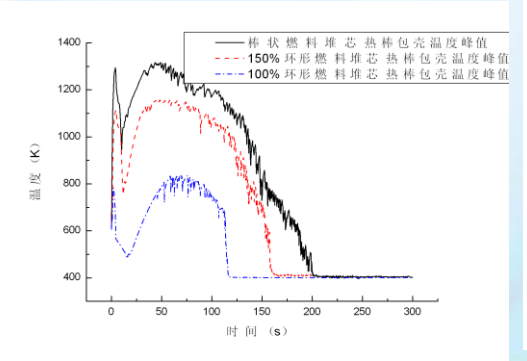
Mixed core design



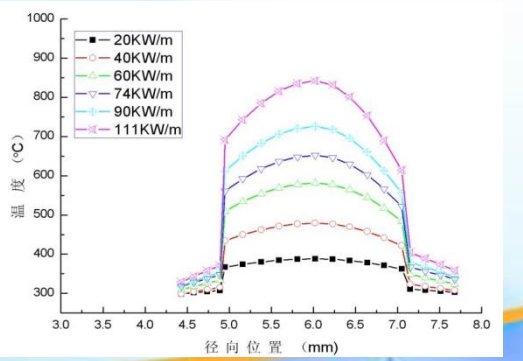
T-H CFD Analysis



Mechanical analysis



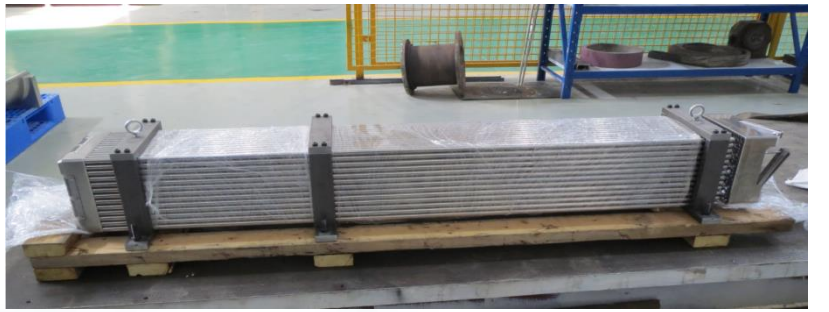
DBA analysis



Fuel Performance analysis

Task 2: Design of annular fuel assembly and related experimental V&V

- **Mechanical design of annular fuel assembly**
  - Fuel rod、upper/ bottom nozzle、Space grid、Gide tube；Design standards for annular fuel assembly；
- **Studies on T/H behavior of AF and validation of the AF design with T/H experiments**
  - T/H experiments for key components of AF ； Heat transfer experiments using small bundles、two phase flow instability and heat split, Flow resistance 、Structural integrate test 、Flow induced vibration and creep test
- **Mechanical test**
  - Key components ： Fuel rod、upper/ bottom nozzle、Space grid、Gide tube, etc..；
  - Fuel assembly：stiffness、impact、vibration、 earthquake tolerance、transportation and operation



Half length experimental fuel assembly



HTHP Test Loop



T-H Test Loop

Task 3：Manufacture of annular fuel assembly

- Fuel pellet：Gd_2O_3 doped pellets
- Fuel rod: Spring, Welding and Inspection
- Upper/Bottom nozzle
- Space grids
- Structural components
- Assembling technology：8 full size assemblies
- Inspection technology



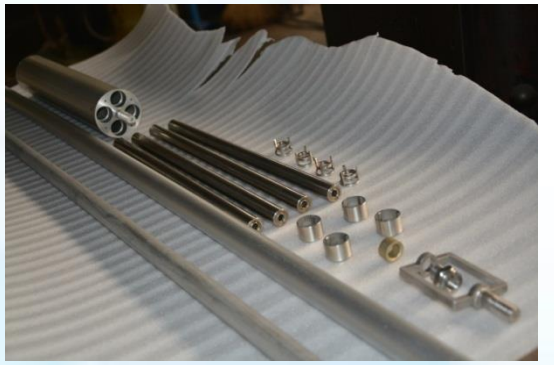
Annular pellets BY CNNFC



Annular rods BY CNNFC



In pile test rods (19.75%)



In-pile test rig components



Full Length Fuel rod

Task 4： Safety performance of annular fuel assembly

- Experimental studies on AF performance under LOCA conditions
- CHF experiments of annular fuel assembly
  - 3X3 full length (heterogeneous heating in axial direction)
- Safety issues related to unbalance of heat transfer between inner and outer channel
- Safety issues related to Growth difference of inner and outer cladding
- Blockage of inner flower channel
- Cladding Safety Performance under current NPP conditions
- Methodology of severe accident analysis with AF reactor core



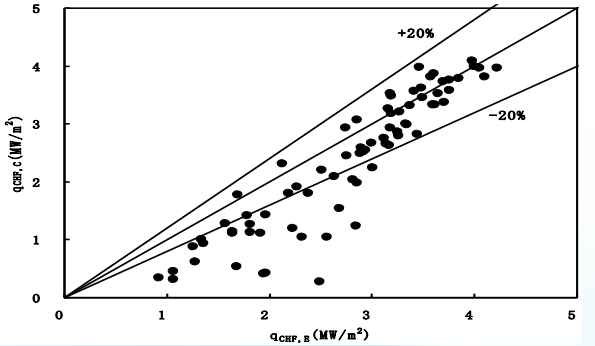
Buckling Samples with different Inner pressure



Samples for cladding safety experiments



Multi-function Test Loop



Calculated CHF vs. experiments data

Inner cladding CHF test

D/L (mm/m)	Pre. (MPa)	Flow (kg/m2s)	Inlet Sub. (K)	Outlet Sub. (K)	CHF (MW/m2)	No. of data
4.62/0.5	1.8－20.6	556－4055	110-354	1-169	0.77－9.3	118
7.98/0.8	2.0－20.4	476－1653	53-361	3-158	0.26 － 4.95	193
10.89/1.1	1.7－20.0	454－1144	169-345	4-141	0.92－3.3	56
8.20/2.4	3.4-19.7	1041-3776	355.2-0	/	0.5-4.4	101

Task 5：In-pile performance of annular fuel assembly

- **Irradiation test and related PIE of small AF bundle in research reactor (load in June 29, 2017)**
- **Irradiation of LTAs in commercial NPPs**
  - Inspection technology for LTA irradiation
  - Transportation Technology of LTAs
  - Licensing for LTAs irradiation in current operating PWR

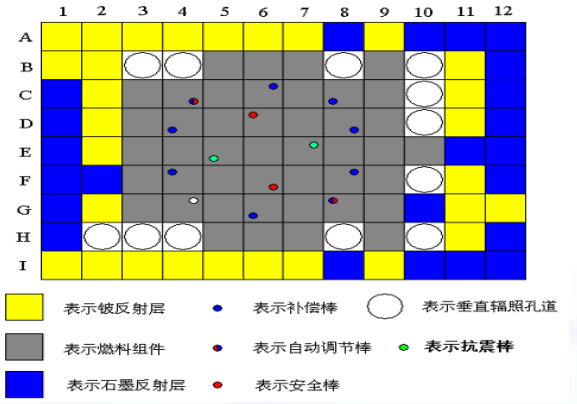
Organization of the Research Project

Manage Unit

中国核燃料有限公司 China Nuclear Fuel Corporation

Execute Unit

中国原子能科学研究院 CHINA INSTITUTE OF ATOMIC ENERGY
中核北方核燃料元件有限公司 CHINA NORTH NUCLEAR FUEL CO.,LTD
环境保护部核与辐射安全中心 Nuclear and Radiation Safety Center
中国核电工程有限公司 China Nuclear Power Engineering Co., Ltd.
NPIC 中国核动力研究设计院 NUCLEAR POWER INSTITUTE OF CHINA
西部新锆核材料科技有限公司 Western Energy Material Technologies Co.,Ltd
Studsvik
中核建中核燃料元件有限公司 CNNC Jianzhong Nuclear Fuel Co.,Ltd.
上海交通大学 Shanghai Jiao Tong University
西安交通大学 XI'AN JIAOTONG UNIVERSITY



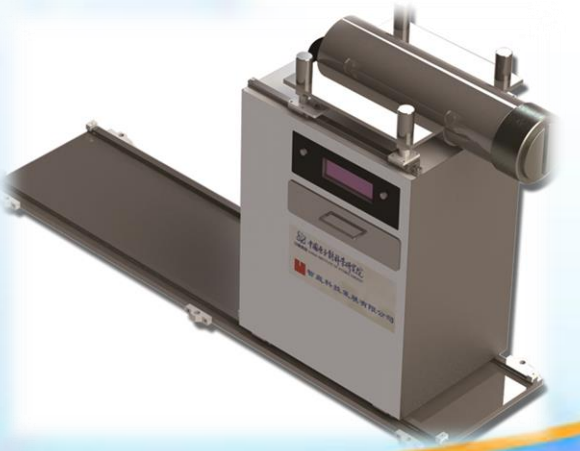
Cross-section of Swimming pool reactor



In-pile test rig



Test rig is loading in 49-2 experimental reactor



FDDS for Fuel failure inspection



Thanks for your attention

Title: Secondary hydriding of zirconium-based fuel claddings at high temperature (LOCA conditions)
Part 1: Multi-scale study of hydrogen distribution
Part 2: Effect of high hydrogen contents on metallurgical and mechanical properties

Thai LE HONG[1,2], Matthieu LE SAUX[1], Jean-Christophe BRACHET[1], Jérôme CREPIN[2] *et al.*

[1] DEN–Service de Recherches Métallurgiques Appliquées (SRMA), CEA, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France

[2] MINES ParisTech, PSL Research University, Centre des Matériaux, CNRS UMR 7633, BP 87, 91003 Evry, France

Abstract

During oxidization in steam at High Temperature (HT) under hypothetical Loss-Of-Coolant Accident (LOCA) conditions, zirconium alloy fuel cladding tubes can absorb, in some conditions (*e.g.* after burst occurrence and during subsequent HT steam oxidation), an important amount of hydrogen, up to thousands of wt.ppm locally. The behavior of the cladding can be affected by this secondary hydriding since hydrogen is known to modify metallurgical and mechanical properties of zirconium alloys. The purpose of this study is to refine the description of secondary hydriding and to improve both experimental database and understanding of the effect of high hydrogen contents up to 3000 wt.ppm, on metallurgical and mechanical properties of zirconium alloys, during and after cooling/quenching from high temperatures (β_{Zr} domain).

In a first part, we will illustrate a recent multi-scale analysis of hydrogen partitioning within Zr-based claddings having experienced secondary hydriding at HT. The objective was to map quantitatively, at different scales, the spatial distribution of hydrogen (and oxygen) within M5™[1] clad segments having experienced ballooning and burst at HT followed by steam oxidation at 1100 and 1200°C, and final direct water quenching down to Room Temperature (RT). This was done by coupling information from several advanced analytical techniques available at CEA Saclay: neutron radiography/tomography, electron probe micro analysis, micro elastic recoil detection analysis, and micro laser induced breakdown spectroscopy. Finally, the hardness of the prior-β_{Zr} structure was correlated to the local oxygen and hydrogen contents, reaching locally 1-1.2 wt.% and 3000-4000 wt.ppm, respectively.

In a second part, we will illustrate a recent study performed on model materials, homogeneously charged in hydrogen at ~1000-3000 wt.ppm, produced from Zircaloy-4 and M5™ cladding tubes. Samples were also prepared from nearly-pure Zr (Van Arkel process) in order to isolate the effect of hydrogen. The samples were heat-treated at ~1000°C to characterize the properties of the transformed β_{Zr} phase and were then cooled down to RT,

[1] M5 is a registered trademark of AREVA NP in the USA and other countries

rapidly (direct water quenching) and slowly to study the potential effects of cooling scenario. Neutron and X-ray diffraction analyses were realized on these model materials to study their structure as a function of chemical composition of the alloy, hydrogen content and cooling scenario. The results were compared to thermodynamic equilibrium predictions using the Thermo-Calc® software with the Zircobase thermodynamic database. The mechanical properties of the (prior-)β_{Zr} structure were investigated by performing uniaxial tensile tests at various temperature between 700°C and 20°C upon cooling from the β_{Zr} temperature domain, on model materials prepared from Zircaloy-4 tubes containing various hydrogen contents, up to 3000 wt.ppm. Material microstructure and fracture surface of the tested specimens were observed in order to identify the failure mechanisms.



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SECONDARY HYDRIDING *OF Zr-BASED FUEL CLADDINGS AT HIGH TEMPERATURE (LOCA):*

PART 1 - MULTI-SCALE STUDY OF HYDROGEN (AND OXYGEN) DISTRIBUTION

JC. Brachet, D. Hamon, M. Le Saux, et al.
DEN-Service de Recherches Métallurgiques Appliquées
***CEA**, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France*

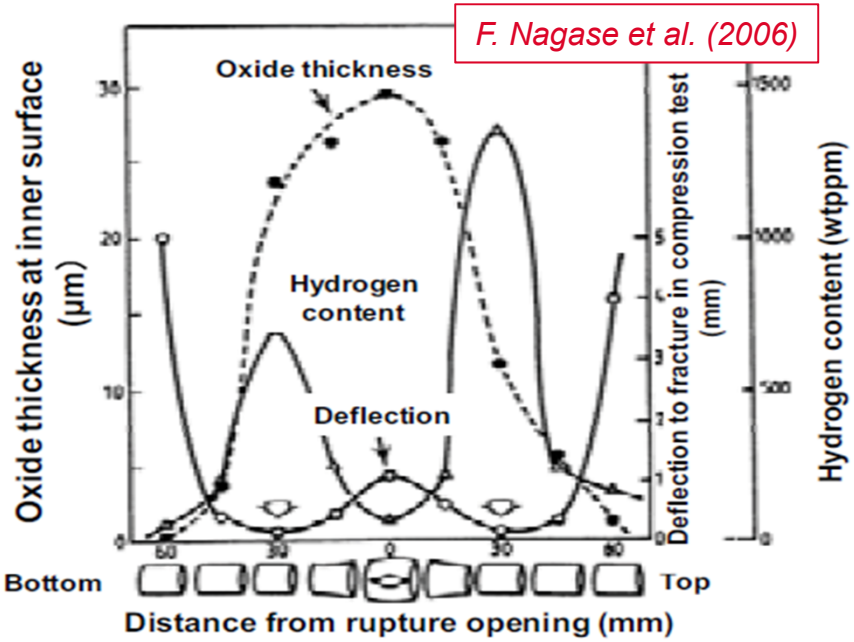
main Refs: JC. Brachet et al. / JNM (2017)
and SCIP-III meeting, Nykoping, Sweden, May 2017

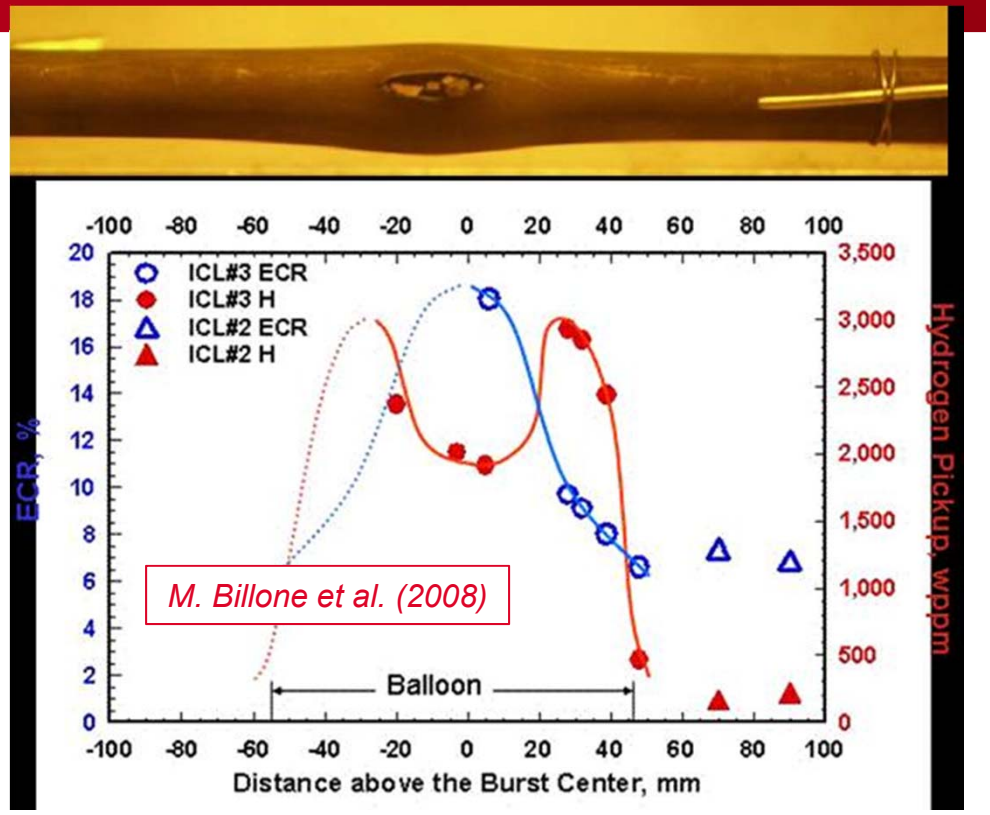
Work performed within the framework of the French CEA-EDF-AREVA Collaborative Program

23RD QUENCH WORKSHOP, OCTOBER 17-19, 2017, KARLSRUHE, GERMANY

INTRODUCTION

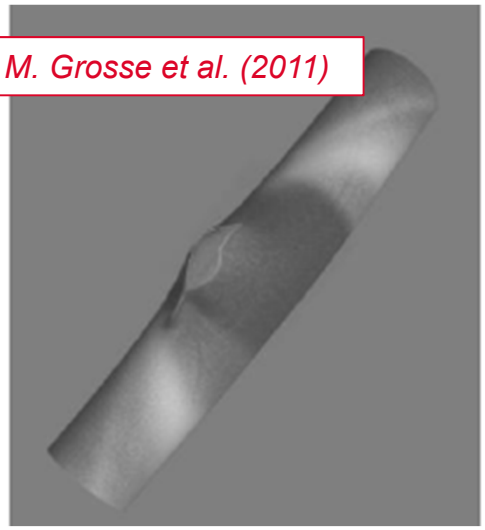
F. Nagase et al. (2006)





M. Billone et al. (2008)

M. Grosse et al. (2011)



- Secondary hydriding ocurring in a few tenths of seconds in outer steam environment at HT, just after burst occurrence

=> due to steam ingress throughout the burst opening and consequently <u>steam starvation conditions</u> inside the gap between the fuel pellet and the inner clad surface

CEA MULTI-SCALE EXPERIMENTAL STUDY OF SECONDARY HYDRIDING:

Objectives and experimental:

- to map quantitatively, at different scales, the distribution of oxygen and hydrogen within M5™ clad segments having experienced ballooning and burst at HT followed by steam oxidation at 1100°C & 1200°C and final direct water quenching down to RT

- then to correlate with the (local) prior-β_{Zr} hardening
(*and further, with the PQ clad mechanical properties*)

⇒ This has been done by coupling information from several advanced analytical tools available at CEA-Saclay (*), that is:
- Neutron Radiography/Tomography (in collab. with Orphée reactor and LLB)
- Electron Probe Micro Analysis (EPMA)
- Micro Elastic Recoil Detection Analysis (μERDA) (in collab. with IRAMIS/LEEL – *nuclear μprobe, which includes a shielded line dedicated to radioactive samples*)
- and Micro Laser Induced Breakdown Spectroscopy (μLIBS) (in collab. with DPC/SEARS)

(*) more details in : JNM (2017) doi: 10.1016/j.jnucmat.2017.03.009

TYPICAL SAMPLE GEOMETRY AND 2-STEPS TESTS PERFORMED

- *M5™ clad segments from Areva-NP have been submitted to 2-steps testing:*

(1) Internal pressure thermal ramp test up to burst occurrence (EDGAR test facility)

(2) Subsequent ~one-sided oxidation at 1100°C for 530s and 1200°C for 160s in flowing steam environment + final direct water quenching down to RT (performed on clad segments with welded end plugs and internal Alumina rod (to simulate the fuel pellets) - DEZIROX facility)

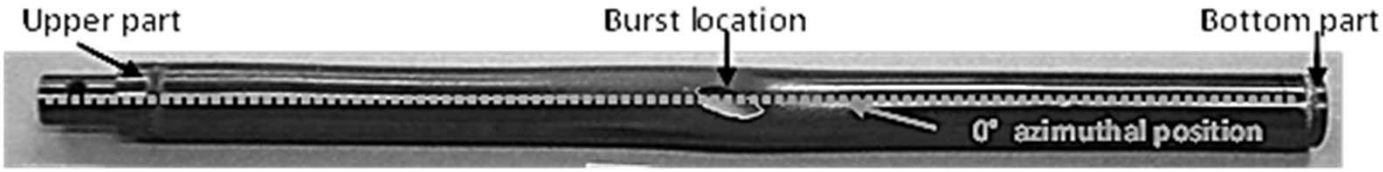
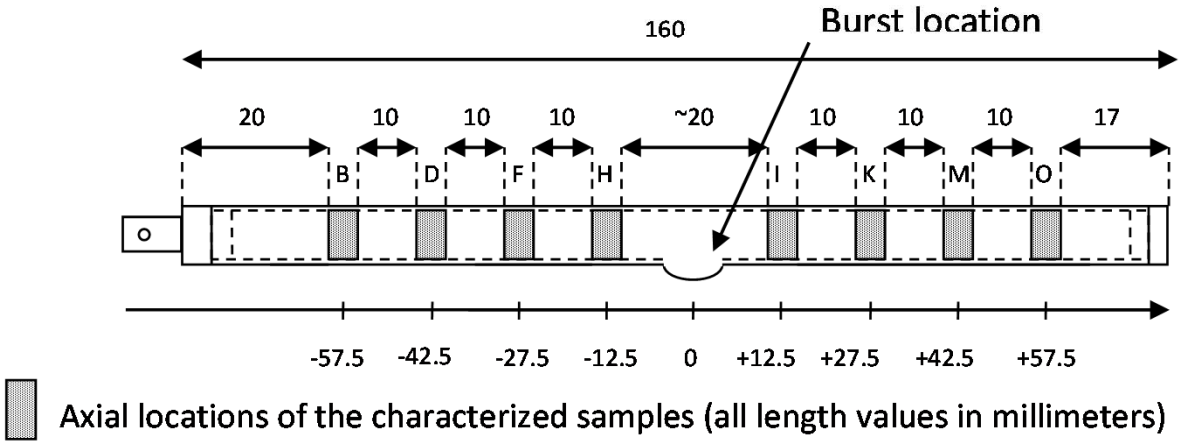


Figure 1 - Macrograph after burst, oxidation and quenching of a 150 mm long clad segment with two end caps and containing an alumina rod



TYPICAL RESULTS FROM NEUTRON RADIOGRAPHY

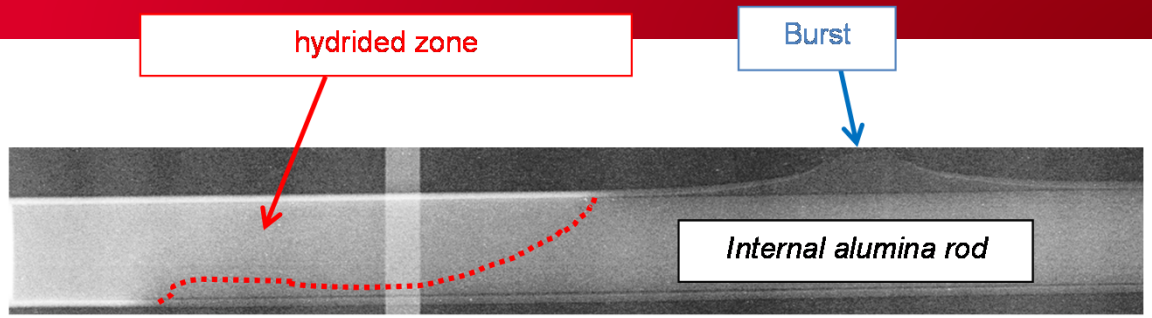
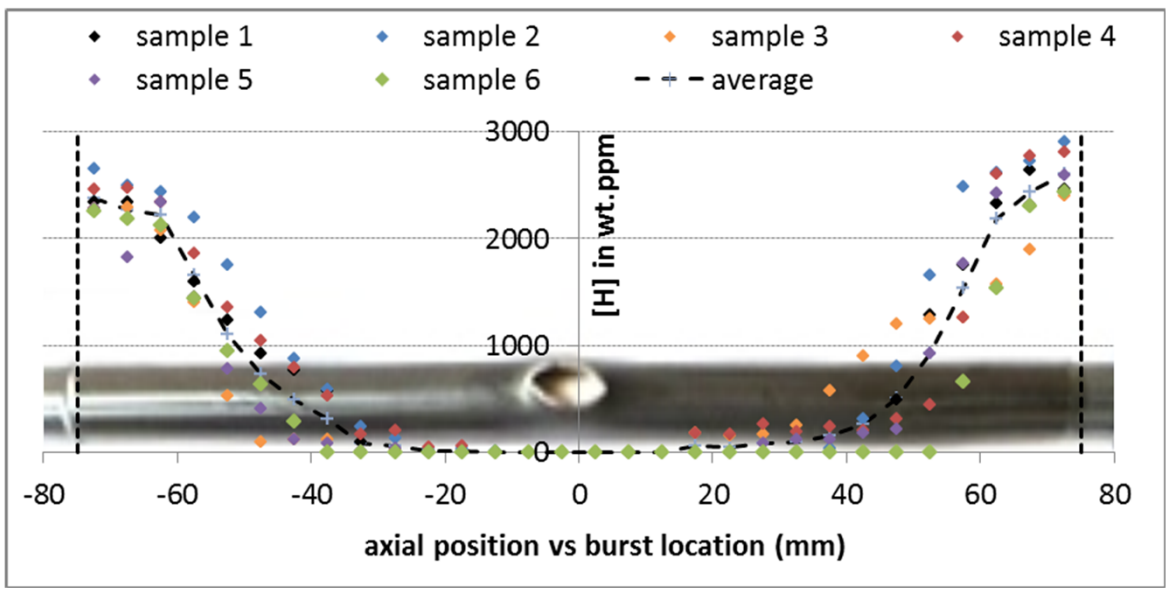


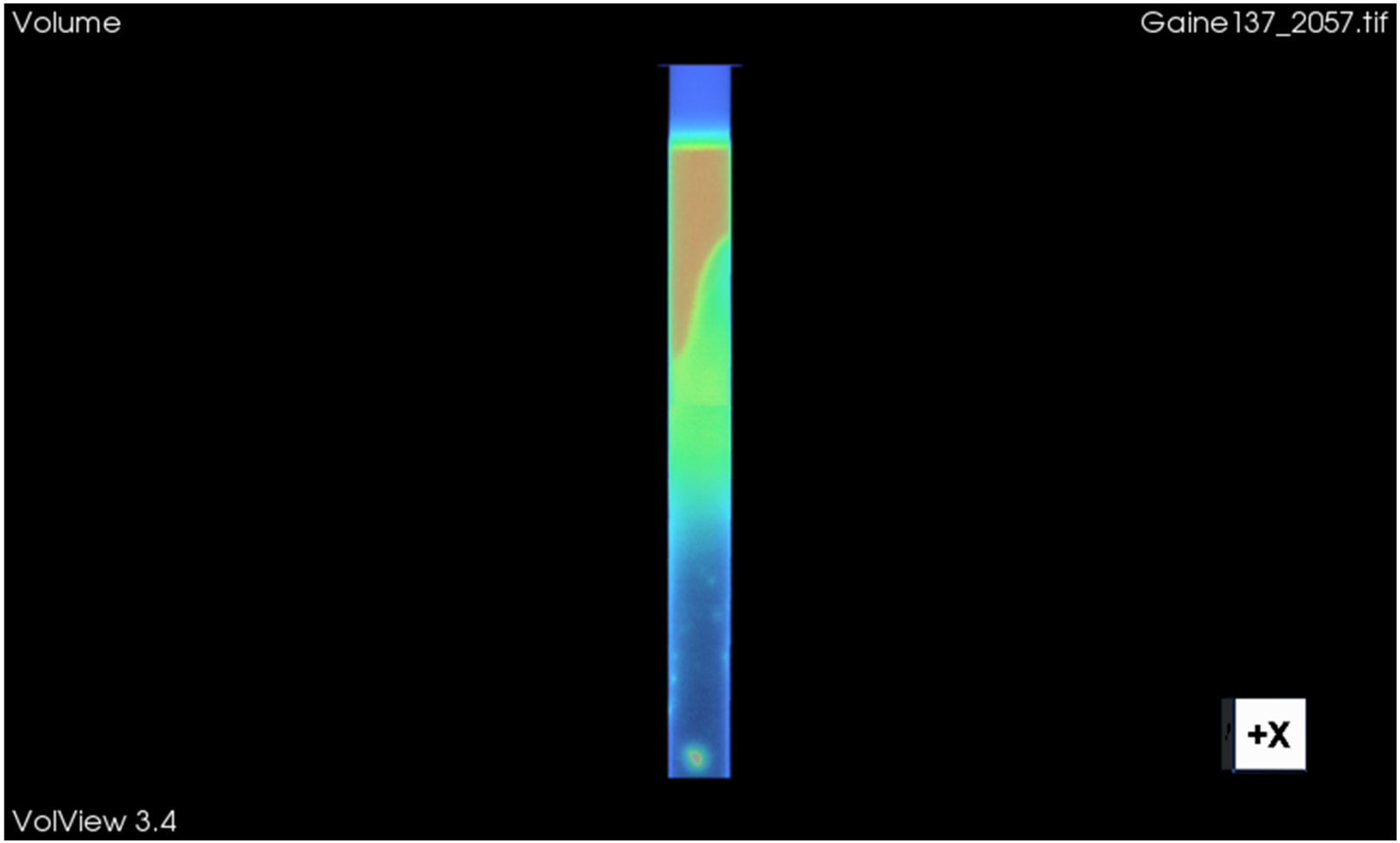
Figure – Typical neutron radiograph of the half of a clad segment having experienced HT secondary hydriding (the lighter the color the higher the hydrogen content)



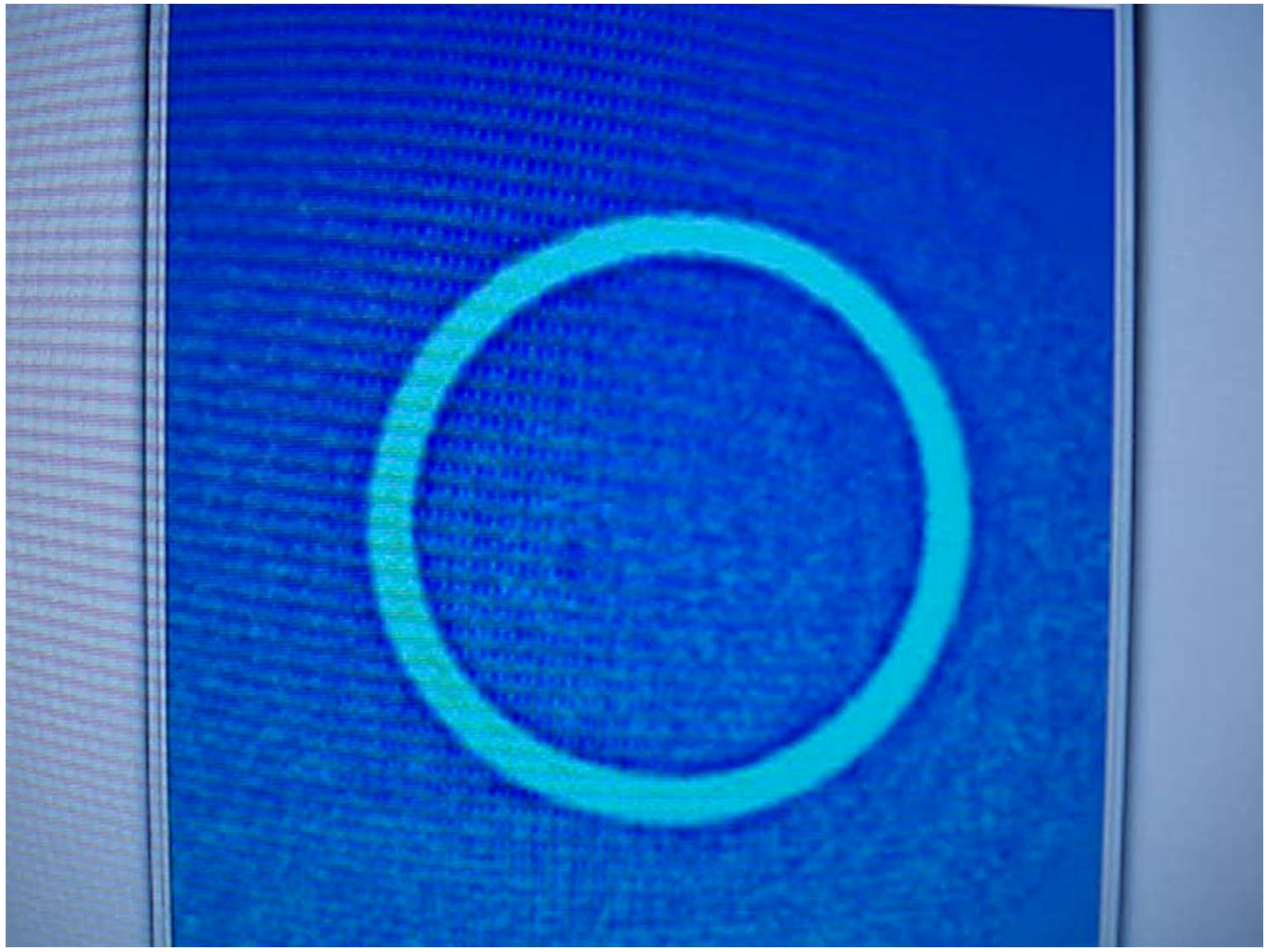
Very reproducible results *(i.e., same locations and amplitudes of the induced secondary hydriding peaks)*

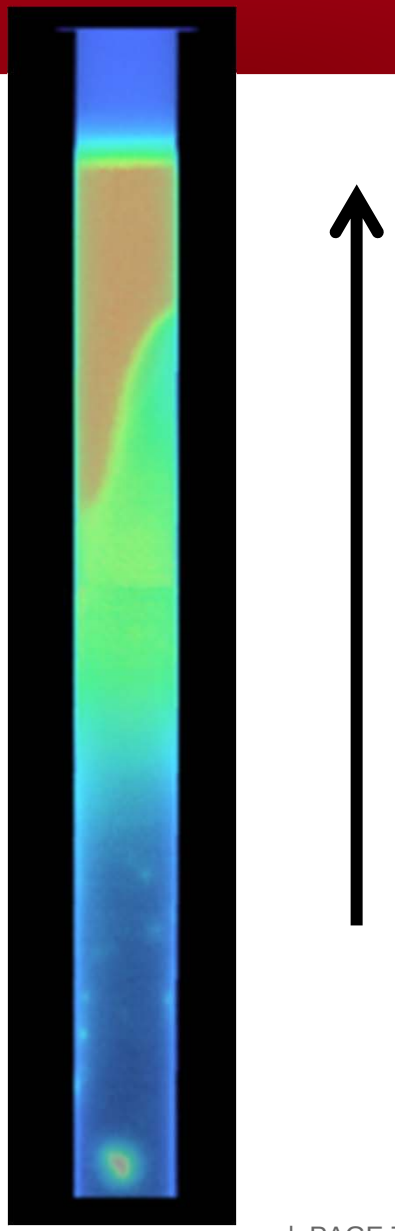
Figure - Circumferentially averaged hydrogen contents as a function of the axial distance from the burst location derived from neutron radiography experiments for six different clad segments that have been oxidized for 160s at 1200°C

TYPICAL RESULTS FROM NEUTRON TOMOGRAPHY (1/3)

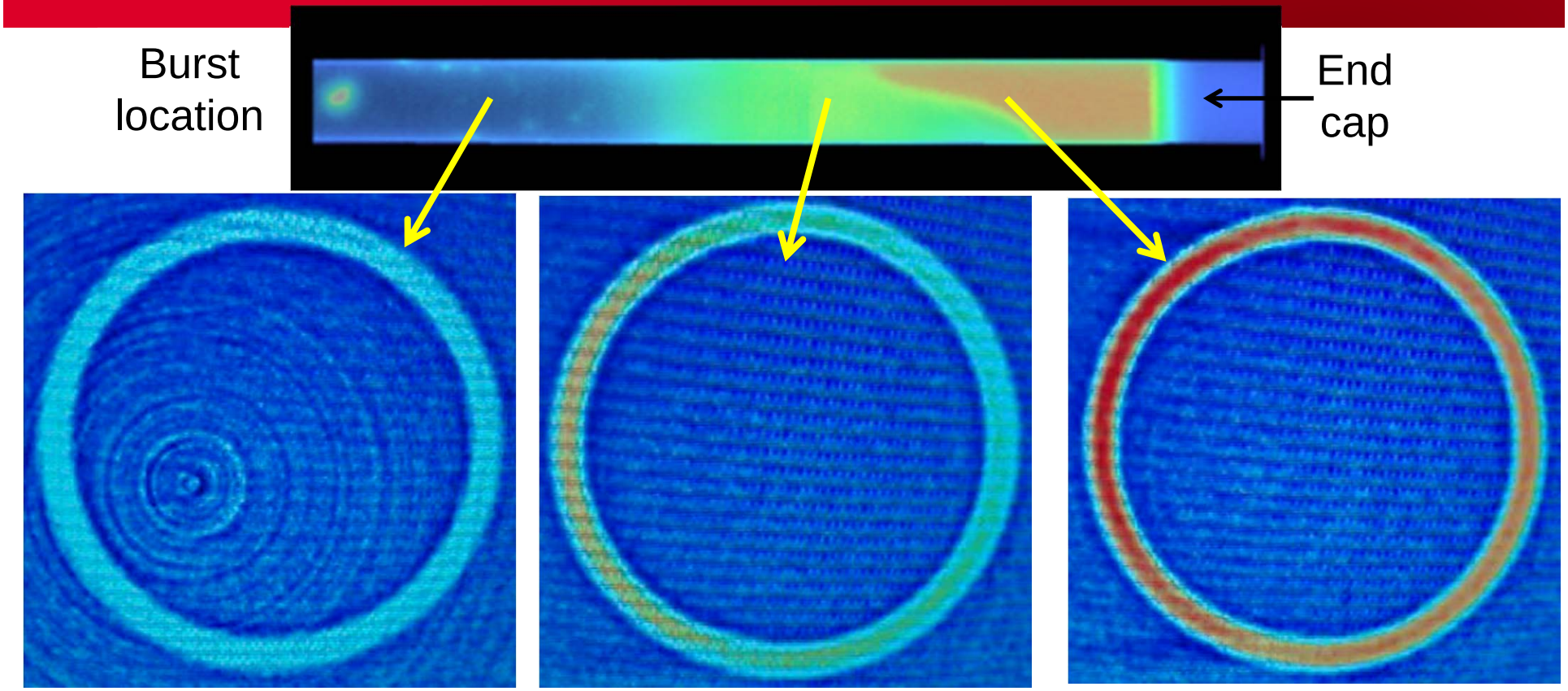


TYPICAL RESULTS FROM NEUTRON TOMOGRAPHY (2/3)





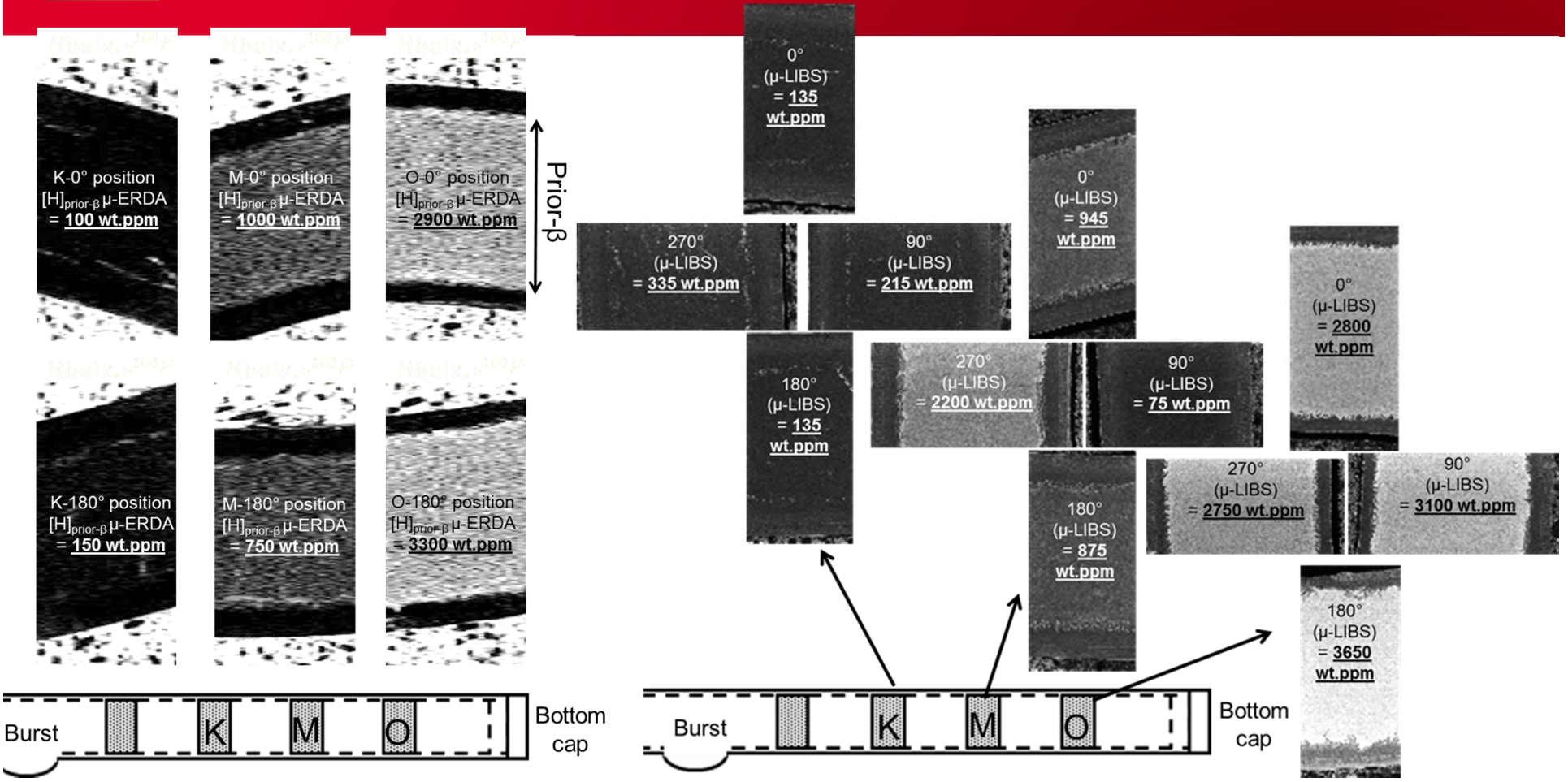
TYPICAL RESULTS FROM NEUTRON TOMOGRAPHY (3/3)



⇒ *consistent with previous work from M. Grosse et al. (KIT) showing (secondary) hydrogen enriched asymmetric bands*

=> Post-LOCA hydrogen measurements using conventional gas hot extraction method give an incomplete view of the H distribution (due to the strong potential circumferential H gradients)

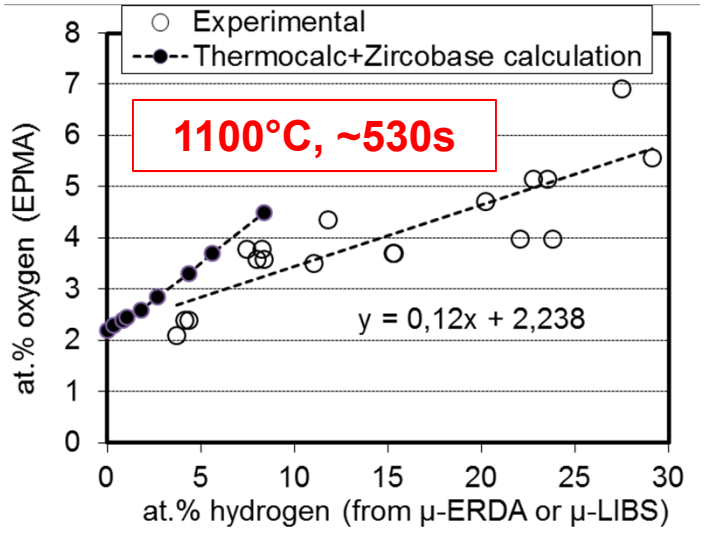
TYPICAL RESULTS FROM µERDA AND µLIBS QUANTITATIVE H MAPPING

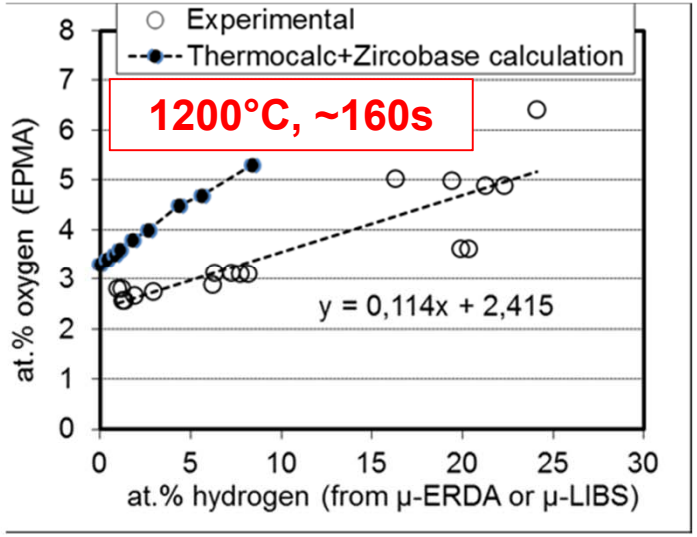


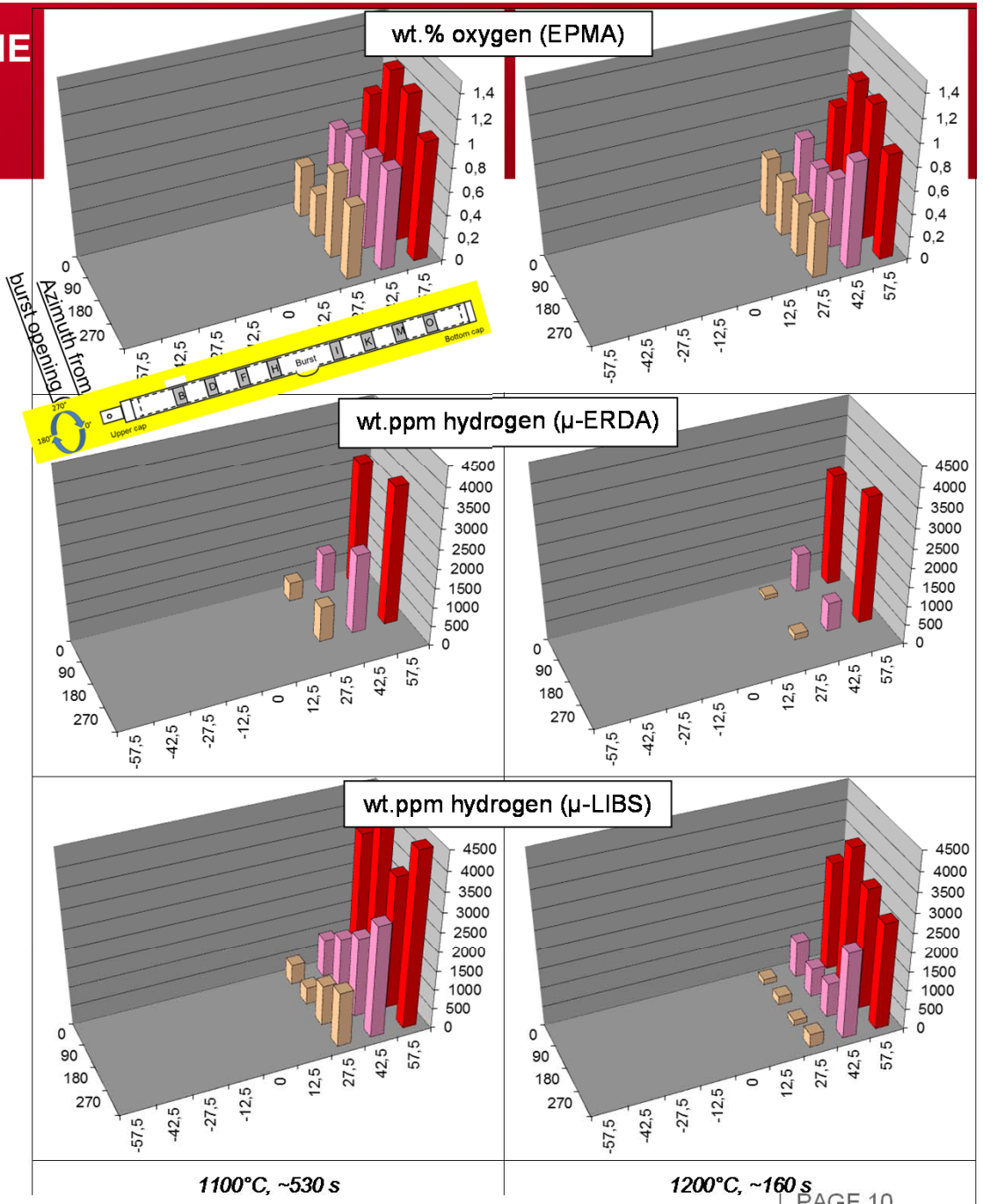
⇒ *Confirmation of strong circumferential H gradients*

⇒ *Most of the hydrogen trapped into the inner prior-β_{Zr} layer*

RELATIONSHIP BETWEEN THE LOCAL PRIOR-β_{Zr} OXYGEN & HYDROGEN CONTENTS

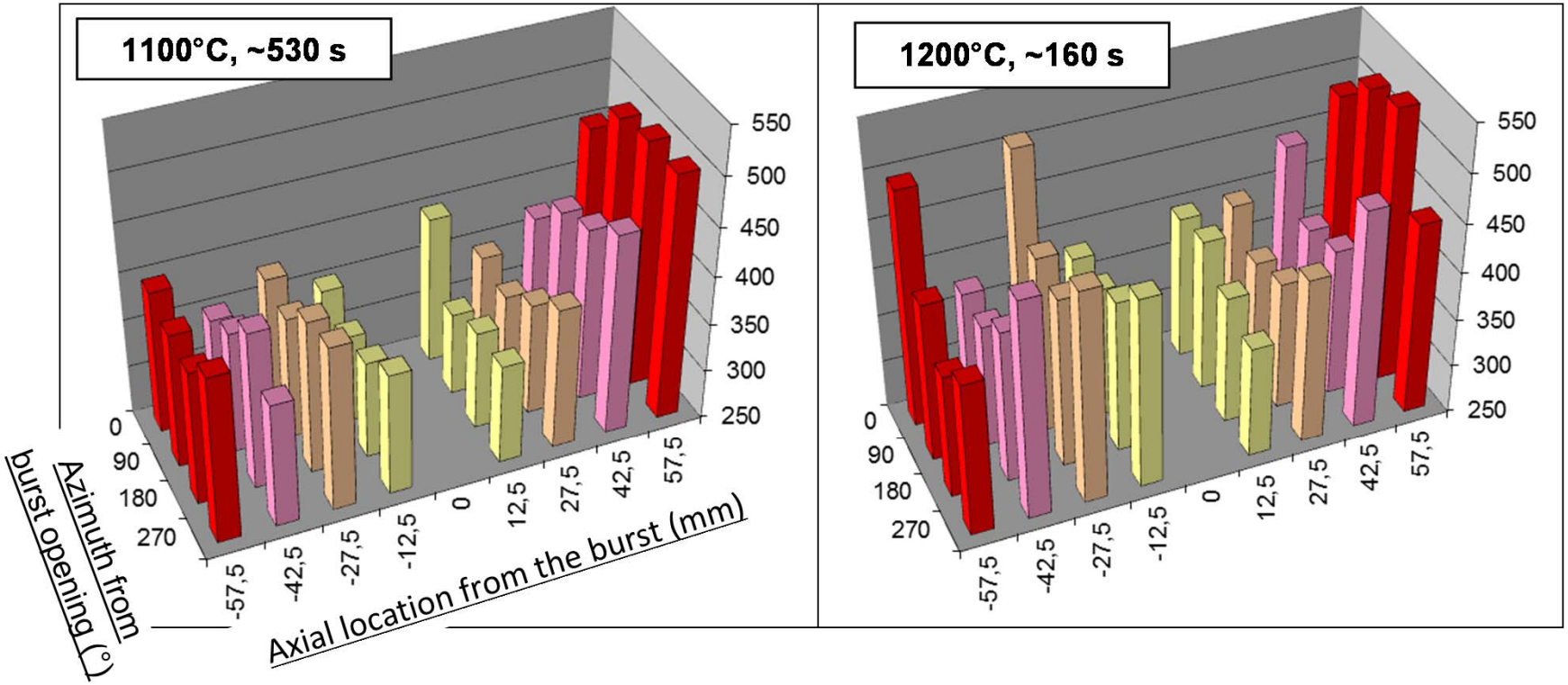






CONSEQUENCES ON THE LOCAL PRIOR-β_{Zr} HARDENING (1/2)



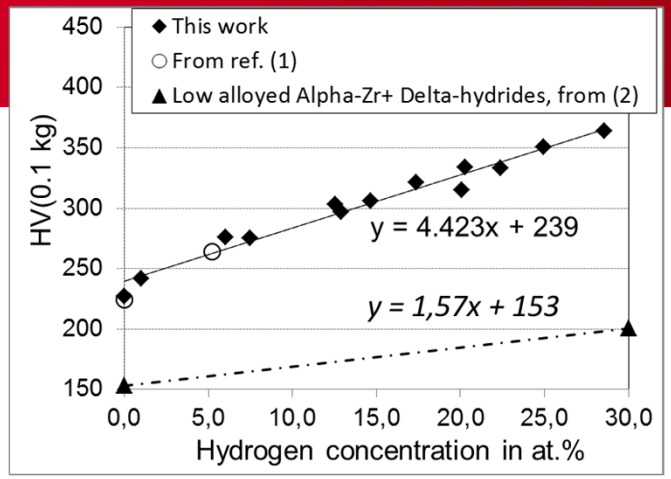


- Post-quenching prior-β micro-hardness (HV(0.1 kg)) measured at different axial and azimuthal locations of tested clad segments

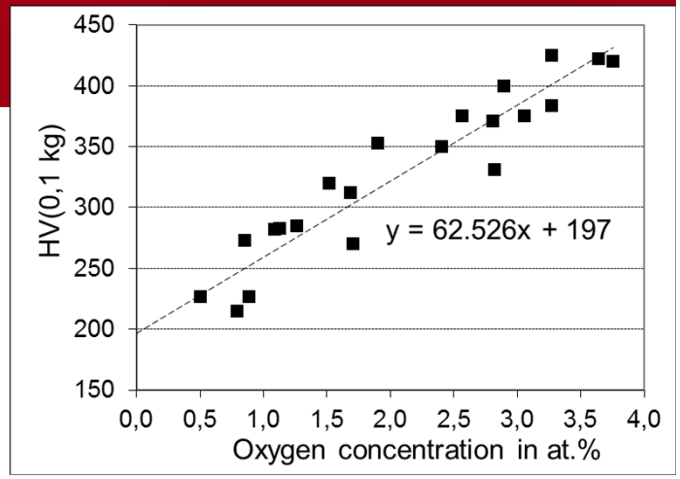
CONSEQUENCES ON THE LOCAL PRIOR-β_{Zr} HARDENING (2/2)

- Prior-β_{Zr} layer (local) hardening mainly due to the (local) oxygen content (only limited hardening due to H itself),

- but indirect effect of H which induces higher prior-β_{Zr} oxygen content (thermodynamic effect: increase of the oxygen solubility and potential impact on oxygen diffusion kinetics)

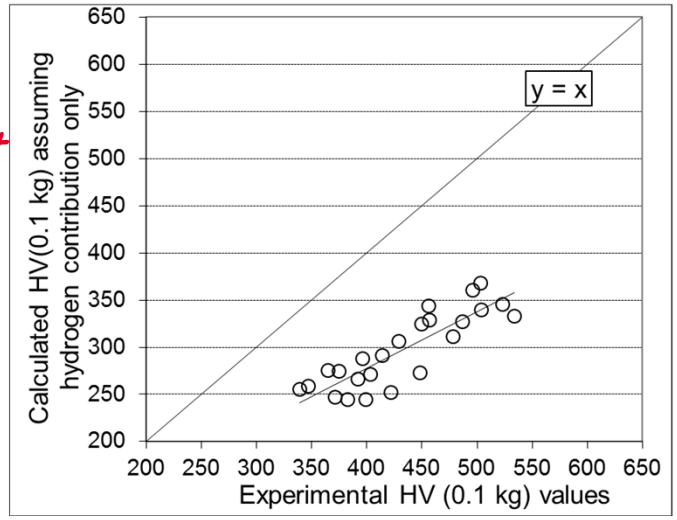


(a)

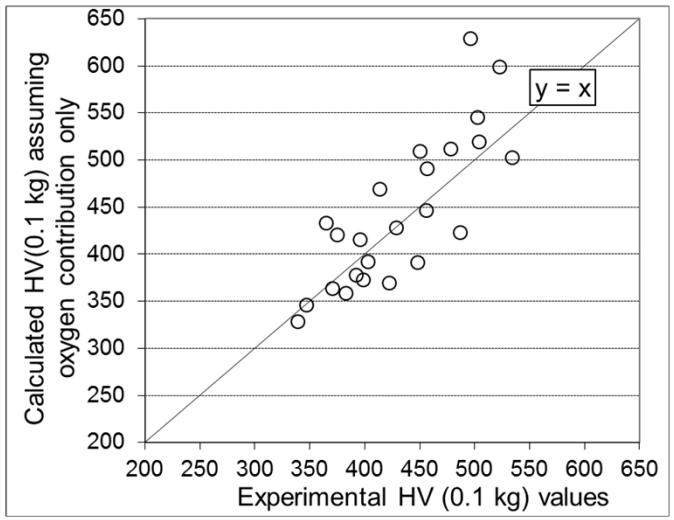


(b)

- Post-quenching prior-β hardening due to the respective contributions of: (a) hydrogen concentration (b) oxygen concentration



(a)



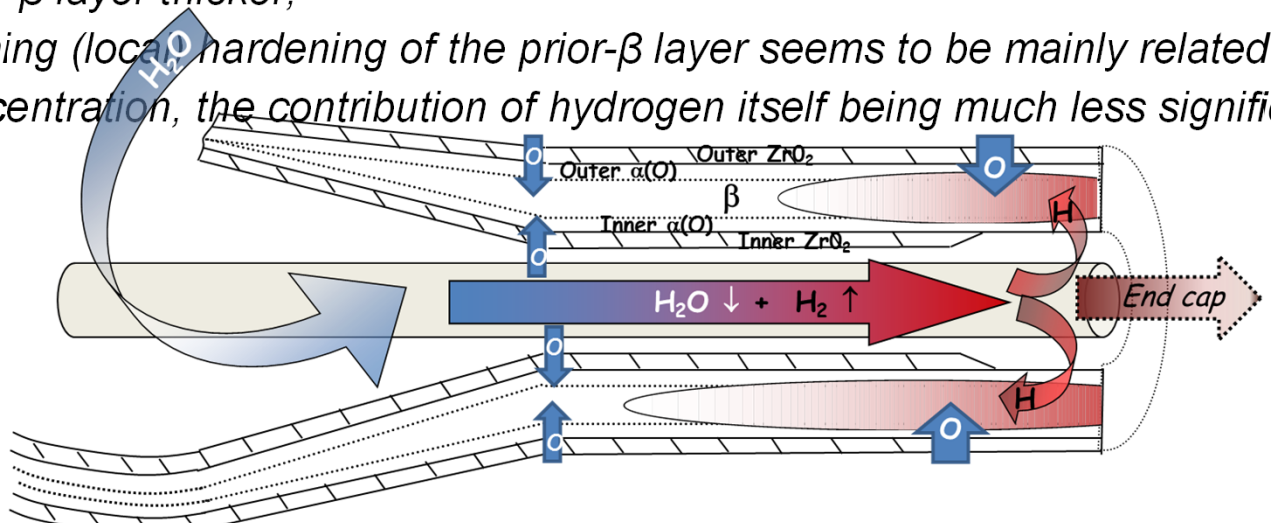
(b)

- Calculated post-quenching micro-hardness of the prior-β phase corresponding to different axial and azimuthal locations of the analyzed clad segments, assuming that: (a) only hydrogen contributes to the measured hardening (b) only oxygen contributes to the measured hardening

SOME CONCLUSIONS FOLLOWING MULTI-SCALE EXPERIMENTAL STUDY OF SECONDARY HYDRIDING UPON LOCA:

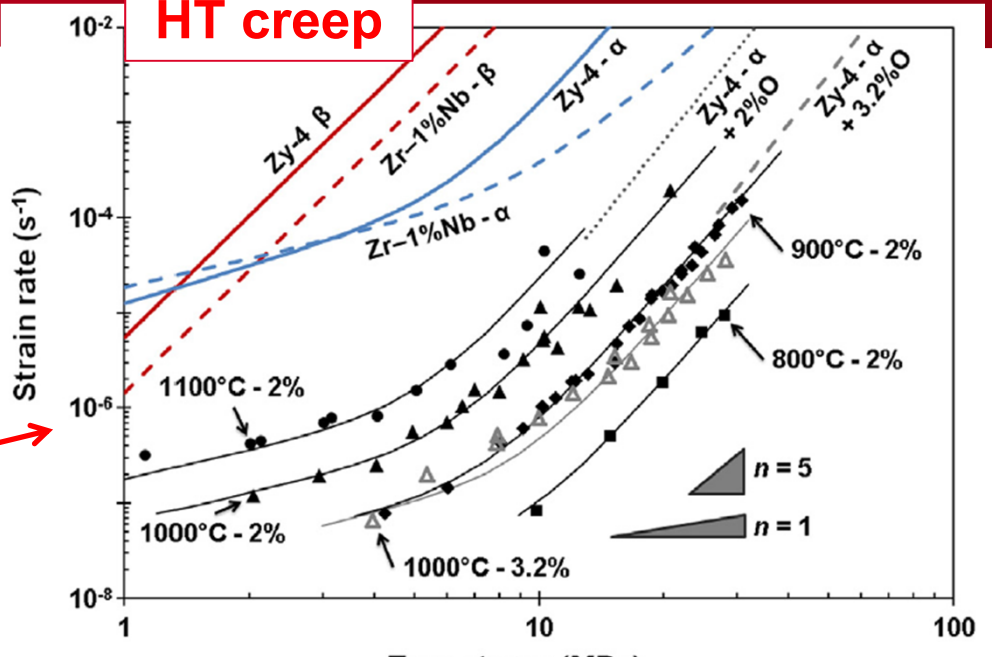
For the specific HT oxidation conditions experienced here, it is shown that:

- *results are very reproducible;*
- *more than 50% of the total gaseous hydrogen quantity coming from the oxidation at HT of the clad inner surface is absorbed by the material and concentrated into the prior-β layer close to the end plugs axial locations (steam starvation conditions);*
- *high axial and azimuthal hydrogen and oxygen concentrations, reaching up to 3000-4000 wt. ppm and 1.0-1.2 wt.%, respectively, measured within the β phase, with strong axial and azimuthal heterogeneities (conventional hot gas extraction analysis gives only a partial view…);*
- *consistent with thermodynamic and kinetics considerations oxygen diffusion into the prior-β layer is enhanced in the regions highly enriched in hydrogen, where the α(O) phase layer is thinner and the prior-β layer thicker;*
- *post-quenching (local) hardening of the prior-β layer seems to be mainly related to the (local) oxygen concentration, the contribution of hydrogen itself being much less significant.*

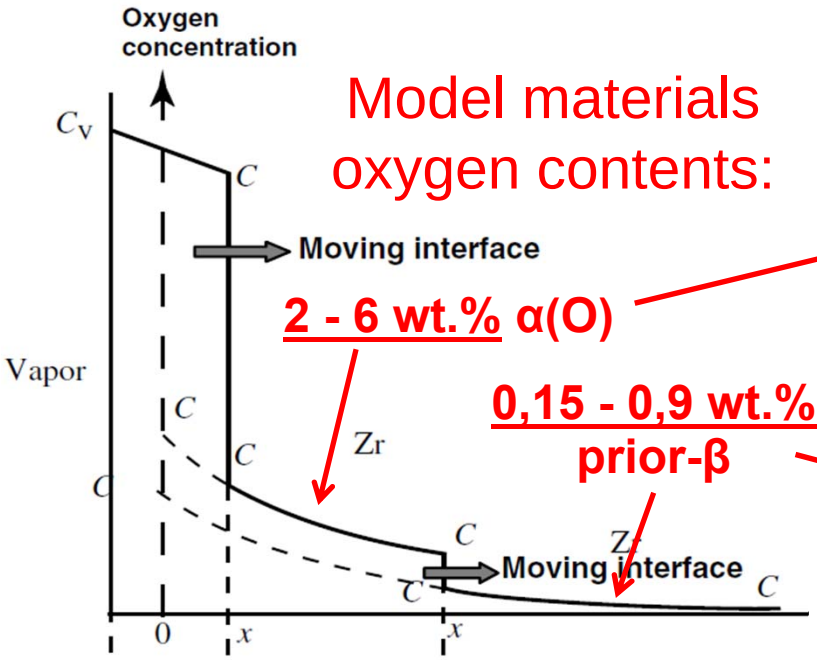


MODEL ALLOYS - HT & PQ MECHANICAL BEHAVIOR OF OXYGEN-ENRICHED MATERIALS 1/2

HT creep



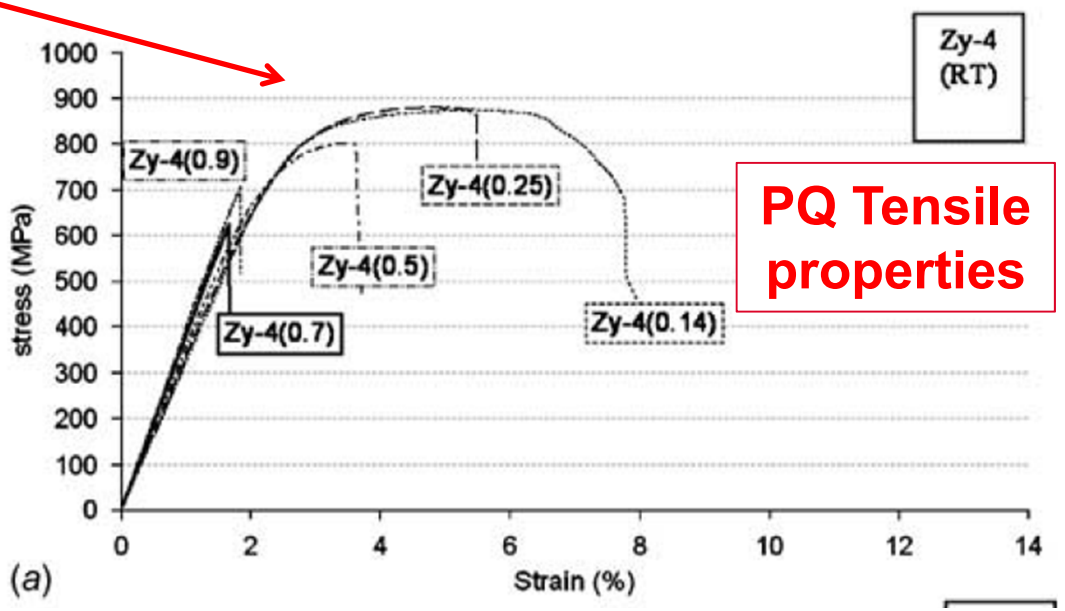
Model materials oxygen contents:



2 - 6 wt.% α(O)

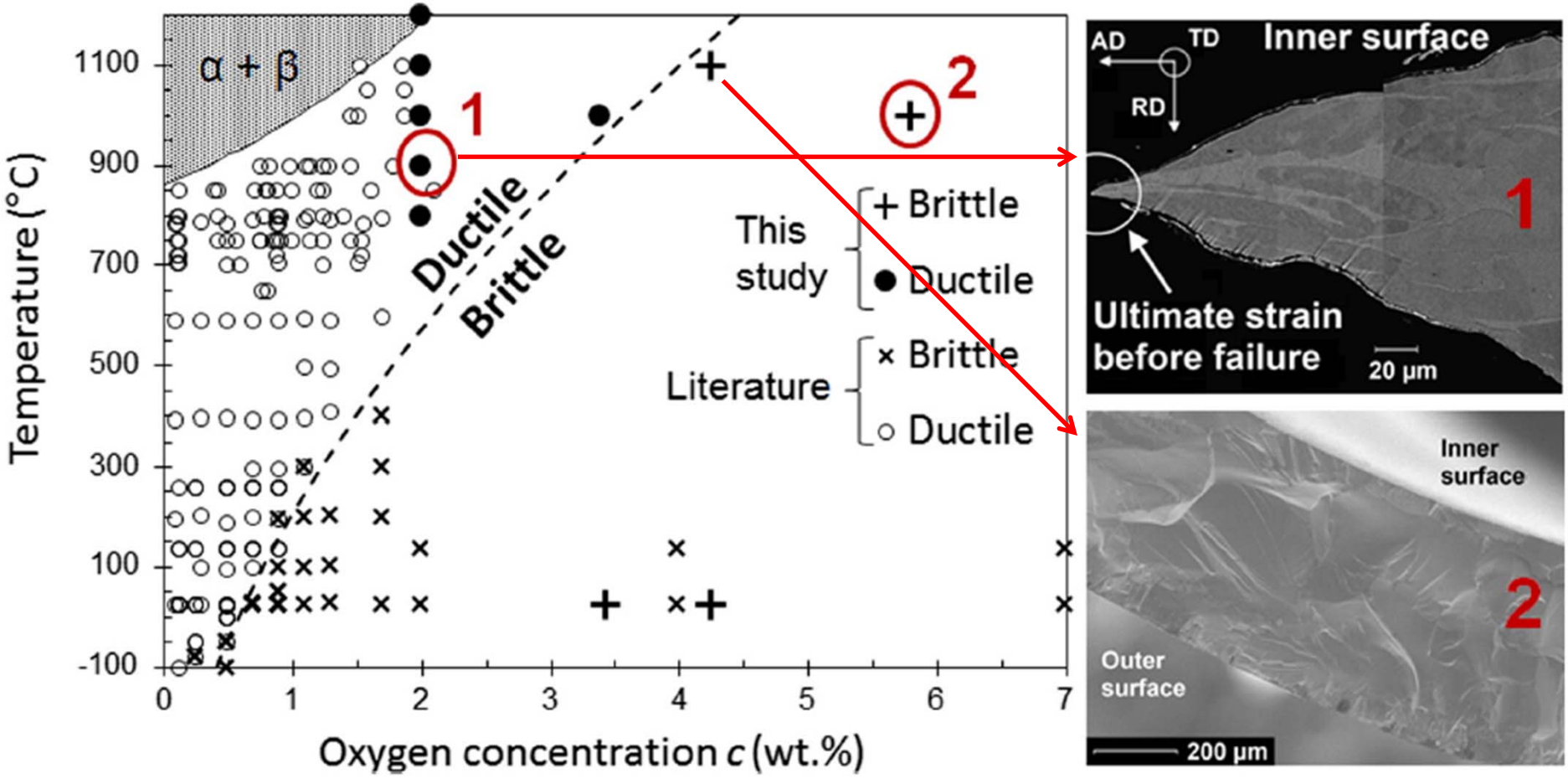
0,15 - 0,9 wt.% prior-β

Ref.: Stern et al. (2008), Chosson et al. (2016),



PQ Tensile properties

« Ductility map » : Ductile-to-Brittle transition Temperature vs. Oxygen content:



Ref.: Chosson et al. (2016)

ON GOING – MODEL ALLOYS - HT MECHANICAL BEHAVIOR OF HIGHLY HYDROGEN-ENRICHED MATERIALS (typical of secondary hydriding)

See next presentation
(Thai LE HONG)

Thank you for your attention

Commissariat à l'énergie atomique et aux énergies alternatives
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Direction de l'Energie Nucléaire
Département des Matériaux pour le Nucléaire
Service de Recherches Métallurgiques Appliquées

Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019

Back-up slides

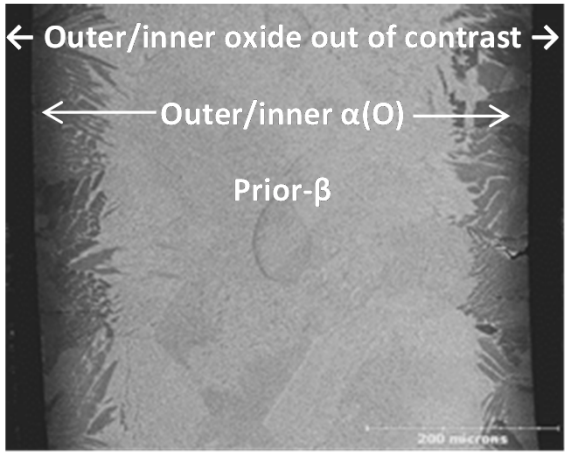
Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Saclay | 91191 Gif-sur-Yvette Cedex
T. +33 (0)1 69 08 12 28 | F. +33 (0)1 69 08 71 67

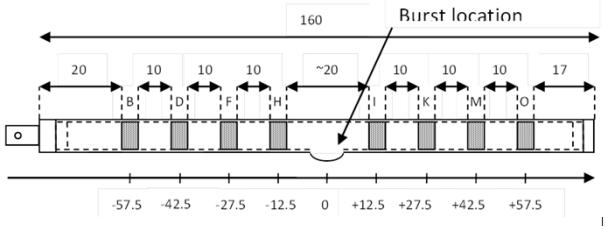
Direction de l'Energie Nucléaire
Département des Matériaux pour le Nucléaire
Service de Recherches Métallurgiques Appliquées

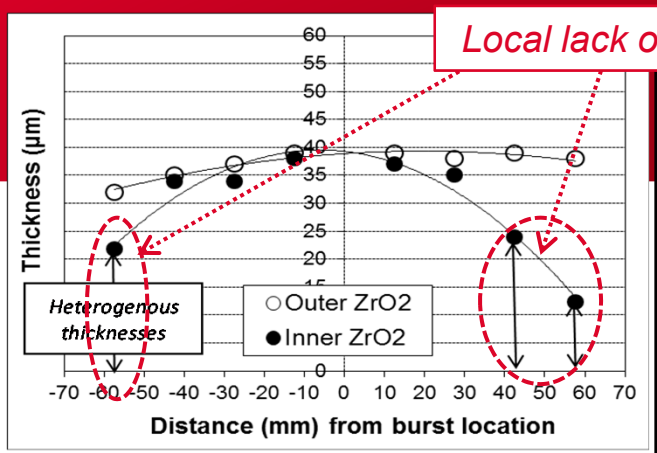
Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019

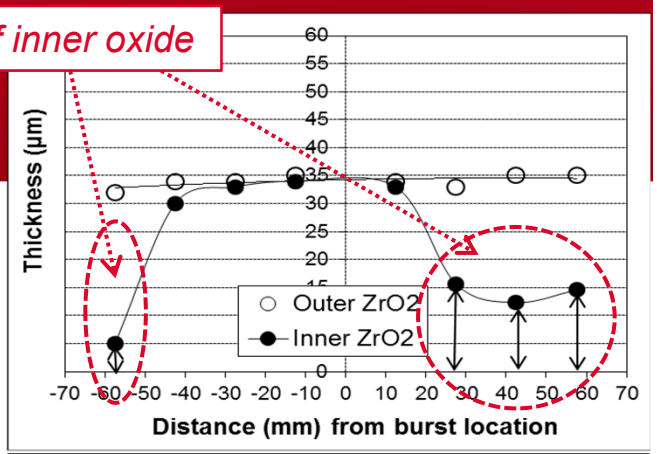
TYPICAL μSTRUCTURE

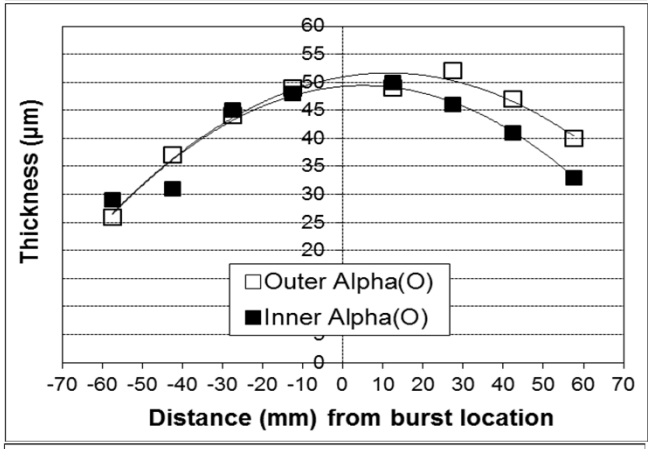
=> Accurate image analysis measurements of the respective outer/inner ZrO_2, $a_{Zr}(O)$ and prior-β_{Zr} thicknesses as a function of clad axial location:

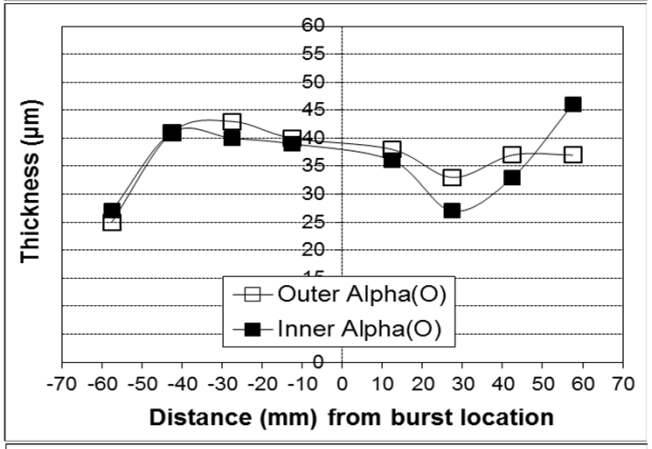


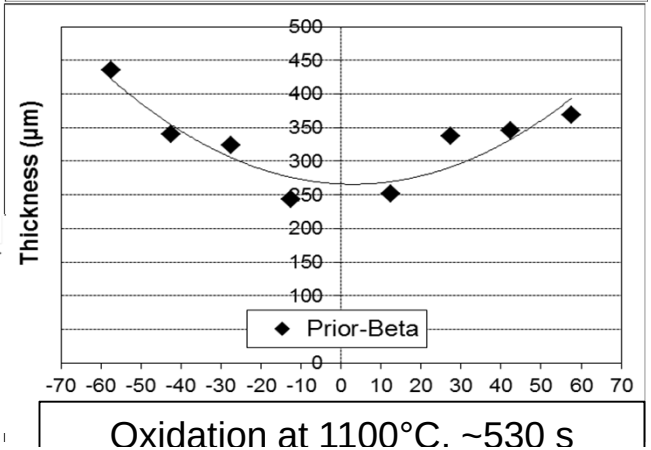




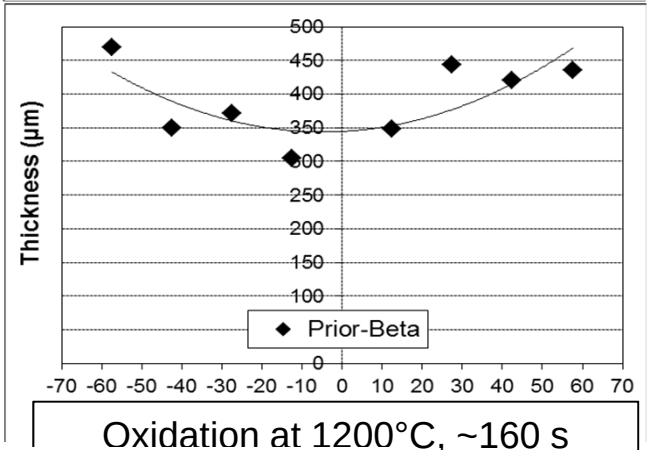






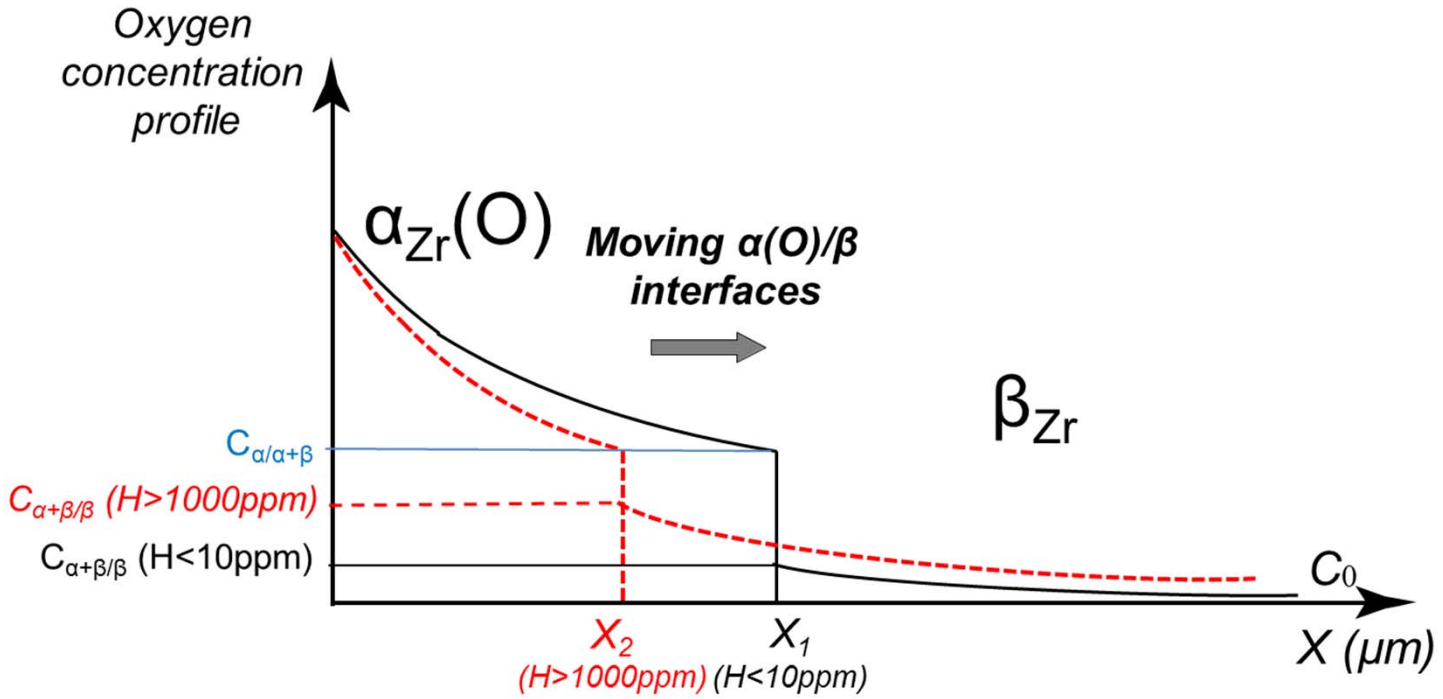


Oxidation at 1100°C. ~530 s



Oxidation at 1200°C, ~160 s

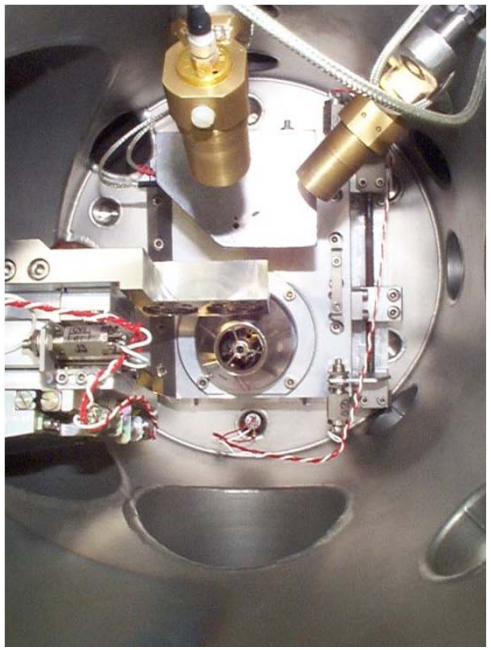
EFFECT OF HIGH H CONTENT (TYPICAL OF SECONDARY HYDRIDING PEAKS) ON THE RESPECTIVE ZrO_2, $\alpha_{Zr}(O)$ AND PRIOR-β_{Zr} (LOCAL) THICKNESSES

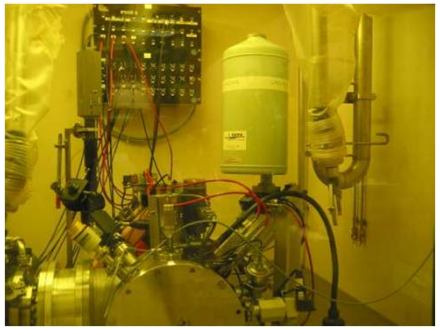


- Schematic illustration of the effect of hydrogen on the relative decrease of the α(O) layer thickness due to an increase of the oxygen solubility in the β phase (for the same oxidation time and temperature)

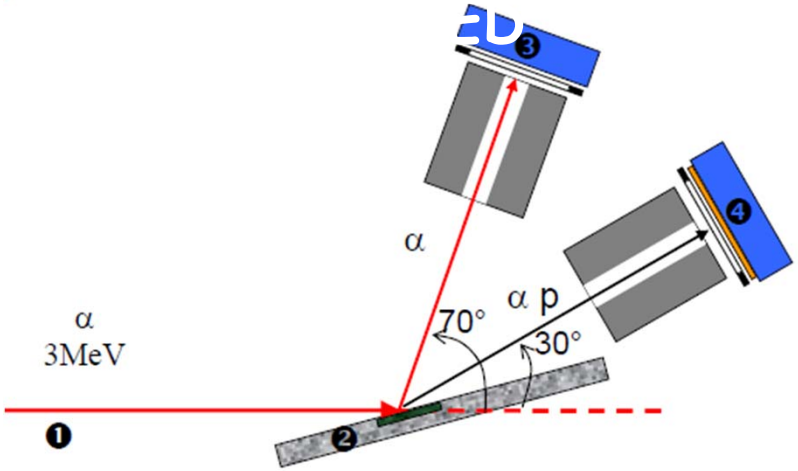
Back-up slides

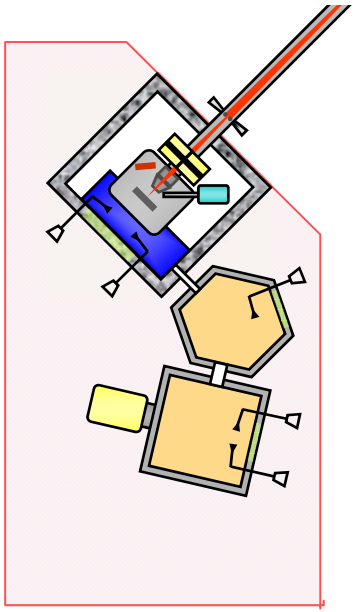
- Chambre d'Analyse de Surfaces et d'Interfaces de Matériaux IRradiants
- Voie à 45° de la microsonde
- Située en zone chaude du LPS
- Manipulation des échantillons en cellules blindées, par télémanipulateur
- Composé de
  - la cellule d'accueil
  - la cellule de transfert
  - la casemate d'analyse







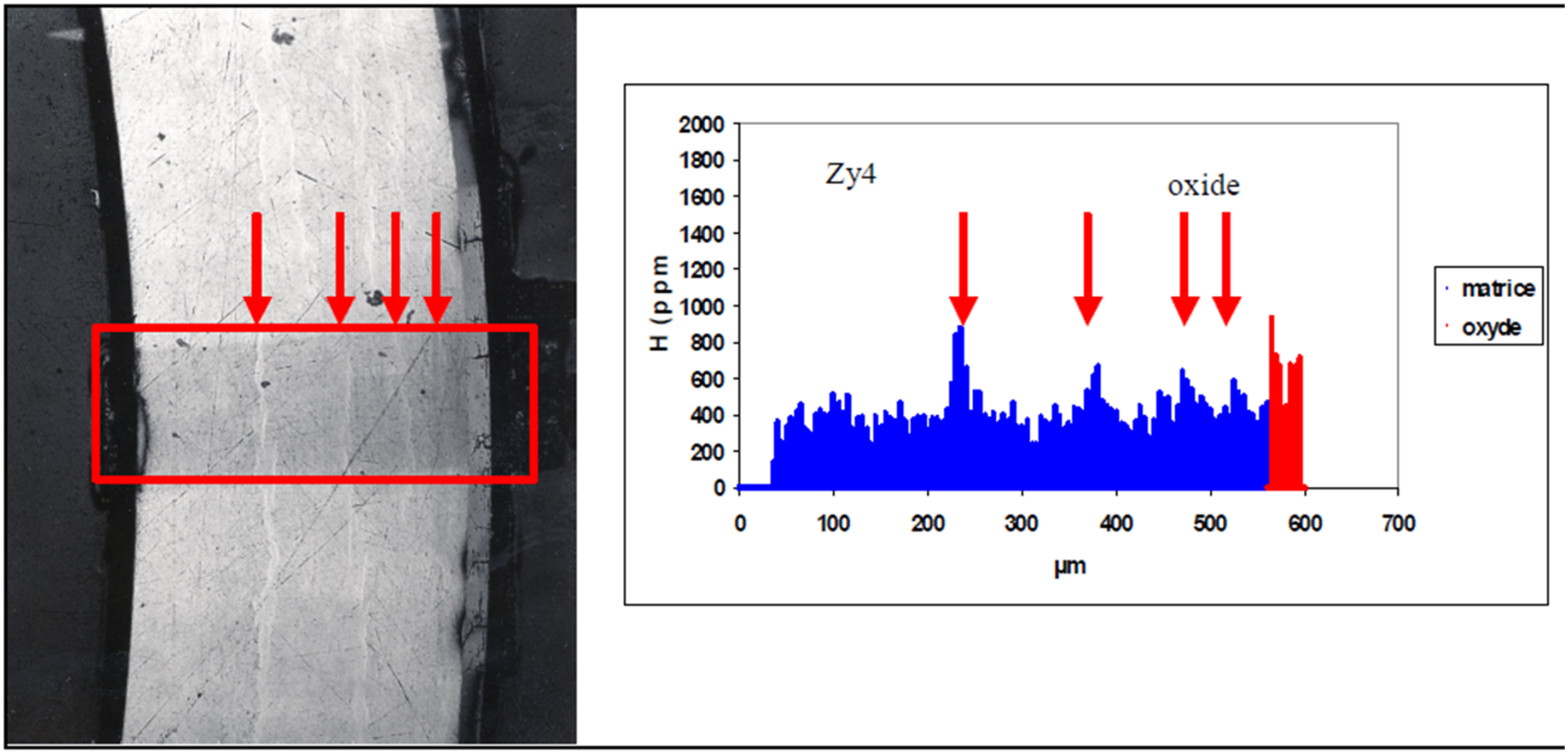




Zone Chaude
(TCPF radioactive)

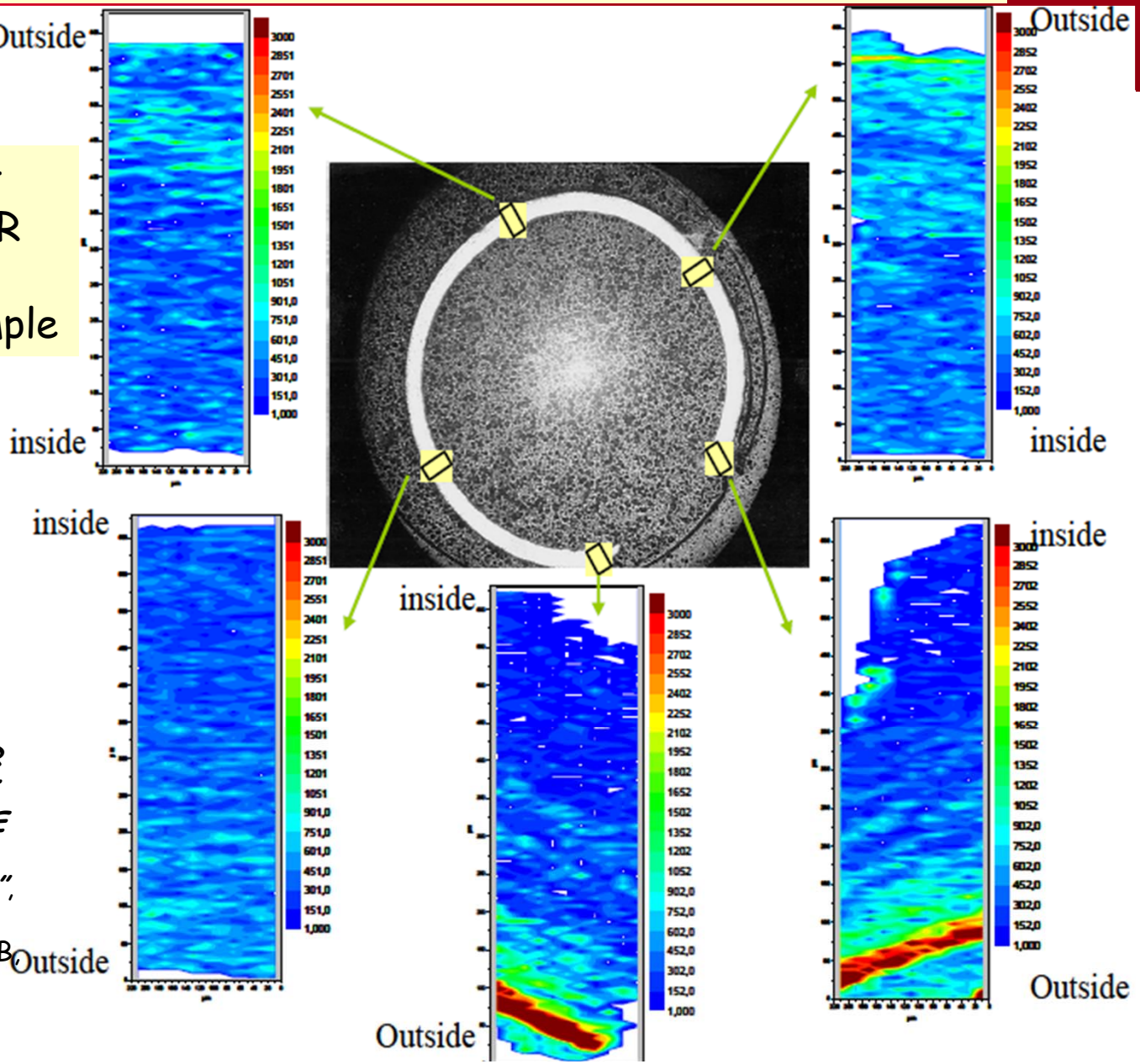


Figure 3: Metallographic observation of the hydride distribution on Zircaloy-4 irradiated for 5 PWR cycles, and corresponding hydrogen profile, obtained by ERDA



From: C. RAEPSAET, et al. ***"QUANTIFICATION AND LOCAL DISTRIBUTION OF HYDROGEN WITHIN ZIRCALOY-4 PWR NUCLEAR FUEL CLADDING TUBES AT THE NUCLEAR MICROPROBE OF THE PIERRE SÜE LABORATORY FROM µERDA"***, Journal of Nuclear Inst. and Methods in Physics Research, B, NIMB55555, (Mai 2008)

Hydrogen content mapping on a 5 PWR cycles irradiated Zircaloy-4 burst sample



From: C. RAEPSAET, et al. ***"QUANTIFICATION AND LOCAL DISTRIBUTION OF HYDROGEN WITHIN ZIRCALOY-4 PWR NUCLEAR FUEL CLADDING TUBES AT THE NUCLEAR MICROPROBE OF THE PIERRE SÜE LABORATORY FROM µERDA"***, Journal of Nuclear Inst. and Methods in Physics Research, B NIMB55555, (Mai 2008)

Title: Secondary hydriding of zirconium-based fuel claddings at high temperature (LOCA conditions)
Part 1: Multi-scale study of hydrogen distribution
Part 2: Effect of high hydrogen contents on metallurgical and mechanical properties

Thai LE HONG[1,2], Matthieu LE SAUX[1], Jean-Christophe BRACHET[1], Jérôme CREPIN[2] *et al.*

[1] DEN–Service de Recherches Métallurgiques Appliquées (SRMA), CEA, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France

[2] MINES ParisTech, PSL Research University, Centre des Matériaux, CNRS UMR 7633, BP 87, 91003 Evry, France

Abstract

During oxidization in steam at High Temperature (HT) under hypothetical Loss-Of-Coolant Accident (LOCA) conditions, zirconium alloy fuel cladding tubes can absorb, in some conditions (*e.g.* after burst occurrence and during subsequent HT steam oxidation), an important amount of hydrogen, up to thousands of wt.ppm locally. The behavior of the cladding can be affected by this secondary hydriding since hydrogen is known to modify metallurgical and mechanical properties of zirconium alloys. The purpose of this study is to refine the description of secondary hydriding and to improve both experimental database and understanding of the effect of high hydrogen contents up to 3000 wt.ppm, on metallurgical and mechanical properties of zirconium alloys, during and after cooling/quenching from high temperatures (β_{Zr} domain).

In a first part, we will illustrate a recent multi-scale analysis of hydrogen partitioning within Zr-based claddings having experienced secondary hydriding at HT. The objective was to map quantitatively, at different scales, the spatial distribution of hydrogen (and oxygen) within M5™[1] clad segments having experienced ballooning and burst at HT followed by steam oxidation at 1100 and 1200°C, and final direct water quenching down to Room Temperature (RT). This was done by coupling information from several advanced analytical techniques available at CEA Saclay: neutron radiography/tomography, electron probe micro analysis, micro elastic recoil detection analysis, and micro laser induced breakdown spectroscopy. Finally, the hardness of the prior-β_{Zr} structure was correlated to the local oxygen and hydrogen contents, reaching locally 1-1.2 wt.% and 3000-4000 wt.ppm, respectively.

In a second part, we will illustrate a recent study performed on model materials, homogeneously charged in hydrogen at ~1000-3000 wt.ppm, produced from Zircaloy-4 and M5™ cladding tubes. Samples were also prepared from nearly-pure Zr (Van Arkel process) in order to isolate the effect of hydrogen. The samples were heat-treated at ~1000°C to characterize the properties of the transformed β_{Zr} phase and were then cooled down to RT,

[1] M5 is a registered trademark of AREVA NP in the USA and other countries

rapidly (direct water quenching) and slowly to study the potential effects of cooling scenario. Neutron and X-ray diffraction analyses were realized on these model materials to study their structure as a function of chemical composition of the alloy, hydrogen content and cooling scenario. The results were compared to thermodynamic equilibrium predictions using the Thermo-Calc® software with the Zircobase thermodynamic database. The mechanical properties of the (prior-)β_{Zr} structure were investigated by performing uniaxial tensile tests at various temperature between 700°C and 20°C upon cooling from the β_{Zr} temperature domain, on model materials prepared from Zircaloy-4 tubes containing various hydrogen contents, up to 3000 wt.ppm. Material microstructure and fracture surface of the tested specimens were observed in order to identify the failure mechanisms.


KIT
Karlsruher Institut für Technologie

DE LA RECHERCHE À L'INDUSTRIE

23rd QUENCH workshop, October 17-19, 2017 Karlsruhe, Germany

SECONDARY HYDRIDING OF ZIRCONIUM-BASED FUEL CLADDINGS AT HIGH TEMPERATURE (LOCA CONDITIONS)

PART 2: EFFECT OF HIGH HYDROGEN CONTENTS ON METALLURGICAL AND MECHANICAL PROPERTIES

Thai LE HONG [1,2], Matthieu LE SAUX[1], Jean-Christophe BRACHET[1], Isabelle TURQUE[1,2,3], Jérôme CREPIN[2]

[1] DEN–Service de Recherches Métallurgiques Appliquées (SRMA), CEA, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France

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Corresponding author, email: hong-thai.le@cea.fr, hong-thai.le@mines-paristech.fr

with financial contributions from

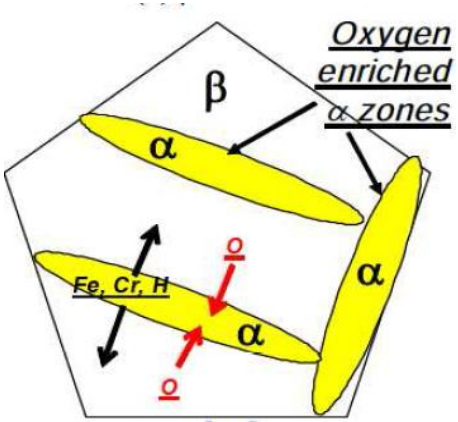
www.cea.fr

INTRODUCTION

- Scientific context of this work and motivation/challenges have been presented by J-C. Brachet in "Secondary hydriding of zirconium based fuel claddings at high temperature (LOCA conditions). Part 1: Multi-scale study of hydrogen distribution"

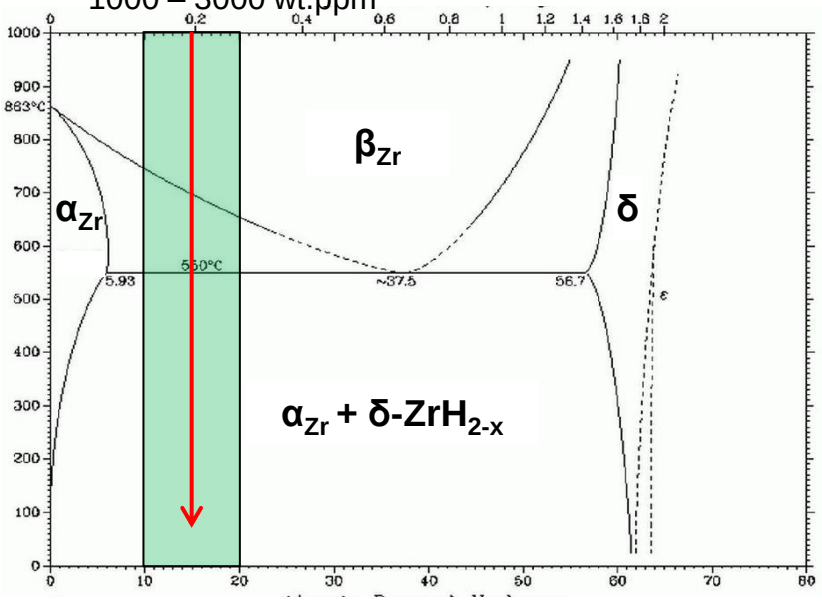
OBJECTIVES OF THIS PHD WORK

- Study the **coupled effects** of **oxygen** (0,14-0,9wt.%) and **high hydrogen contents** (1000-3000 wt.ppm) on the **metallurgical and mechanical properties** of fuel claddings in zirconium based alloys, **Zircaloy-4 and M5**, during and after **cooling/quenching from high temperature (β_{Zr} domain)**, *i.e.* LOCA conditions



Heterogeneous "micro-composite" prior-β_{Zr} microstructure

1000 – 3000 wt.ppm



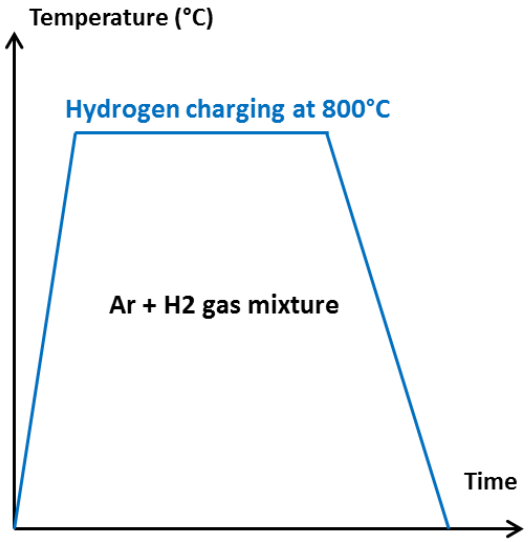
10 – 20 at.%

Zuzek et al., 1990

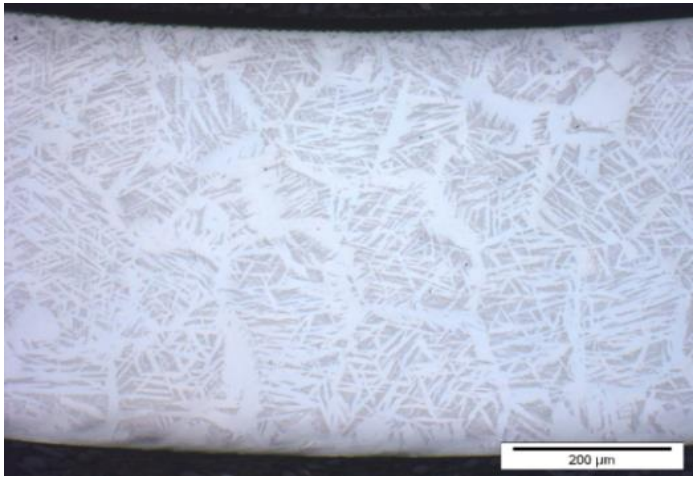
This presentation: Effects of high hydrogen contents (up to 3000 wt.ppm)

Model materials **homogeneously** charged in hydrogen at **1000-3000 wt.ppm** under **Ar + H2 at 800°C**, prepared from **Zircaloy-4 and M5™* fuel cladding tubes** (6cm long)

- **Heat-treatment** in the **β_{Zr} phase temperature** domain (1000°C) to simulate β transformation
- Cooling down to RT, **water quenching or slow cooling**, for studying the **effect of cooling scenario**



Material	%Sn	%Fe	%Cr	%O	%Nb	%Zr
Zy-4	1,3	0,22	0,12	0,13	-	Bal.
M5™	-	0,04	-	0,14	1	Bal.

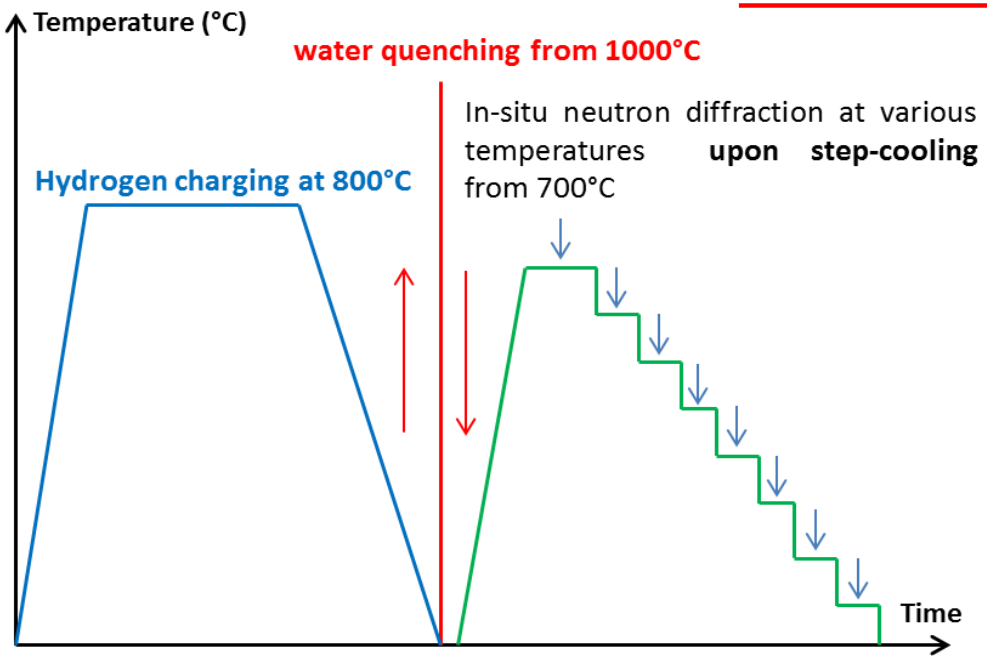


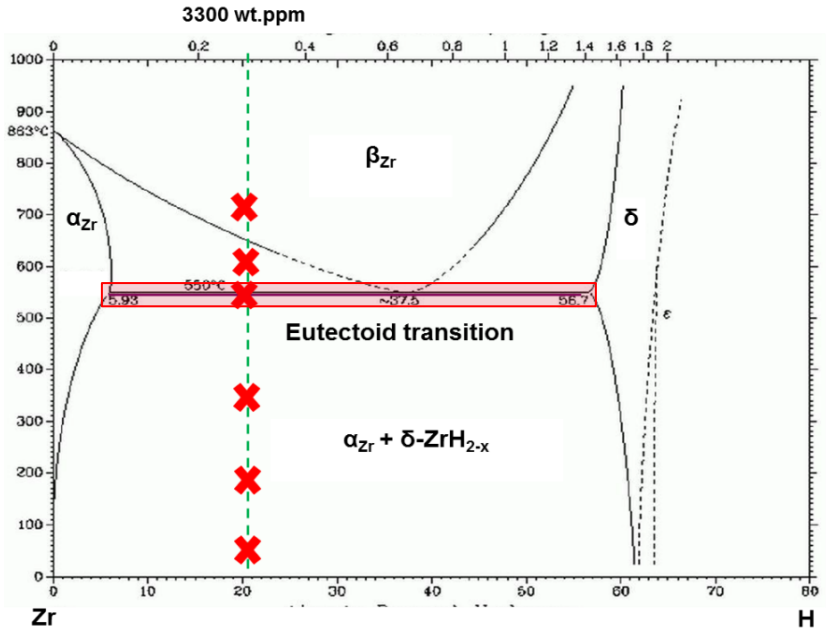
Microstructure of a tube in M5 prehydrided at 2000 wt.ppm under Ar + H2 gas mixture at 800°C

** M5 is a registered trademark of AREVA NP in the USA or others countries*

METALLURGICAL CHARACTERIZATION OF PREHYDRIDED MATERIALS

In situ neutron diffraction

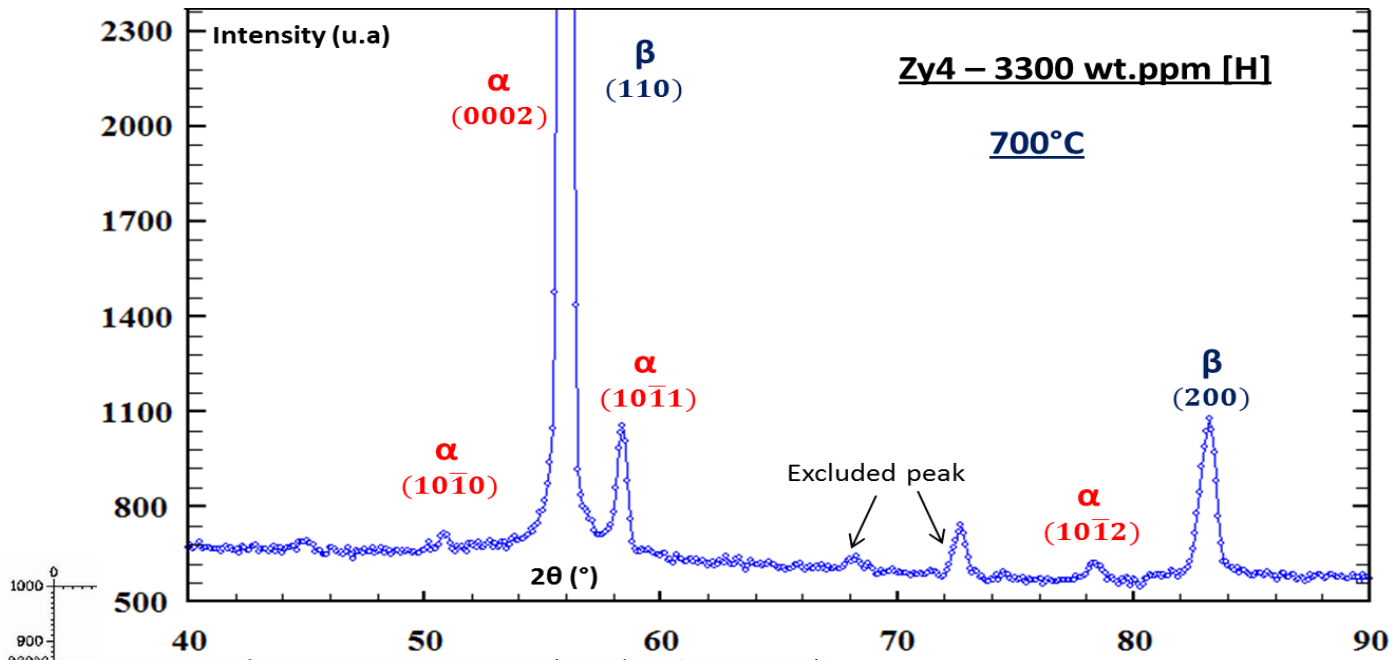


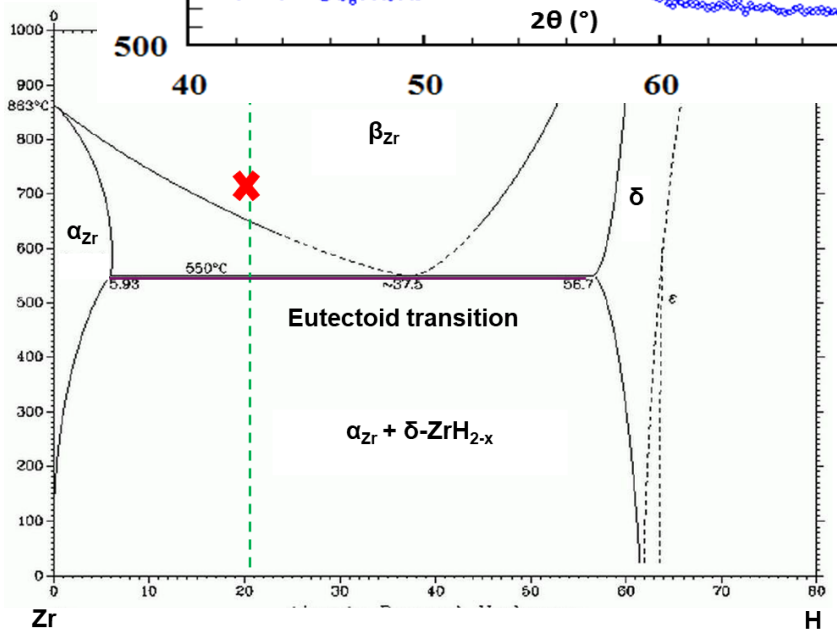


- **Experimental conditions**: Data acquisition at **various temperatures** during **step-cooling from 700°C** (near-equilibrium conditions) to RT
- **Materials**: Cladding tubes in Zircaloy-4 and M5 (5cm long)
- **Objectives**: Metallurgical evolution with temperature
- **Refinement method**: Rietveld refinement and Fullprof software

Neutron diffraction analysis performed in Orphée reactor, in collaboration with Léon Brillouin Lab (LLB)

Metallurgical evolution with temperature



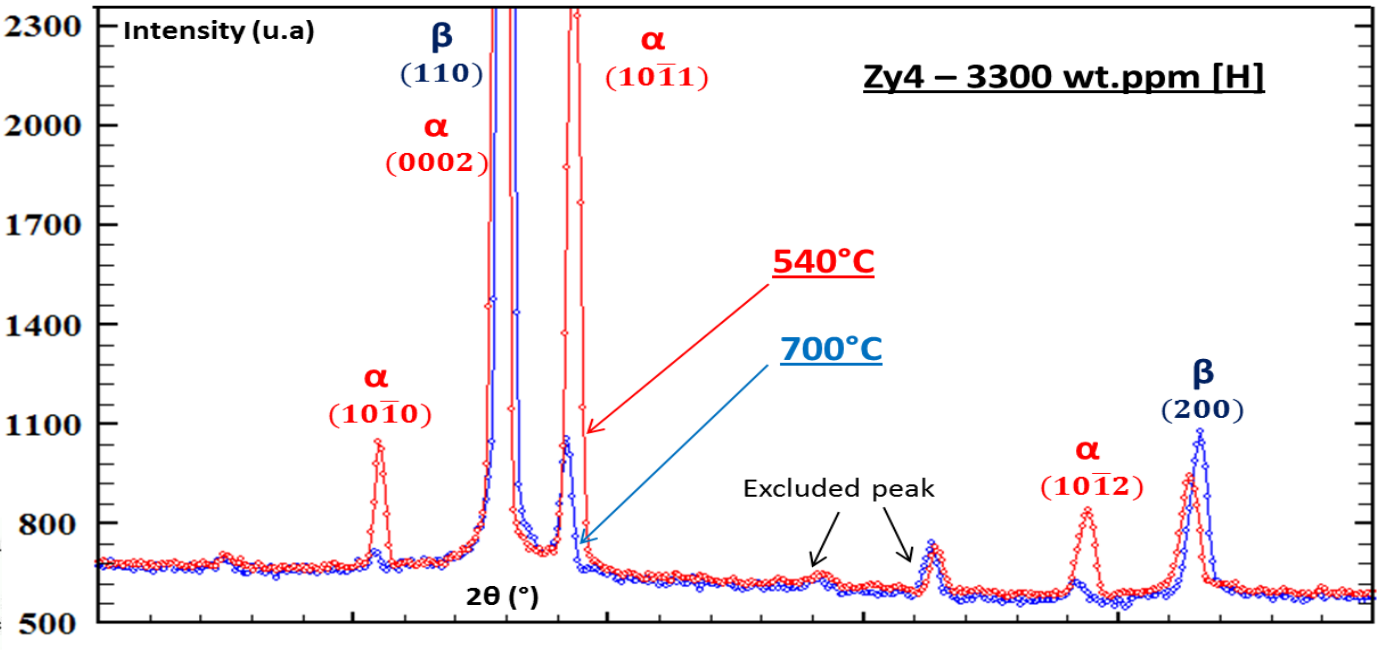


Neutron diffraction diagrams obtained for Zy4 prehydrided at 3300 wt.ppm, during **step-cooling from 700°C** after **heat-treatment at 1000°C** in steam

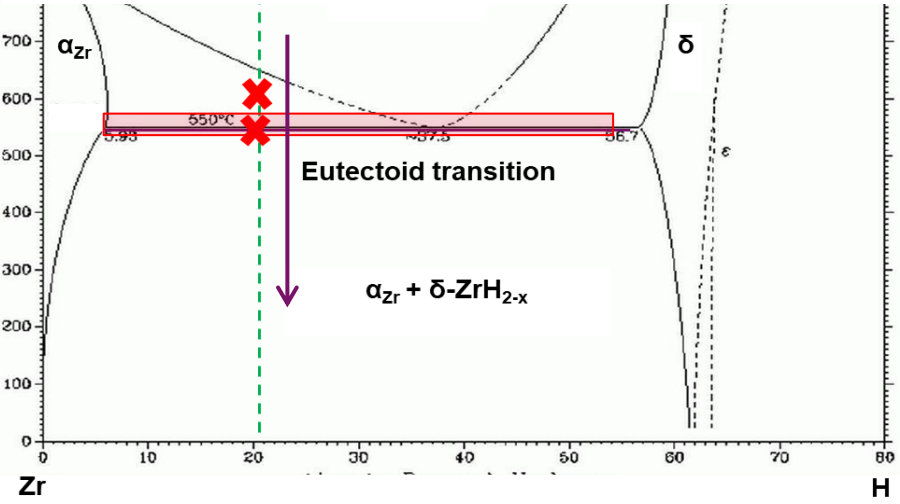
At 700°C: 2 phases

- α zirconium (minority phase)
- β zirconium (majority phase)

Metallurgical evolution with temperature



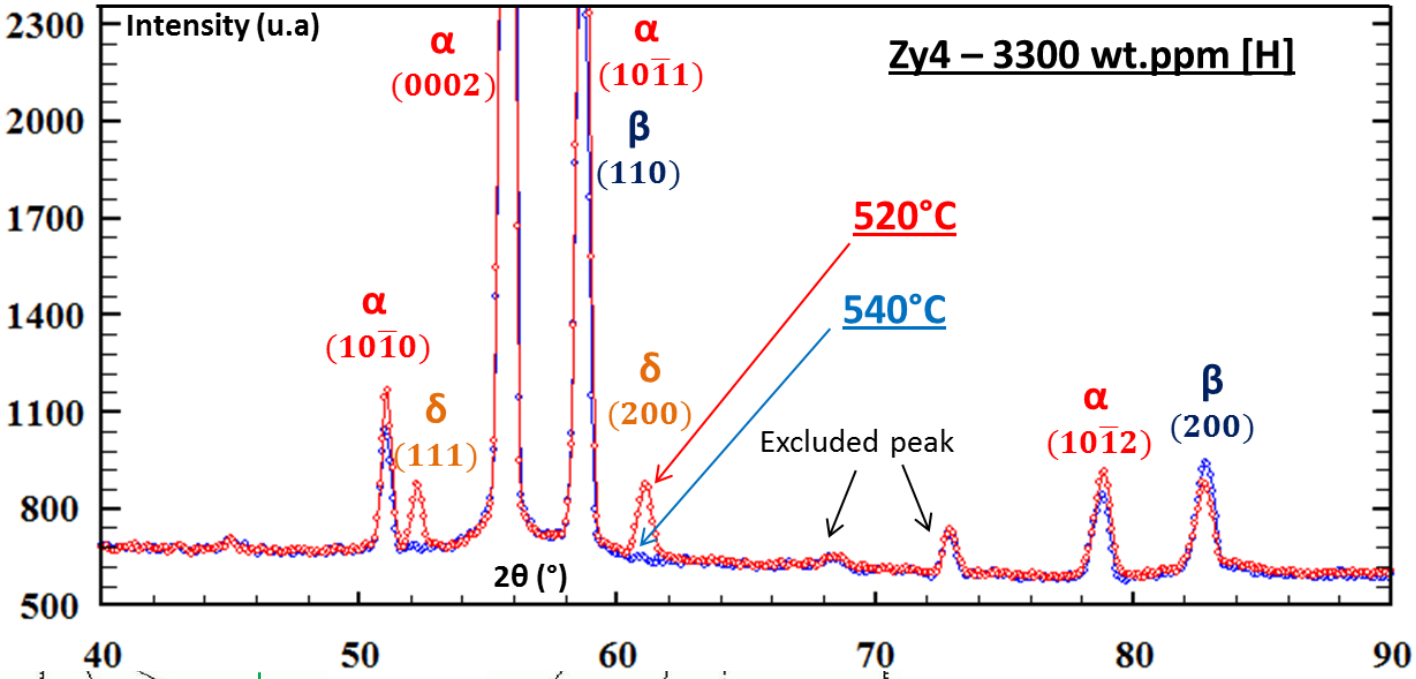
Neutron diffraction diagrams obtained for Zy4 prehydrided at 3300 wt.ppm, during **step-cooling from 700°C** after **heat-treatment at 1000°C** in steam

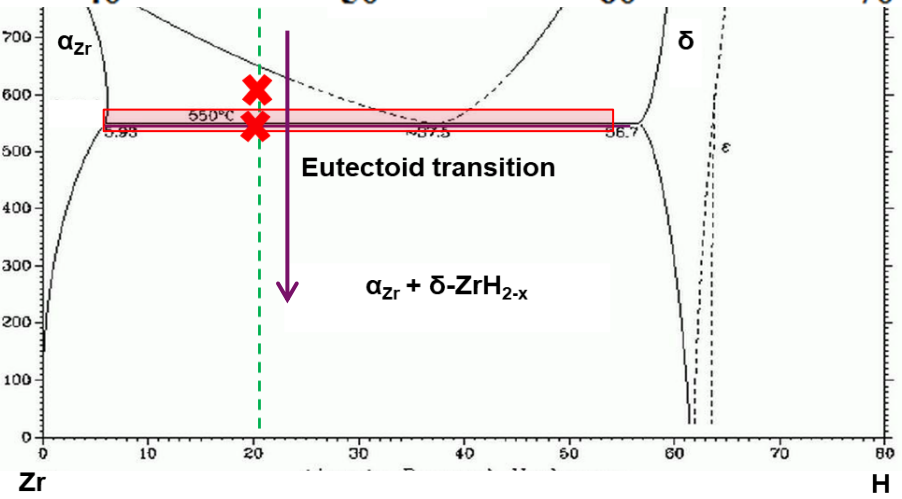


At 540°C: 2 phases α zirconium and β zirconium

- Peak intensity of β zirconium decreases
- β (at 700°C) → α + β (at 540°C)

Hydrides precipitation temperatures



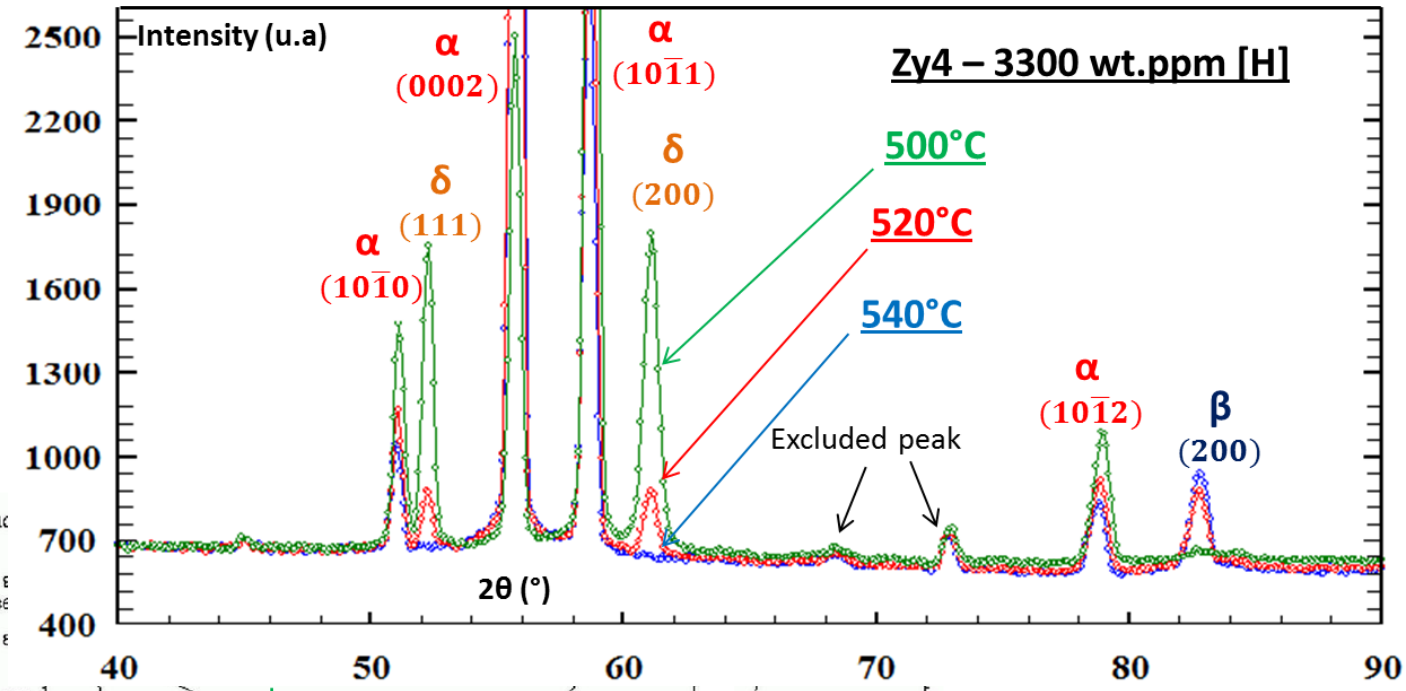


Neutron diffraction diagrams obtained for Zy4 prehydrided at 3300 wt.ppm, during **step-cooling from 700°C** after **heat-treatment at 1000°C** in steam

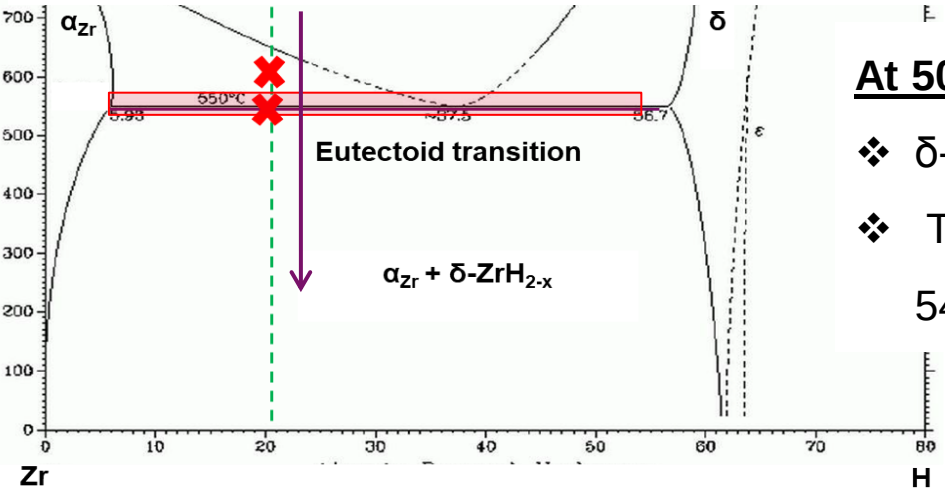
At 520°C: 3 phases

- α zirconium
- β zirconium
- δ-ZrH2-x hydrides

Hydrides precipitation temperatures



Neutron diffraction diagrams obtained for Zy4 prehydrided at 3300 wt.ppm, during **step-cooling from 700°C** after **heat-treatment at 1000°C** in steam

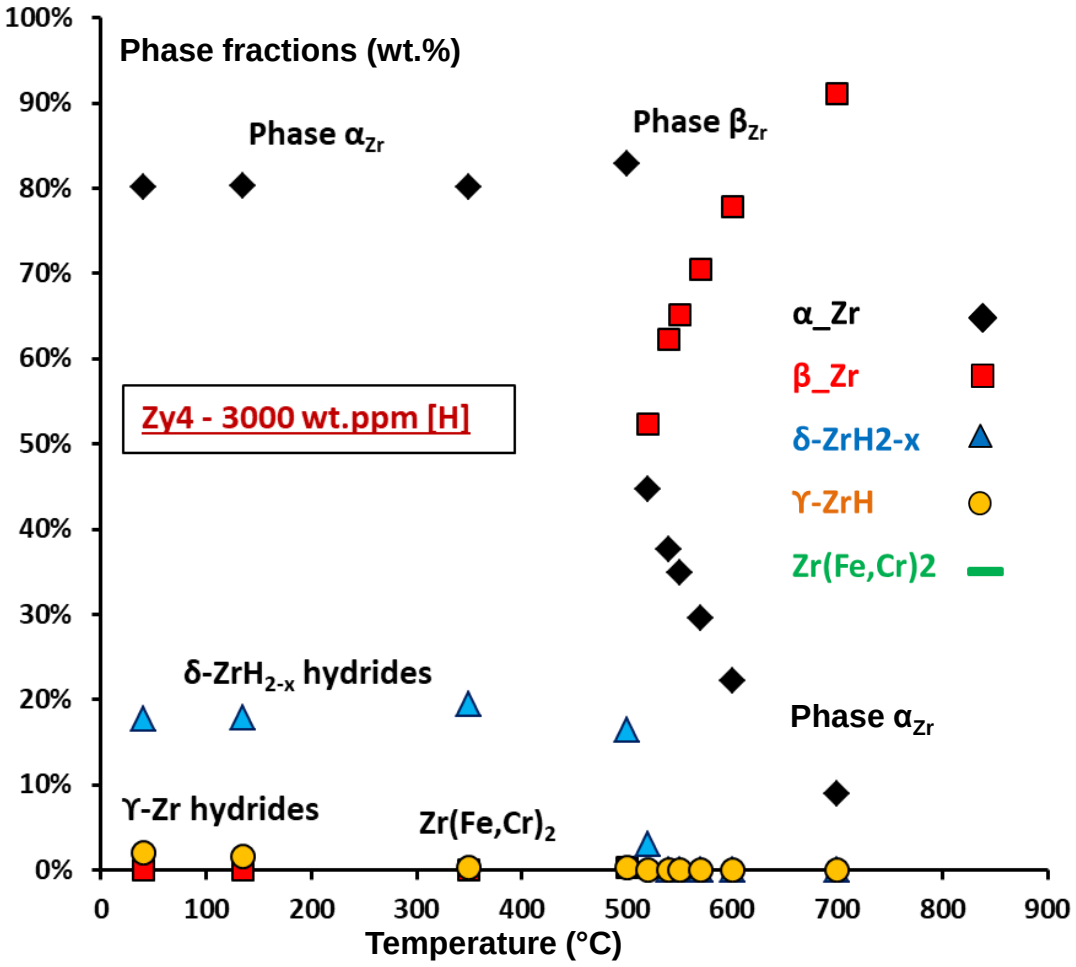


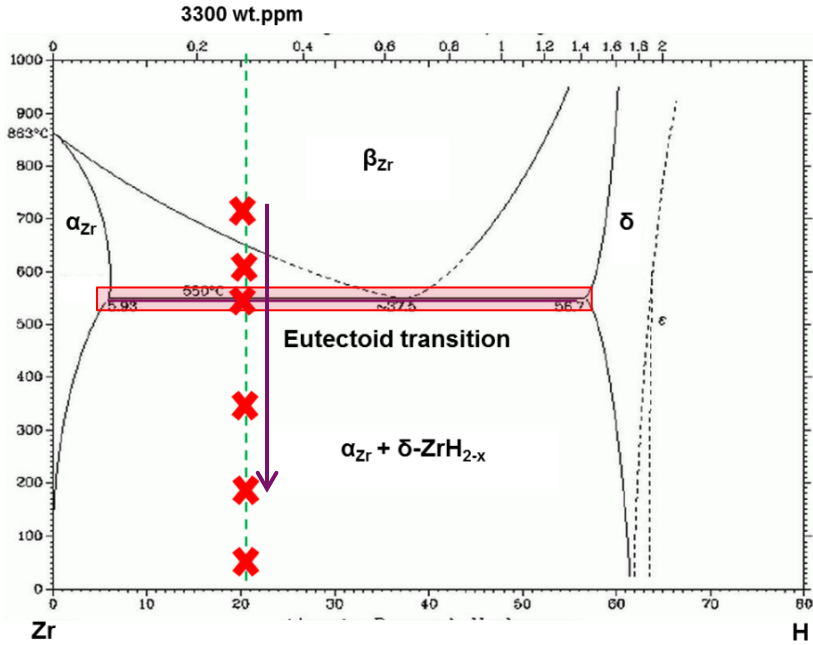
At 500°C: 2 phases **α zirconium** + **δ-ZrH2-x hydrides**

- δ-ZrH2-x precipitation temperature ~540°C
- Temperature range of **co-existence α+β+δ**: 500-540°C

M5: cf. back-up slides

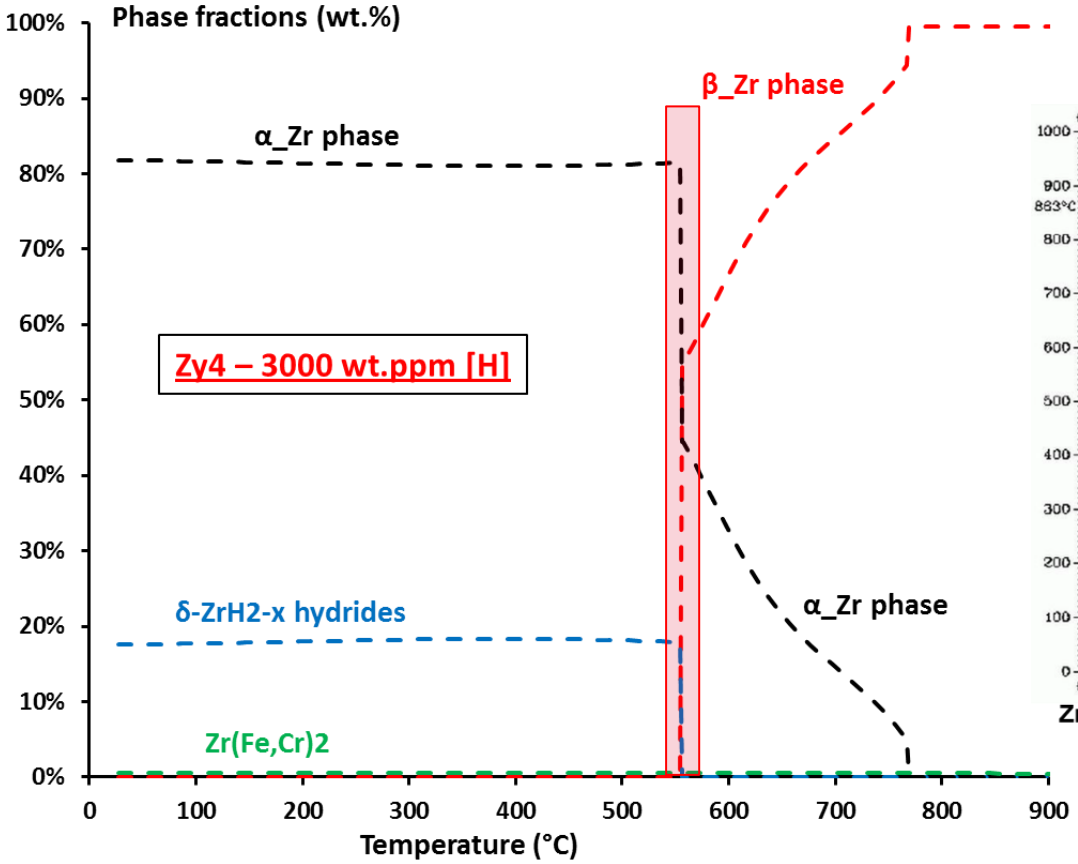
Evolution of phase fractions with temperature

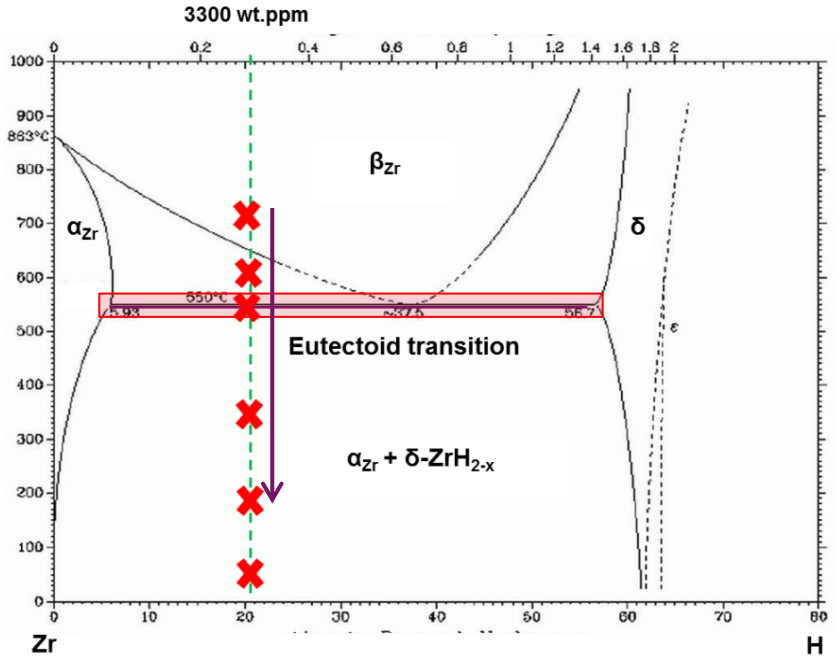




❖ **Phase weight fractions** obtained by **in situ neutron diffraction**

Evolution of phase fractions with temperature

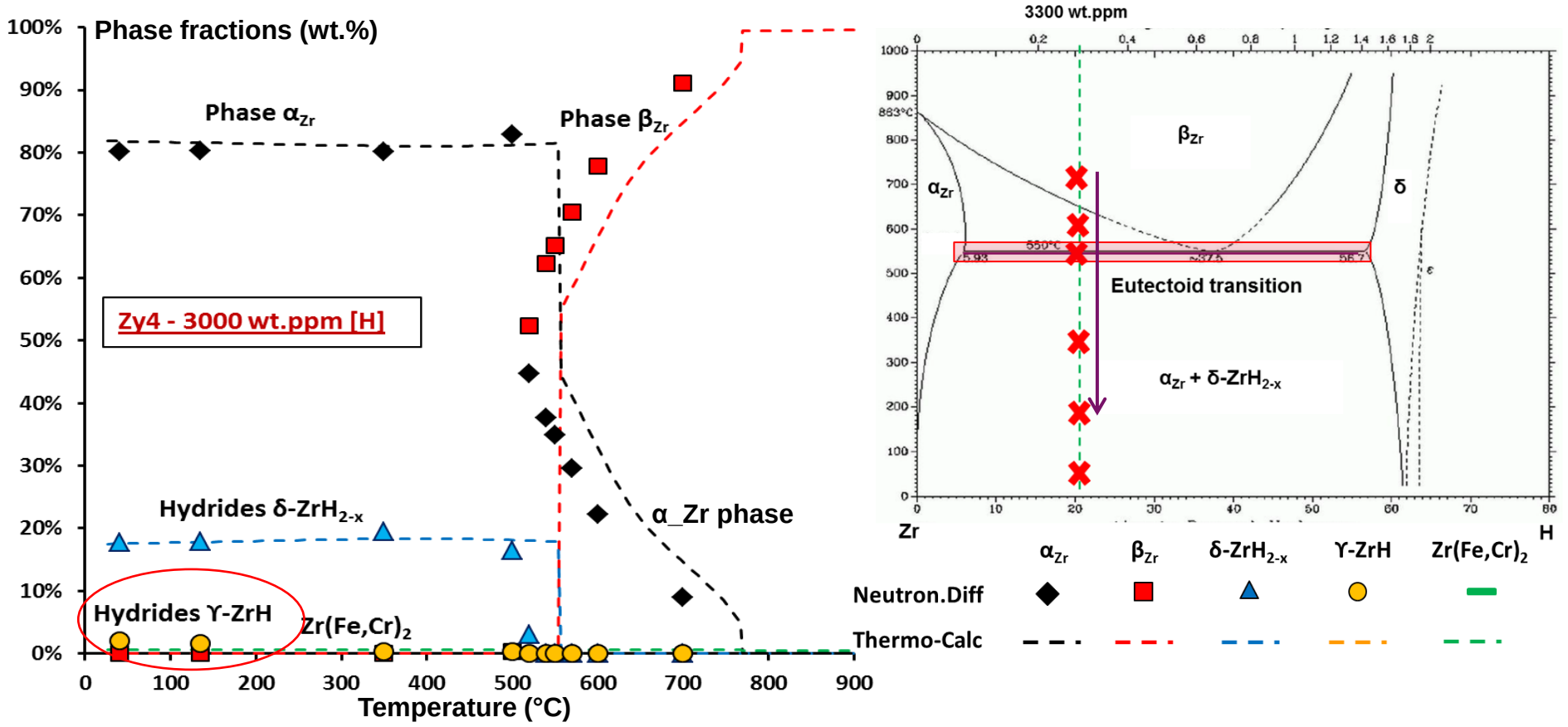




❖ **Phase weight fractions** obtained by **Thermo-Calc** and **Zircobase** in **equilibrium conditions**

Zy4 (Zr, Cr, Fe, Sn, O) – 3000wt.ppm H system

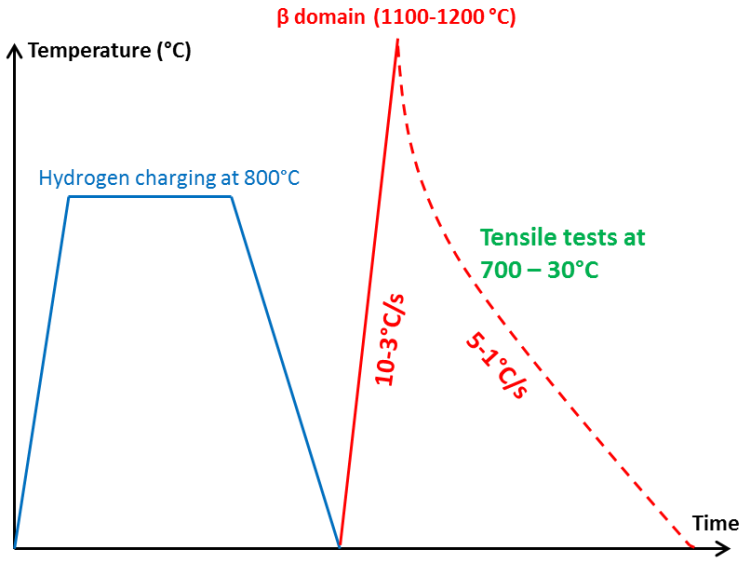
Evolution of phase fractions with temperature



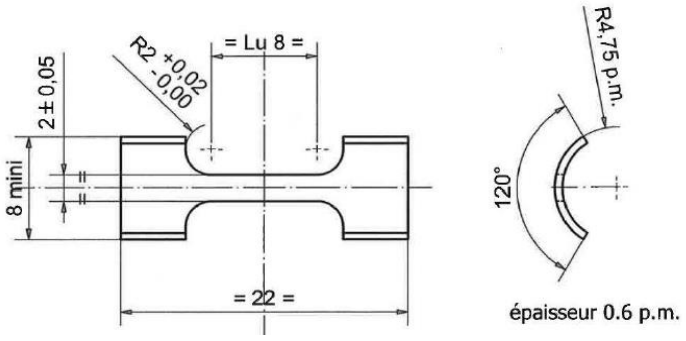
- **Good agreement** between **Thermo-Calc prediction** and **neutron diffraction data**
- **Apparition of ϒ-ZrH hydrides** (~2 wt.%) (potentially metastable) at the end of cooling

M5: cf. back-up slides

MECHANICAL BEHAVIOR OF THE HIGHLY HYDROGEN-CHARGED (PRIOR-) β PHASE DURING COOLING FROM β DOMAIN TEMPERATURES



Thermal cycles for tensile tests



Uniaxial tensile tests in air

Temperatures: **700 °C – 30°C** on cooling from **β_{Zr}** domain **(1100-1200°C)**

Geometry: Specimens were fabricated **from the cladding tubes**

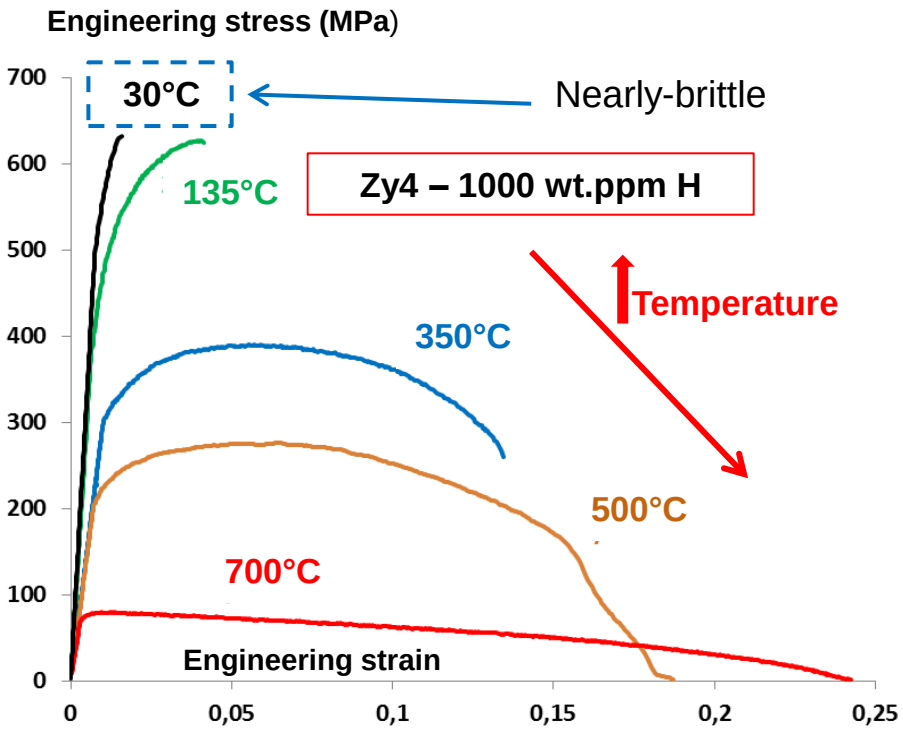
Materials: Zy4 et M5™

- **as-received**
- **prehydrided** at **1000, 2000 and 3000 wt.ppm**

Strain rate: **$\dot{\varepsilon} = 10^{-1}\ s^{-1}$** (test time < 1s: avoid oxidation + microstructural evolution during tensile tests)

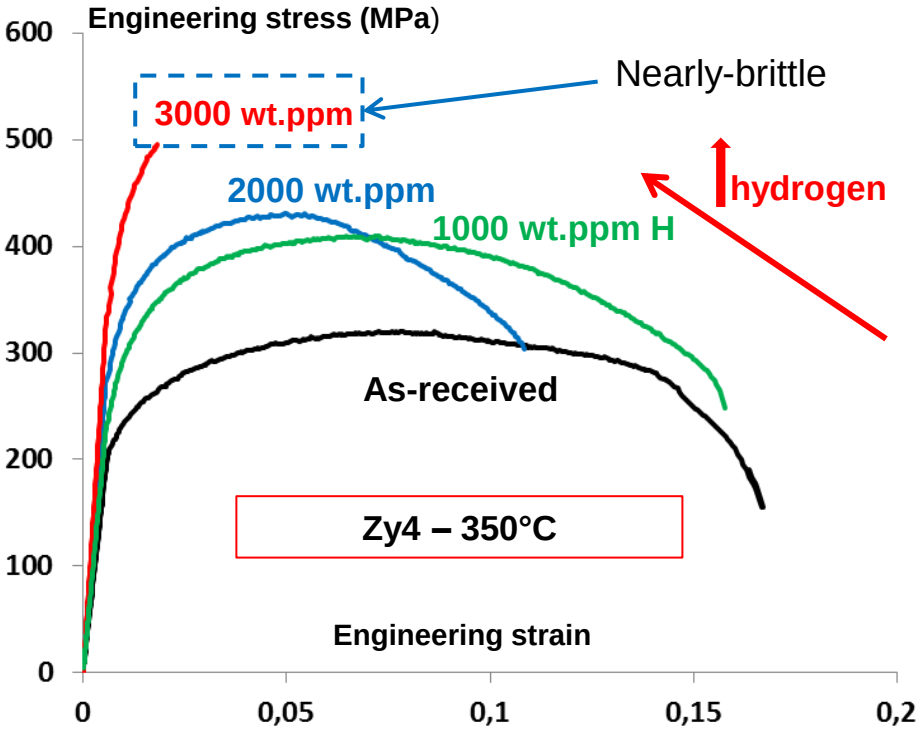
Fast camera (250 images/second) → **Digital Images Correlation**

Temperature effect



Engineering stress-strain curves of tensile tests on Zy4 prehydrided at 1000 wt.ppm

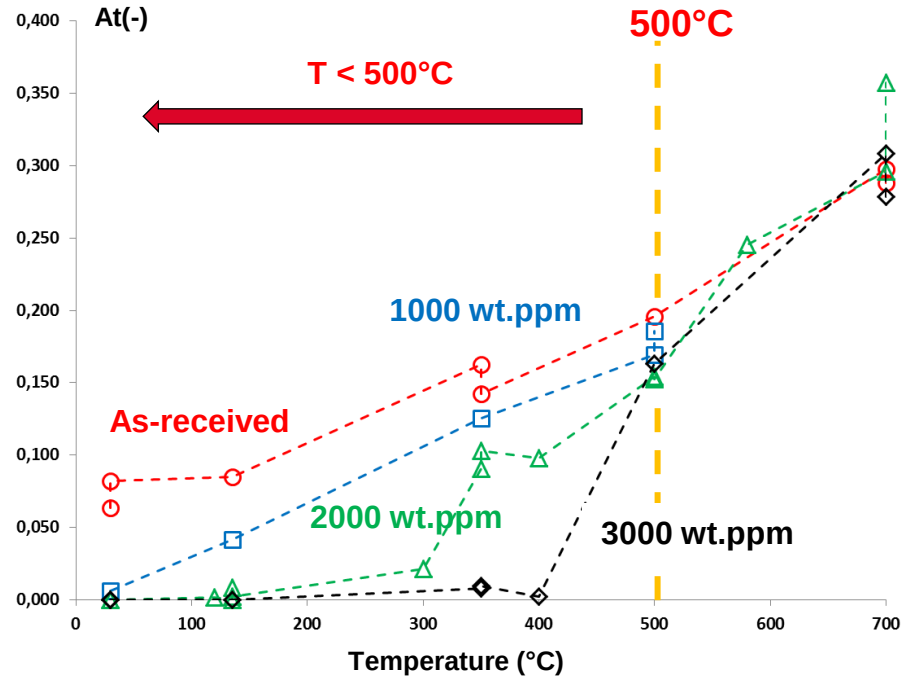
Hydrogen embrittlement effect

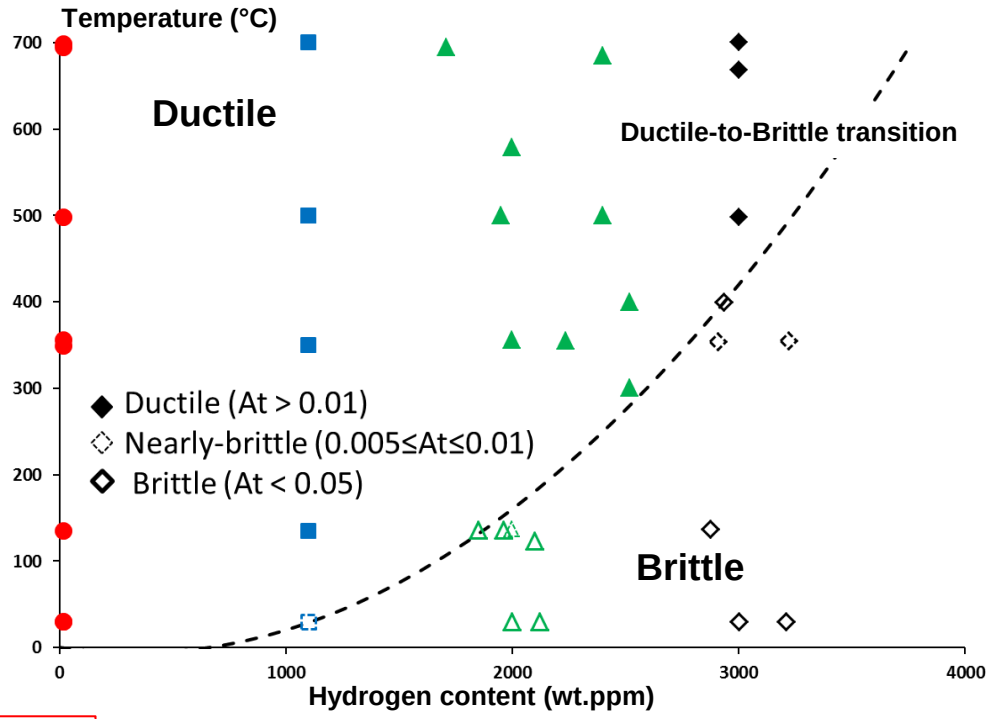


Engineering stress-strain curves of tensile tests on as-received and Zy4 prehydrided at 1000, 2000 and 3000 wt.ppm, tested at 350°C

Turque et al., 2016. Mechanical behavior at high temperature of highly oxygen- or hydrogen-enriched α and (prior-) β phases of zirconium alloys. In Zirconium in the Nuclear Industry: 18th International Symposium, ASTM STP 1597, Hilton Head, SC, USA, 240-280.

Plastic elongation of as-received and Zy4 prehydrided at 1000, 2000 and 3000 wt.ppm, tested in tension on cooling after heat-treatment in the β_{Zr} domain





- T > 500°C, no effects of hydrogen on ductility
- T < 500°C, significant embrittlement effect of hydrogen

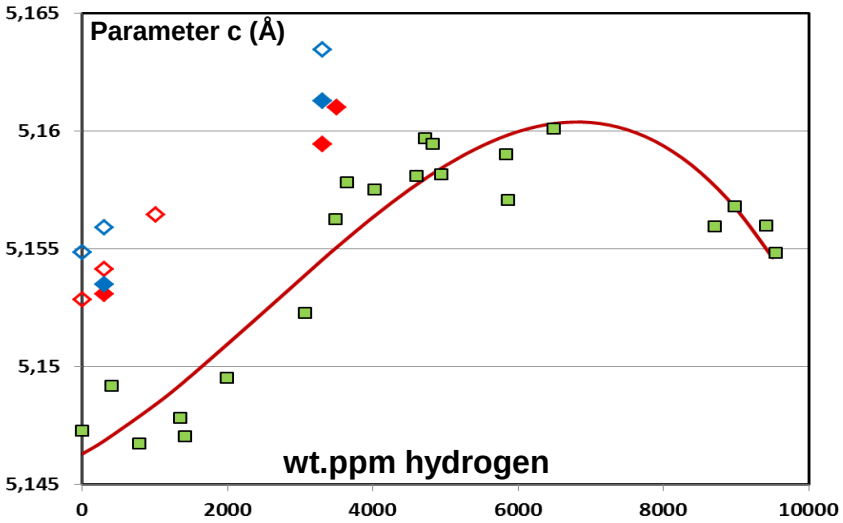
CONCLUSION AND FURTHER WORK

Metallurgical behavior:

- Concerning the **evolution of phase weight fractions** with temperature, **good agreement between neutron diffraction data and thermodynamic prediction** obtained by Thermo-Calc software and thermodynamic database Zircobase of CEA. **Only slight differences between Zy4 and M5**

- The **precipitation of metastable γ-ZrH hydrides** was **observed** at low temperatures in materials containing **high** (1000 and 3000 wt.ppm) and **lower** (300 wt.ppm) **hydrogen contents** (cf. back-up slides)

- **Same observations** made by KIT (Pshechnikov et al., 2015) for the **evolution of lattice parameters** (a and c) at room temperature as a **function of mean hydrogen content**. Need further investigation



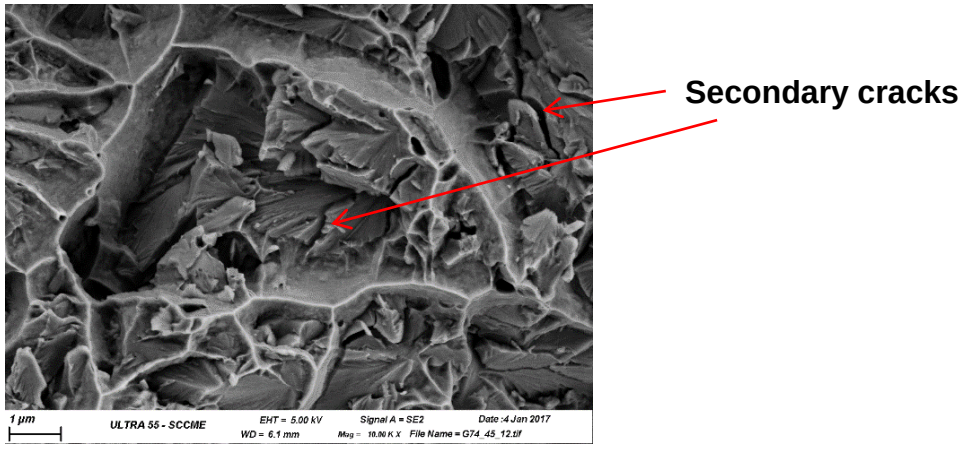
Evolution of c *as a function of* ***hydrogen*** *content on* ***Zy4*** *samples (neutron and X-ray diffraction). Comparison to XRD at RT obtained on Zy4 by Pschenichnikov et al., 2015 (c.f. back-up slides)*

CONCLUSION AND FURTHER WORK

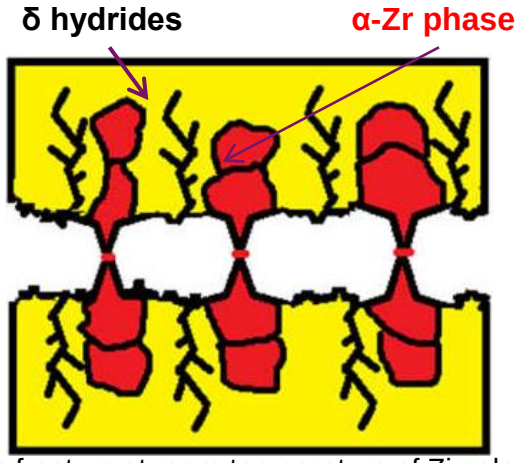
Mechanical behavior:

- **Embrittlement effect of hydrogen** for temperatures **below 500°C**
- **T ≤ 500°C**, **↑[H] → mechanical resistance increases** (in case of ductile materials)
- **700°C**, **mechanical resistance decreases** when **↑[H]**
- **Ductile-to-Brittle transition map** as a function of **temperature** and **hydrogen content** was established from experimental data

- **Further work**: **fracture mechanisms** of **highly hydrogen-charged materials** and **coupled effects** of **oxygen** and **high hydrogen contents** on **mechanical behavior** and **metallurgical properties**.
- **Mechanical behavior law** as a function of **oxygen, hydrogen** and **temperature** → simulation/modeling



Fracture surface obtained by SEM of Zy4 with 2200 wt.ppm H, tested at 350°C in tension on cooling after heat-treatment in β domain



Schematic fracture at room temperature of Zircaloy-4 hydrogenated at high temperature, *Pschenichnikov et al., 2015*

Pshenichnikov et al., 2015. Hydrides and fracture of pure zirconium and Zircaloy-4 hydrogenated at temperatures typical for loss-of-coolant accident conditions. Proceeding of 23th International Conference on Nuclear Engineering, May 17-21, 2015, Chiba, Japan.

Thank you for your attention

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EXPERIMENTAL PROCEDURE

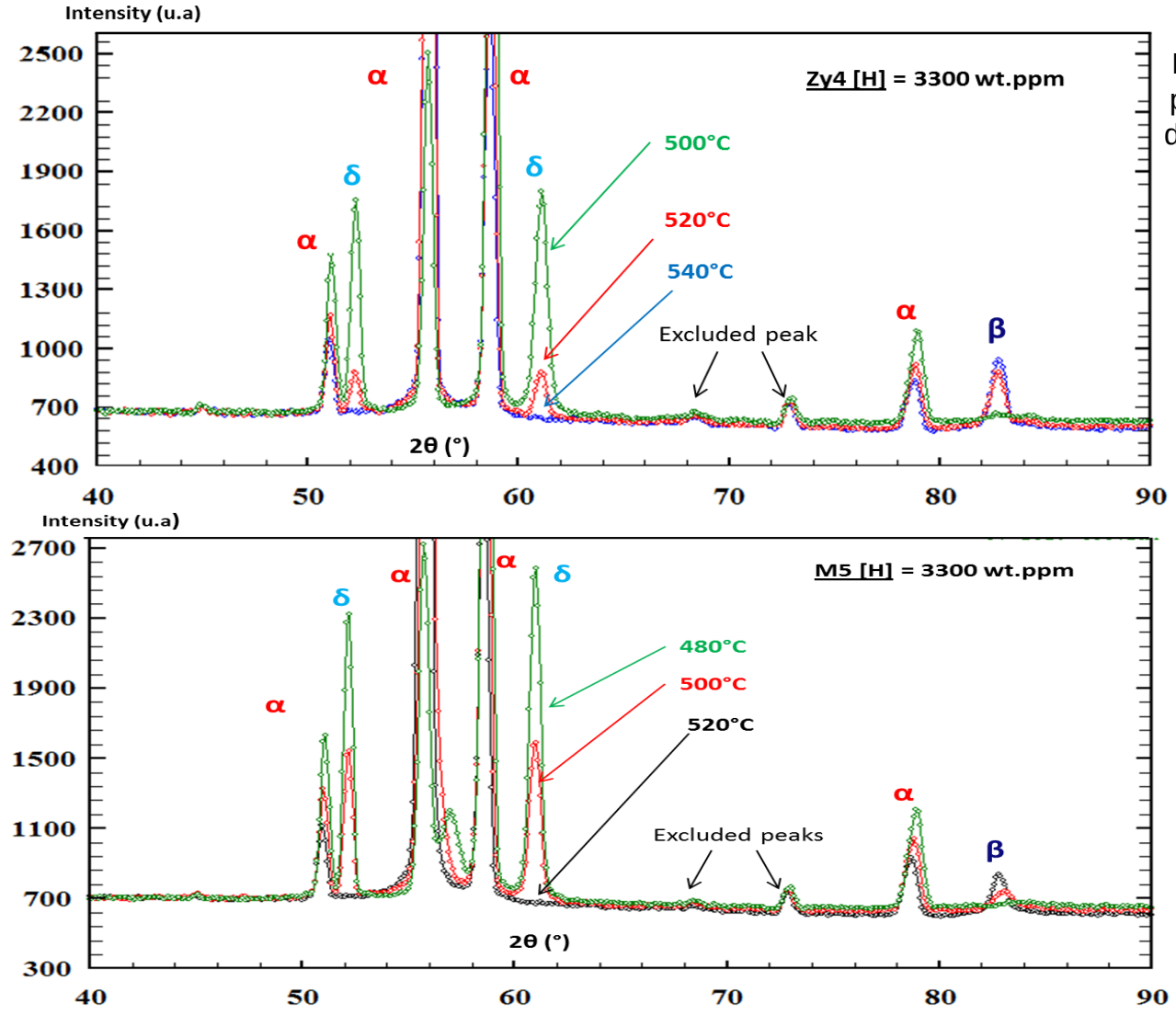
Metallurgical characterization (phase fractions, phase transition temperatures and evolution of lattice parameters):

- In situ neutron diffraction during cooling from 700°C down to 20°C
- Neutron and X-ray diffraction at RT
- Comparison to thermodynamic equilibrium prediction obtained by Thermo-Calc and Zircobase thermodynamic database

Mechanical characterization:

- Uniaxial tensile tests at various temperatures (700°C-30°C) during cooling from high temperatures

Hydrides precipitation temperatures

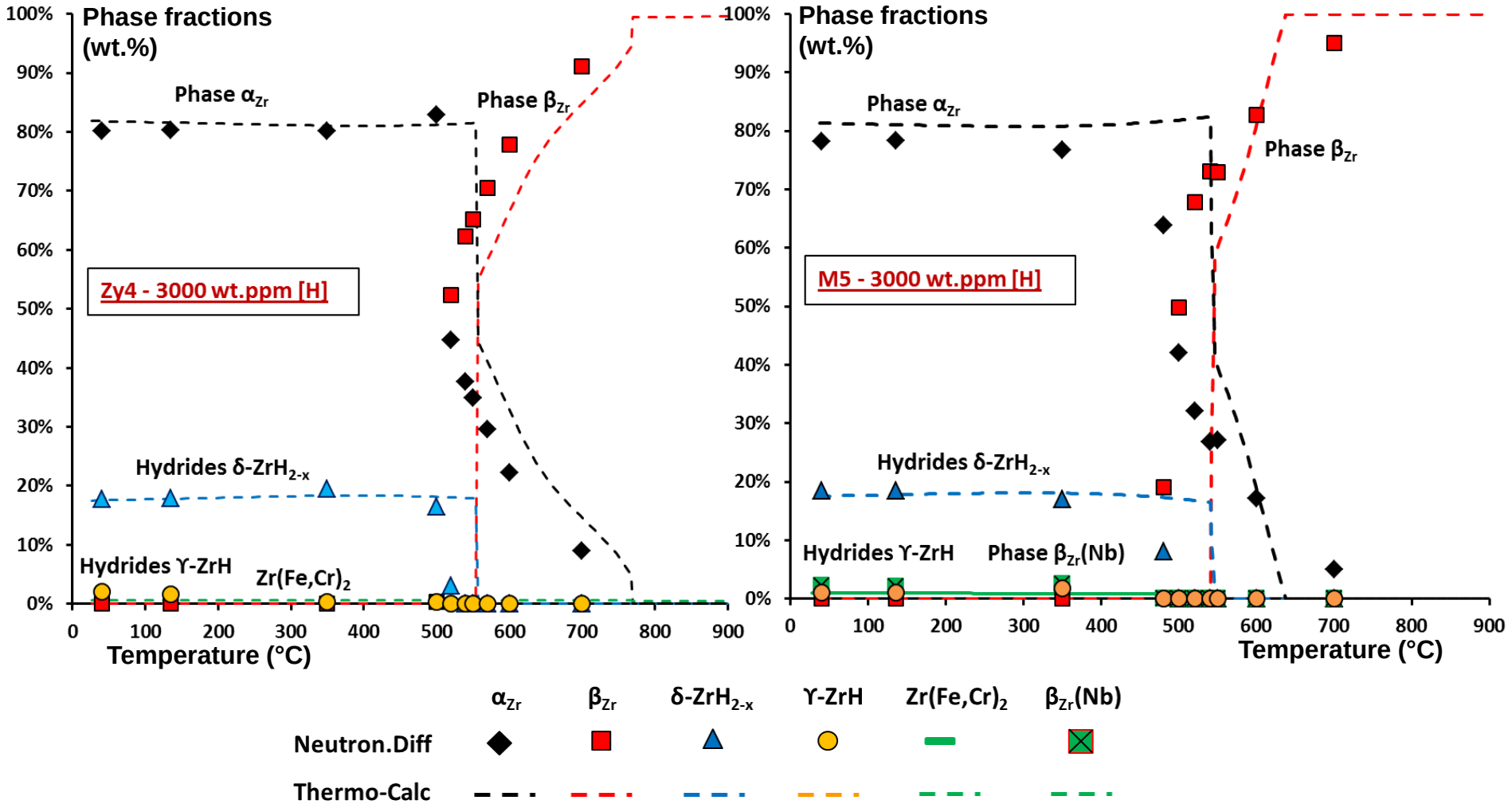


Neutron diffraction diagrams obtained for prehydrided Zy4 and M5 at 3300 wt.ppm, during step-cooling from 700°C after heat-treatment at 1000°C in steam

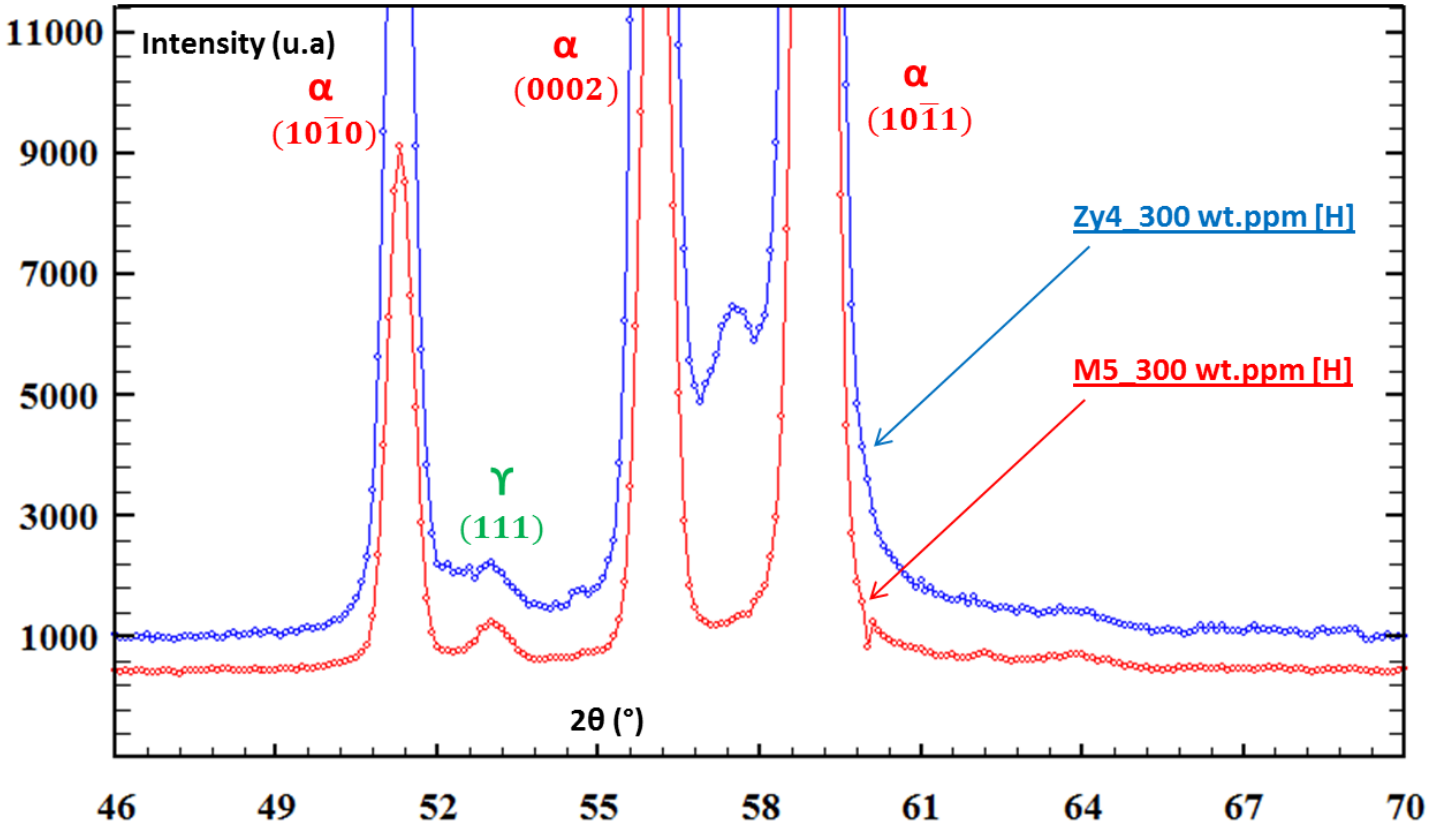
- δ-ZrH_{2-x} starts to precipitate in Zy4-3300H at ~ 540°C
- Temperature range of co-existence α + β + δ : 500°C-540°C

- Apparition of δ-ZrH_{2-x} hydrides in M5-3300H at ~520°C
- Temperature range of co-existence α + β + δ : 480°C-520°C

Evolution of phase fractions with temperature

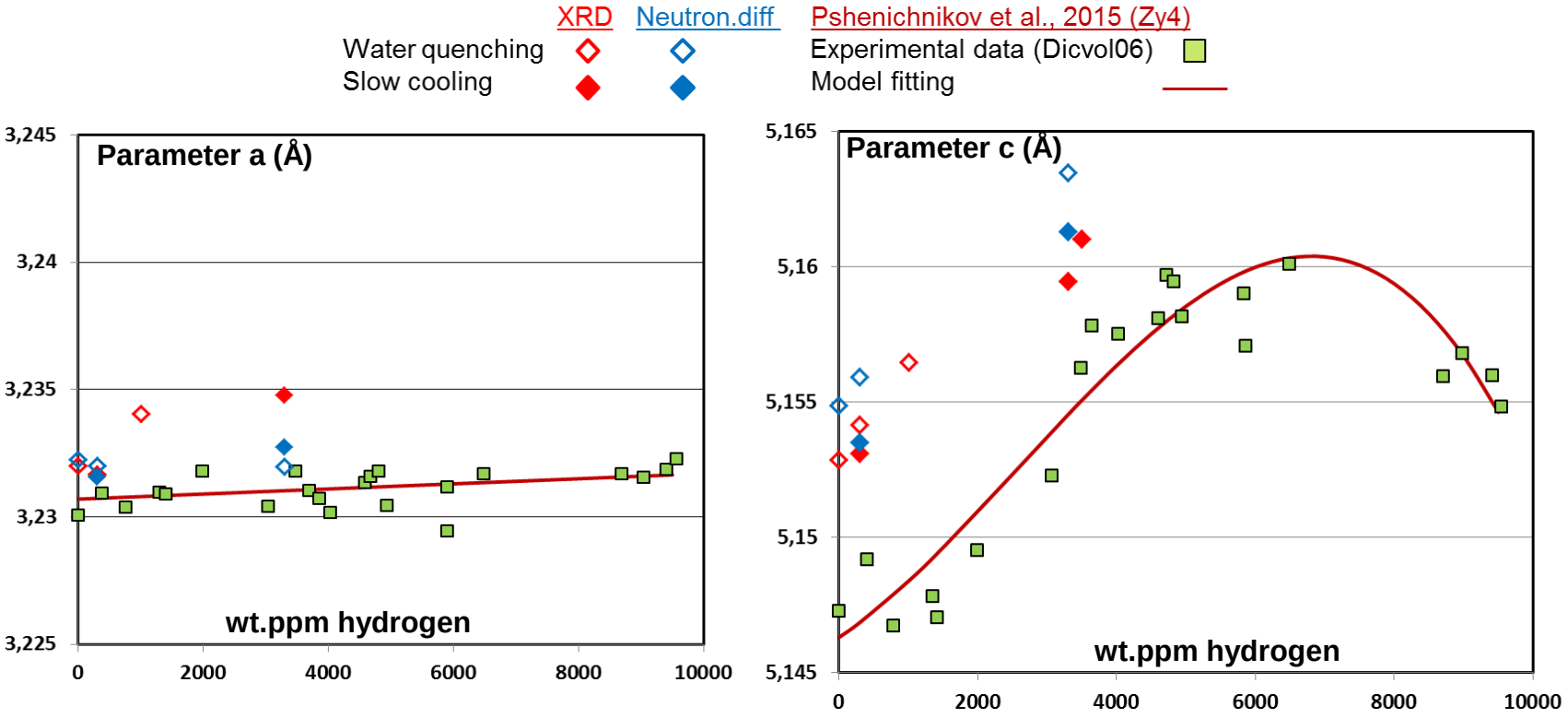


Presence of γ-ZrH hydrides at low temperatures



Neutron diffraction diagrams obtained at RT for Zy4 and M5 prehydrided at **300 wt.ppm**, water quenched after heat-treatment at 1000°C in steam

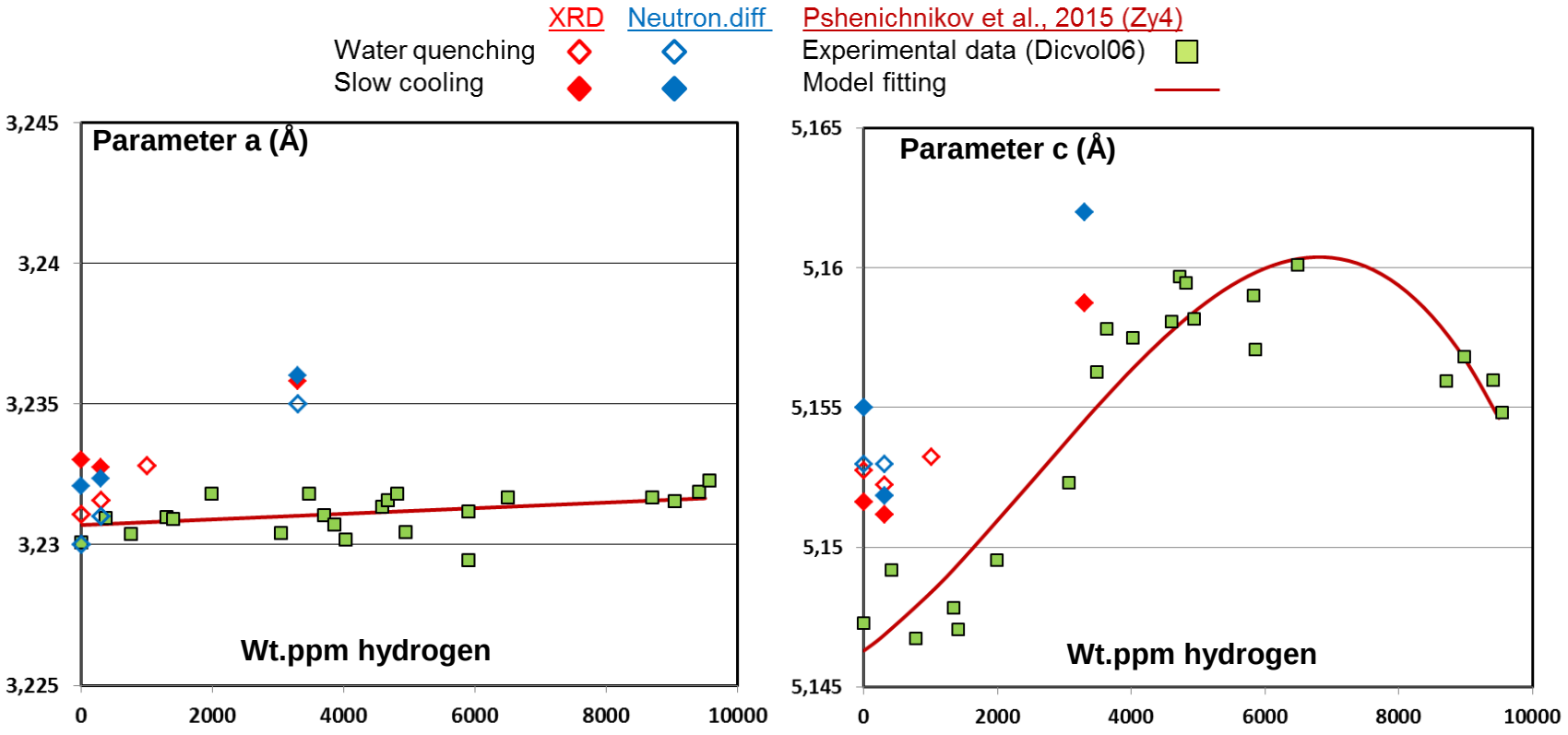
Evolution of lattice parameters – Diffraction at RT (case Zy4)



The results of both CEA XRD and neutrons diffraction at room temperature show a relative a and c parameters increase as a function of the mean H content, as already observed by KIT for Zy4

Pshenichnikov et al., 2015. Microstructure and mechanical properties of Zircaloy-4 cladding hydrogenated at temperature typical for loss-of-coolant accident (LOCA) conditions. Nuclear Engineering and Design 238, 33-39.

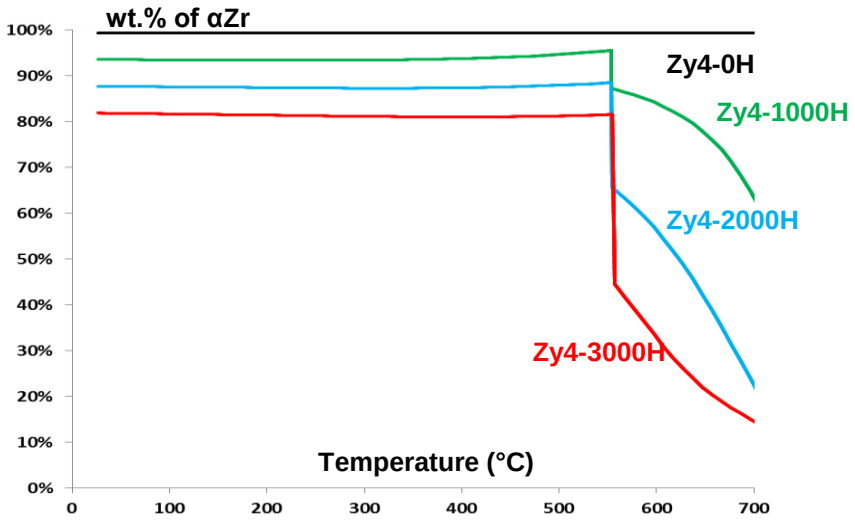
Evolution of lattice parameters – Diffraction at RT (case M5)

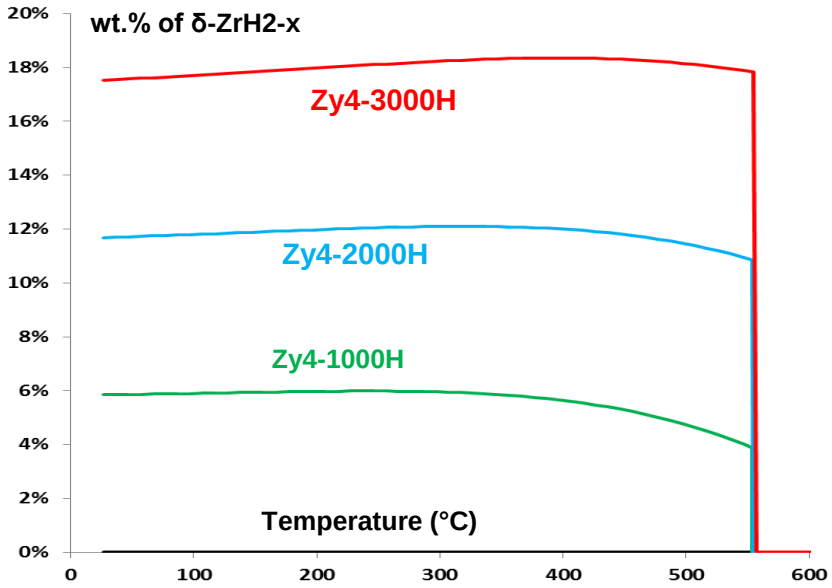


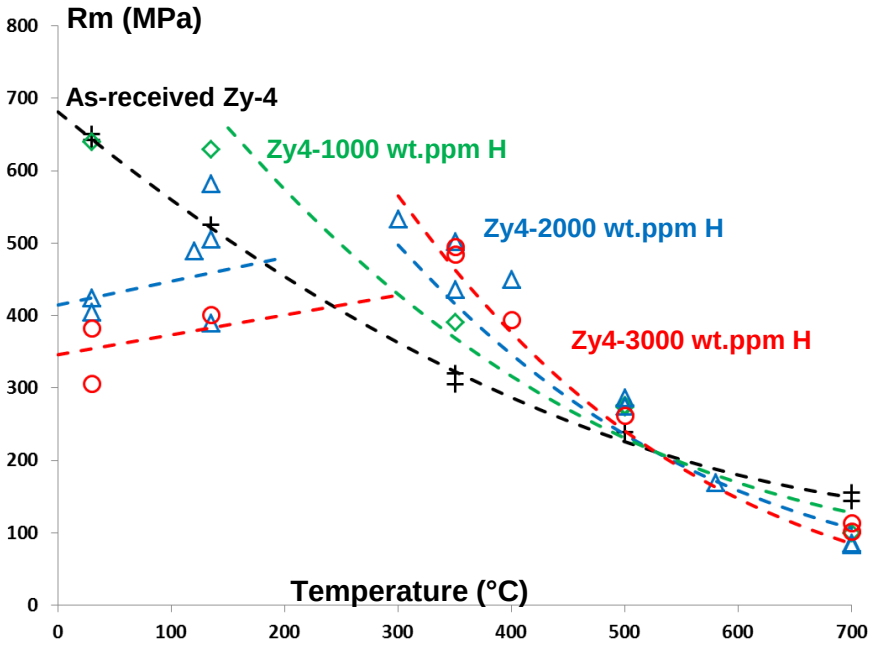
Similar results obtained for M5

Pshenichnikov et al., 2015. Microstructure and mechanical properties of Zircaloy-4 cladding hydrogenated at temperature typical for loss-of-coolant accident (LOCA) conditions. Nuclear Engineering and Design 238, 33-39.

Evolution of phase weight fractions obtained by thermodynamic calculation at equilibrium by **Thermo-Calc** and thermodynamic database **Zircobase**



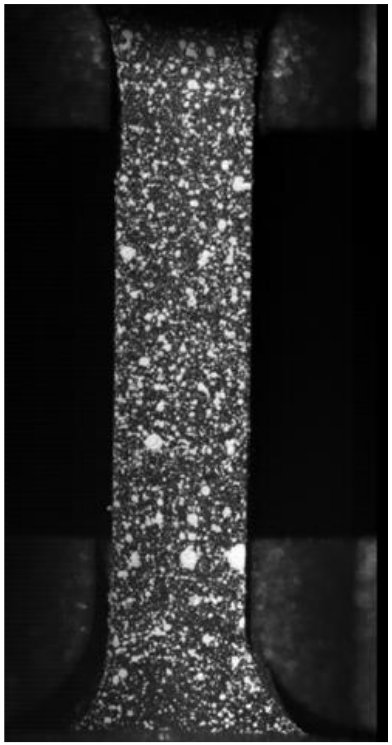


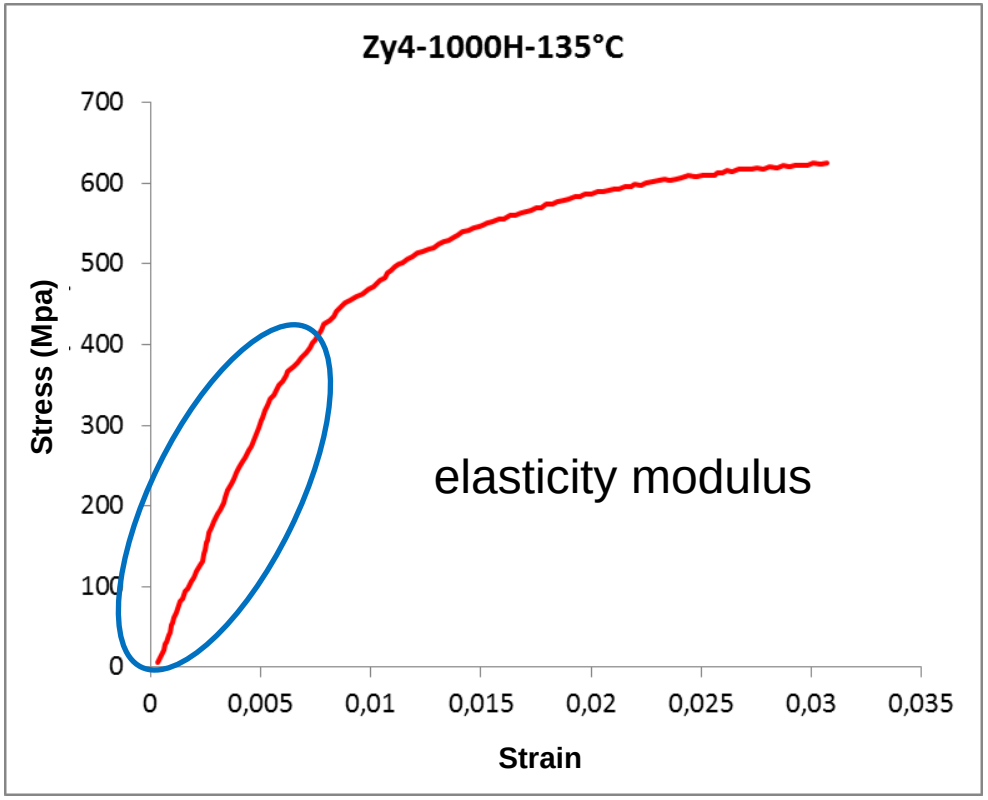


Ultimate stress of as-received and Zircaloy-4 prehydrided at 1000, 2000, 3000 wt.ppm, tested in tension on cooling after heat-treatment in β domain

- ↓ mechanical resistance when ↑ temperature
- T ≤ 500°C, ↑[H] → ↑ mechanical resistance (in case of ductile materials)
- 700°C, ↓ mechanical resistance when ↑[H]

MECHANICAL CHARACTERIZATION





Stress-strain curve of Zy4 prehydrided at 1000 wt.ppm, tested at 135°C, obtained by Digital Images Correlation

Air oxidation of sponge based E110 cladding alloy at high temperatures

Jakub Krejčí[1,2], **Pavel Vrbka[1,2]**, Jitka Kabátová[1], Jan Kočí[1], František Manoch[1], Věra Vrtílková[1]

[1] UJP PRAHA a.s., Nad Kamínkou 1345, 156 10 Praha - Zbraslav, Czech Republic
[2] Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague, V Holešovičkách 2, 180 00 Praha 8, Czech Republic

Abstract: The reaction mechanism between a sponge based E110 cladding alloy and air at temperatures ranging from 700 to 1200°C was studied. Air attack under prototypical conditions with air ingress during a severe reactor accident or a spent fuel pond accident was investigated. Oxidation in air atmosphere leads to major degradation of the cladding material. Exposure times covered both pre- and post-breakaway regimes.
Autoclave pre-oxidized samples were exposed as well. Pre-oxidation prevents cladding surface against nitrogen attack and leads to significant decrease of the oxidation rate.
Metallographic observations along with microhardness testing were carried out to study the material microstructure. The oxide layer thickness and the α-Zr(O) sublayer thickness were measured. The hydrogen content was measured and residual mechanical properties were studied using Ring Compression Tests.
A comparison of oxidation kinetics with existing models used in severe accident computer codes was provided.

Key words: E110, high-temperature oxidation, air, mechanical properties



Air Oxidation of Sponge Based E110 Cladding Alloy at High Temperatures

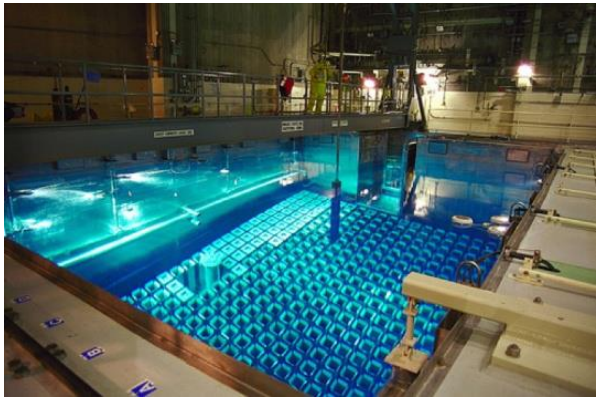
P. Vrbka, J. Krejčí, J. Kabátová, J. Kočí, F. Manoch, V. Vrtílková

Presentation for 23rd International QUENCH Workshop

Air Oxidation Experiments

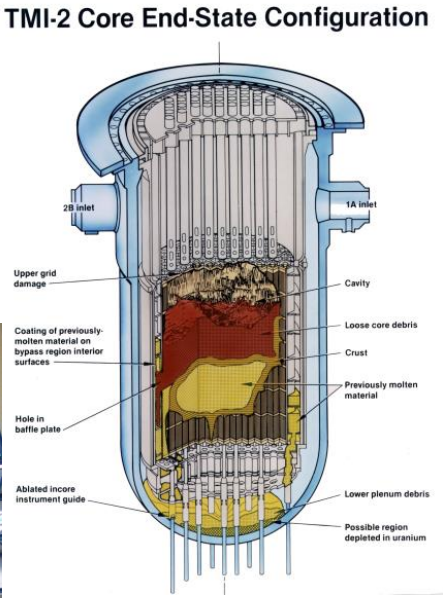
19th October 2017

Introduction



- Scenarios for high temperature air oxidation:
  - Loss of cooling accident in spent fuel pool
  - Air ingress during loss of integrity reactor vessel
  - Loss of integrity of spent fuel transportation cask





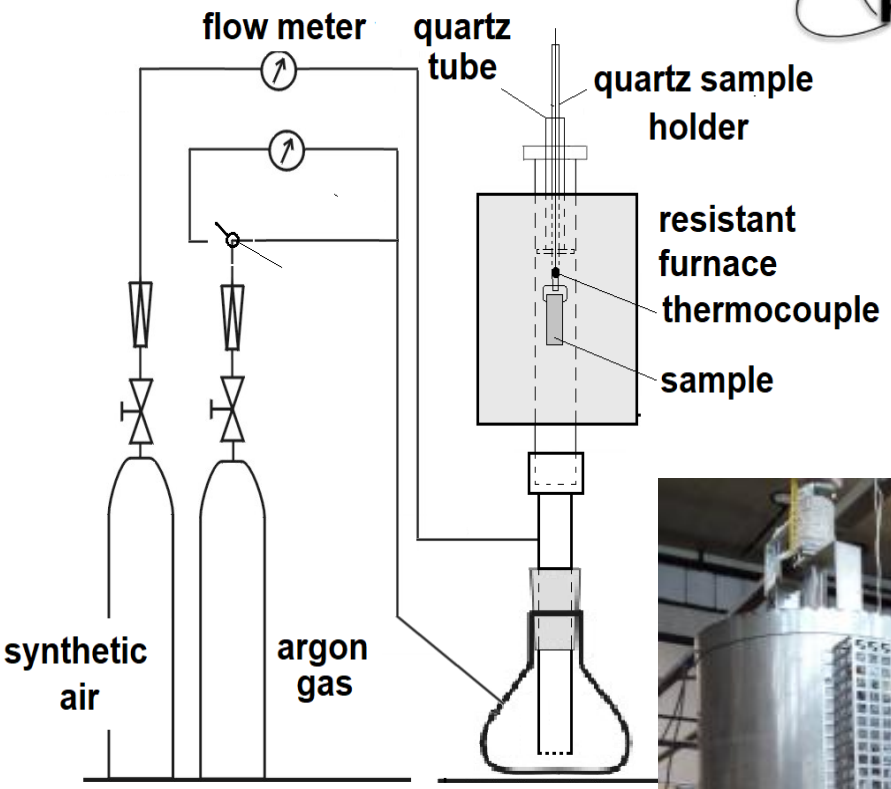
Specimens

- Alloy E110 (sponge zirconium)
- Size: Ø 9.1×7.93 mm, length 30 mm
- Chemical composition:

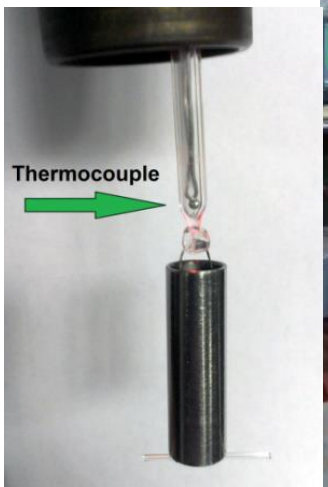
	Zr	Nb[%]	Fe [ppm]	H [ppm]
E110	Balance	1,01	500	3

- Double-side oxidation
- As-received and pre-oxidized samples
- Pre-oxidation:
  - oxide thickness: 5 μm
  - steam 425°C/10.7MPa, 42 days
  - H: 20 ppm

Test procedure and equipment



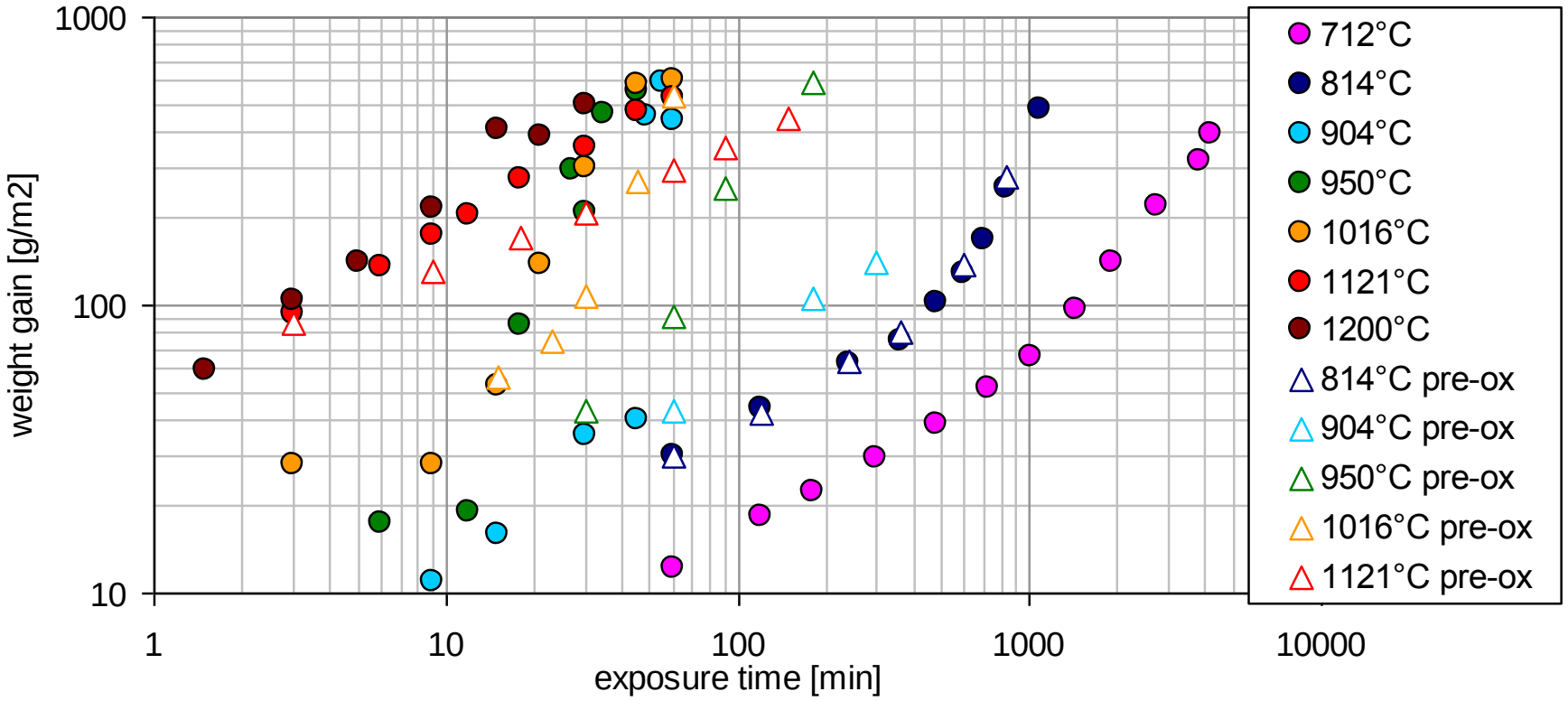
- **Temperature:** 700–1200 °C
- **Oxidizing gas:**
  - Air (79%N_2 21%O_2) 18 l/h + argon 6 l/h (30 +10 l/h; 6+2 l/h)
  - Atmospheric pressure
  - Cooling sample by argon 10 l/min





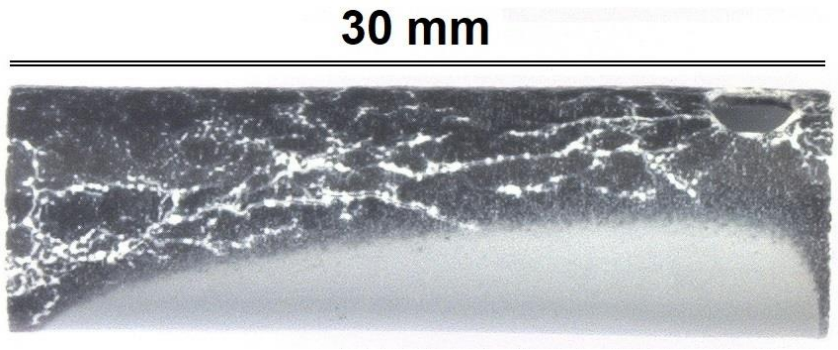
E110 oxidation kinetics

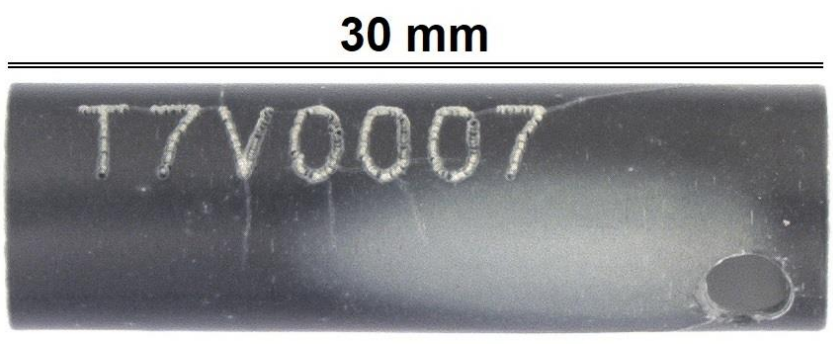
- Lower weight gain of pre-oxidized sample from 900 °C



* Real temperature was slightly higher than 700, 800,… °C

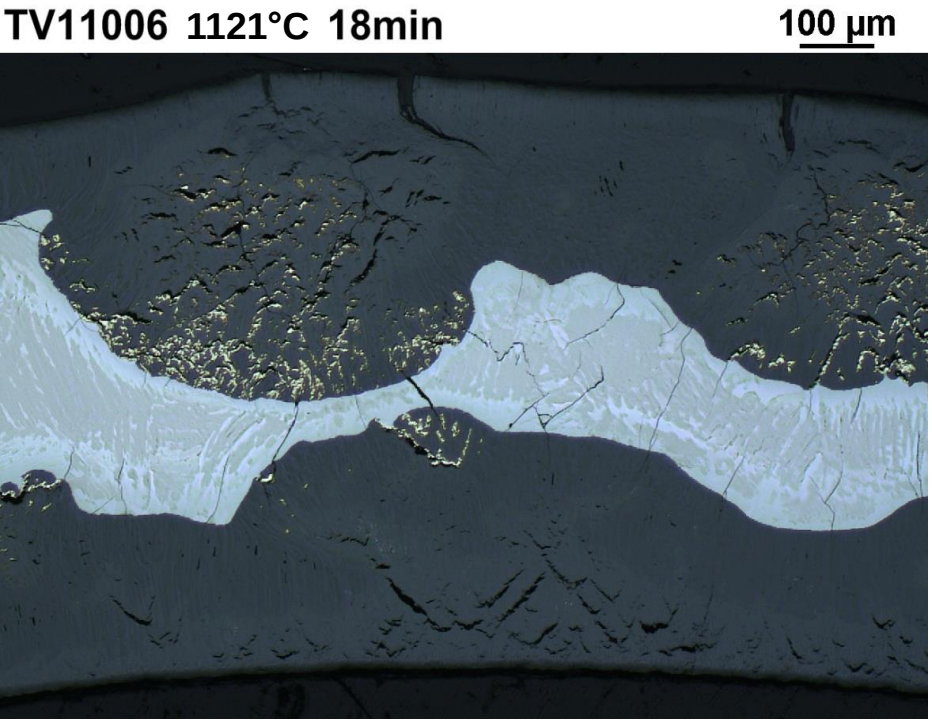
Pre-oxidation effect

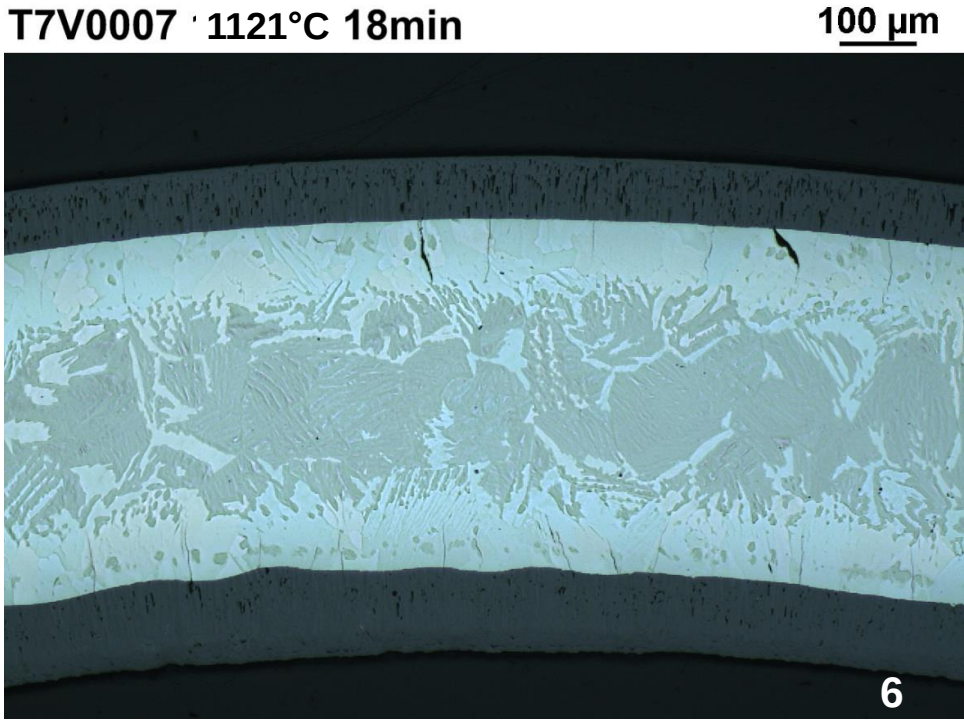




As-received sample

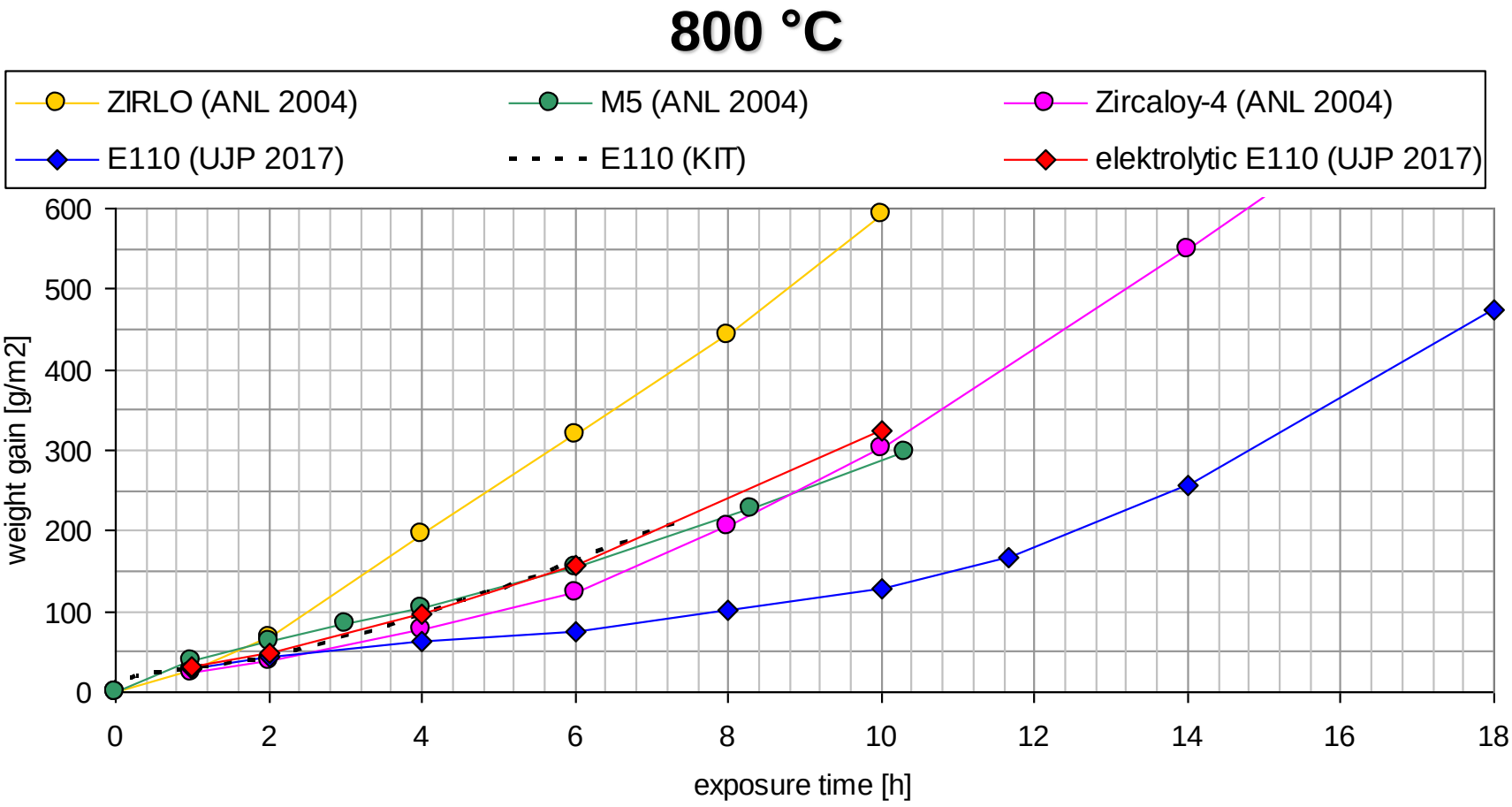
Pre-oxidized sample





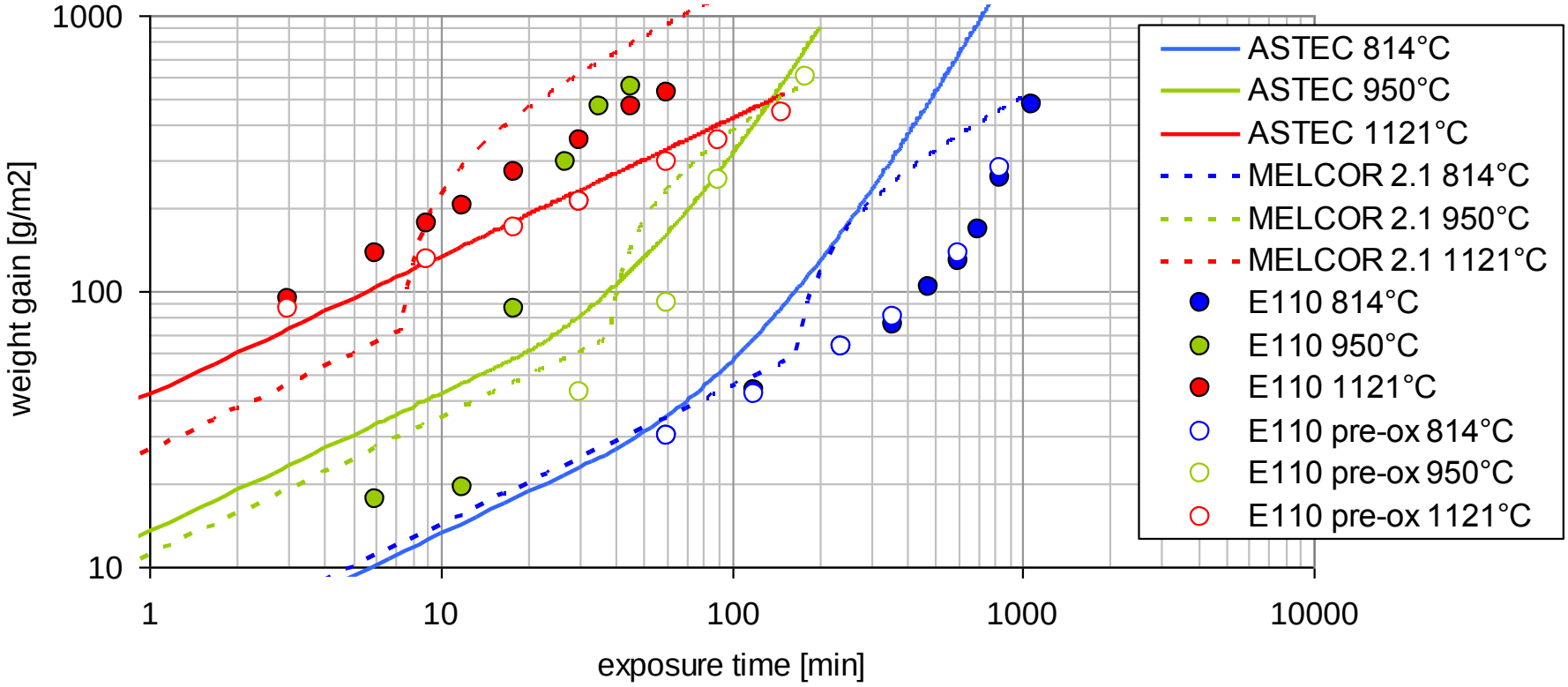
Comparison with other alloys

- E110 similar to M5 for 700 °C
- E110 the lowest weight gain among compared alloys



Comparison with models

- Models ASTEC, MELCOR 2.1 for Zircaloy-4 → disagreement with experiment

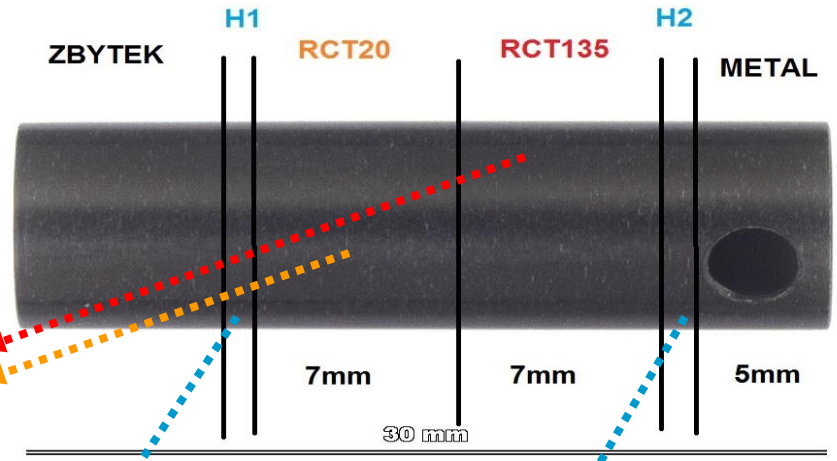


Post-experimental examination





Ring compession test at 20 °C, 135 °C



ZBYTEK: remaining part



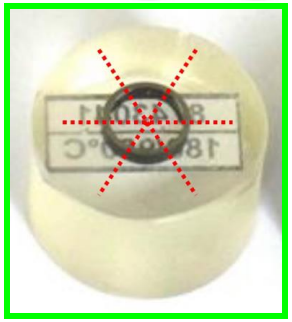
Hydrogen content measurements

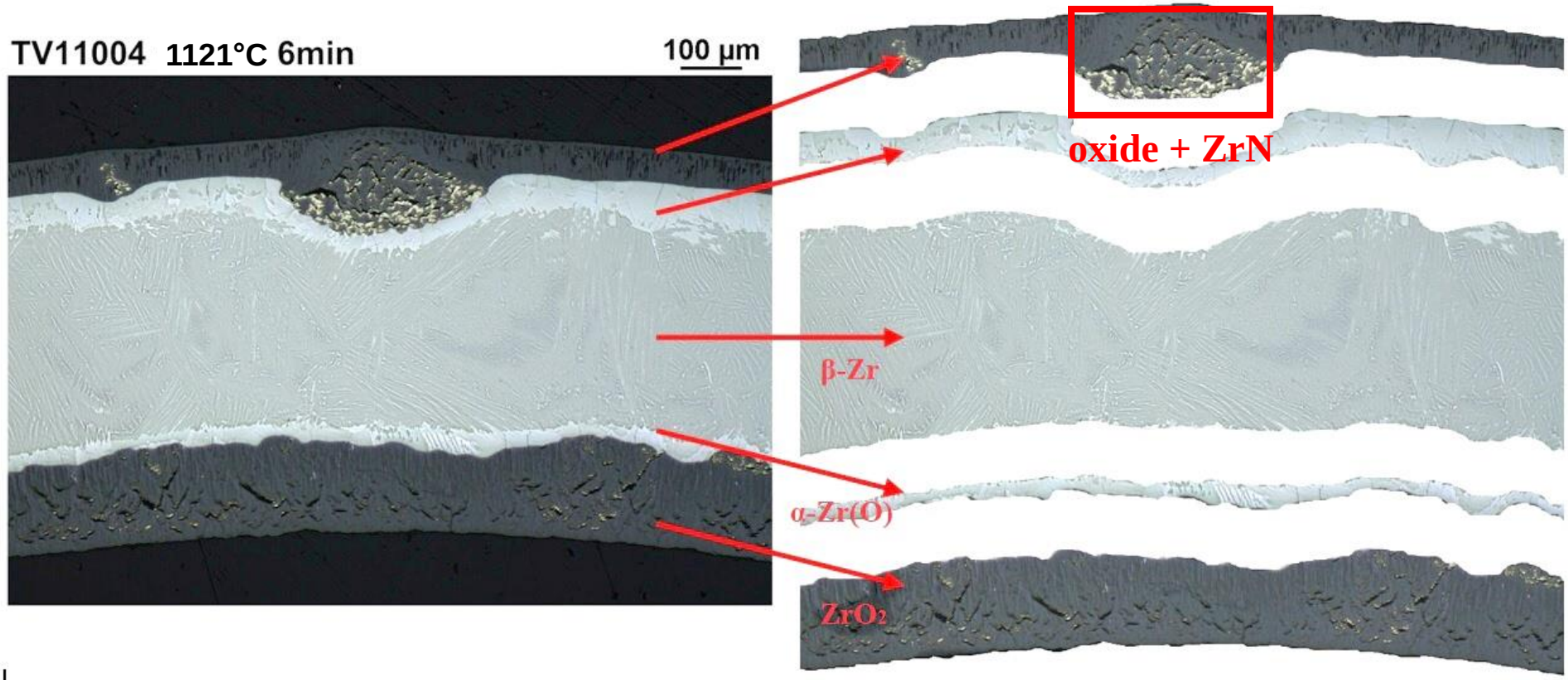


Metalography and microhardness evaluation

Microsture evaluation

- Thickness of oxide (outer, inner), α-Zr(O), β-Zr layer
- Thicknesses were analyzed in 6 directions

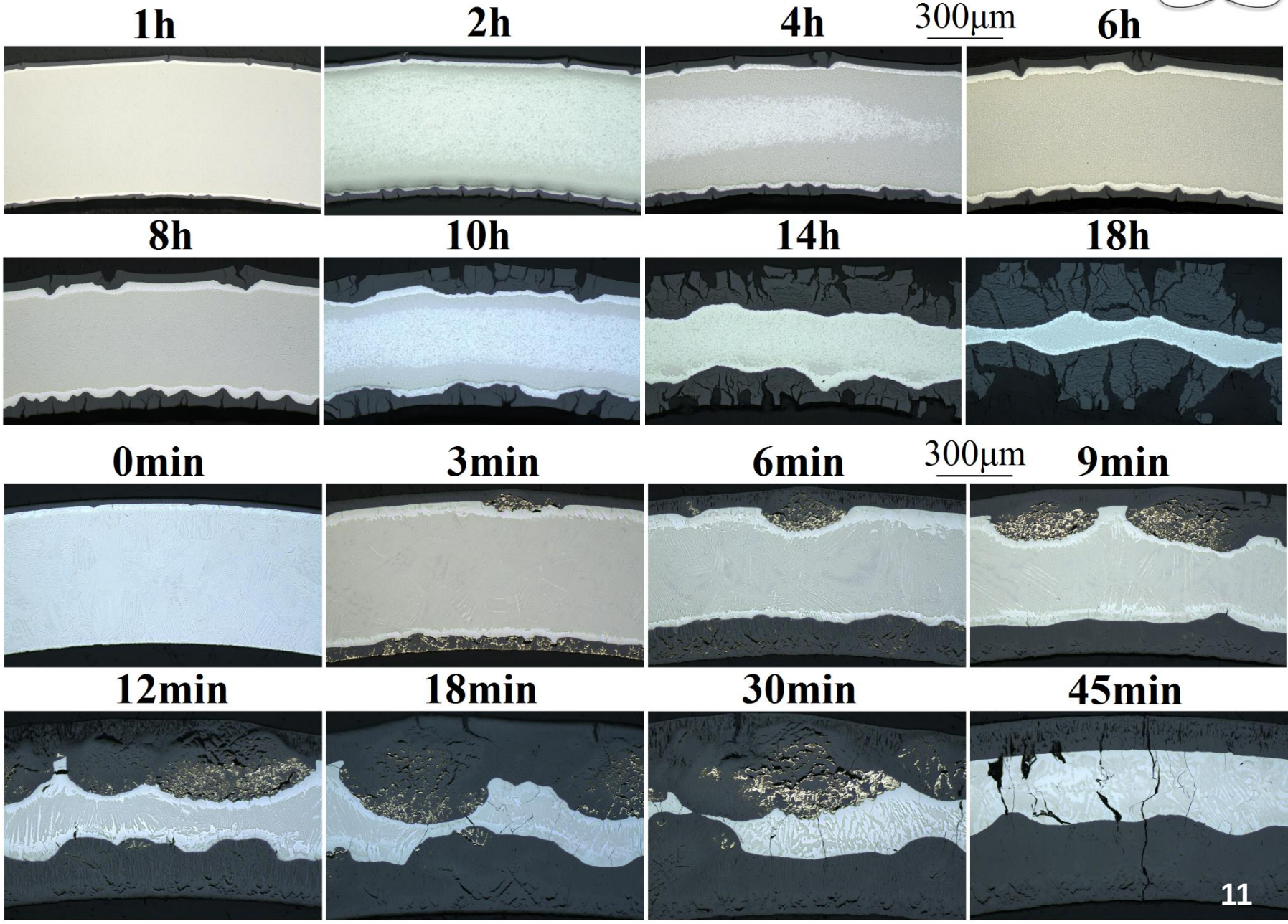




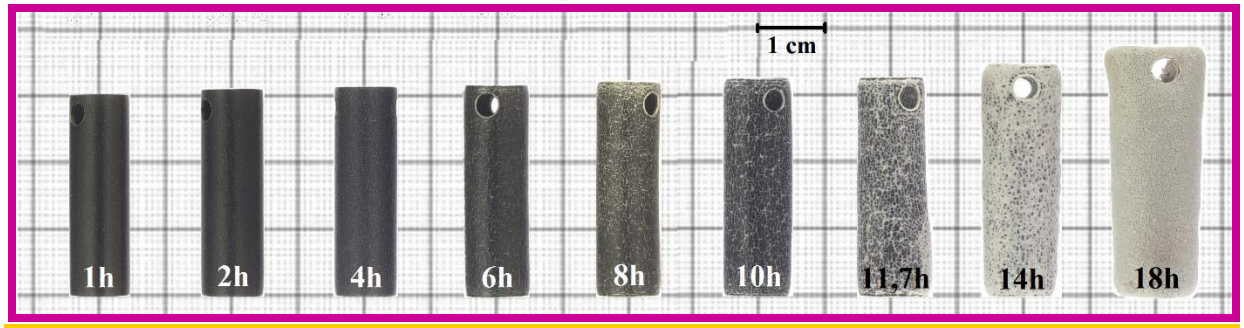
Microstructure

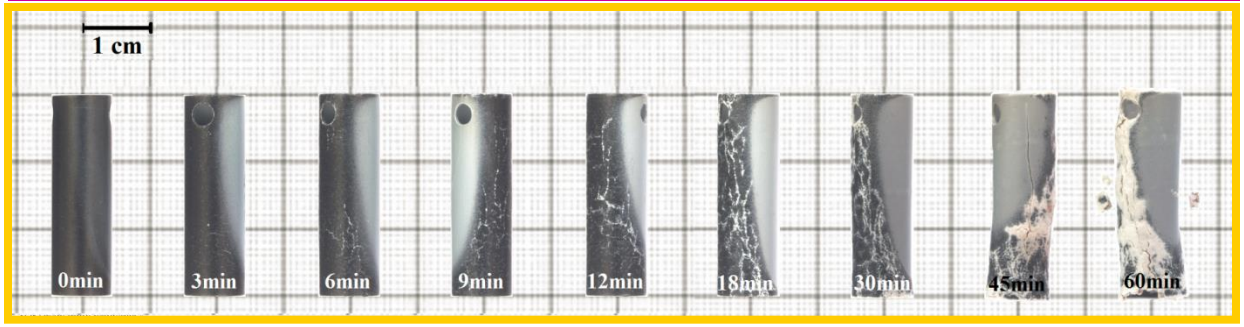
814 °C

1121 °C

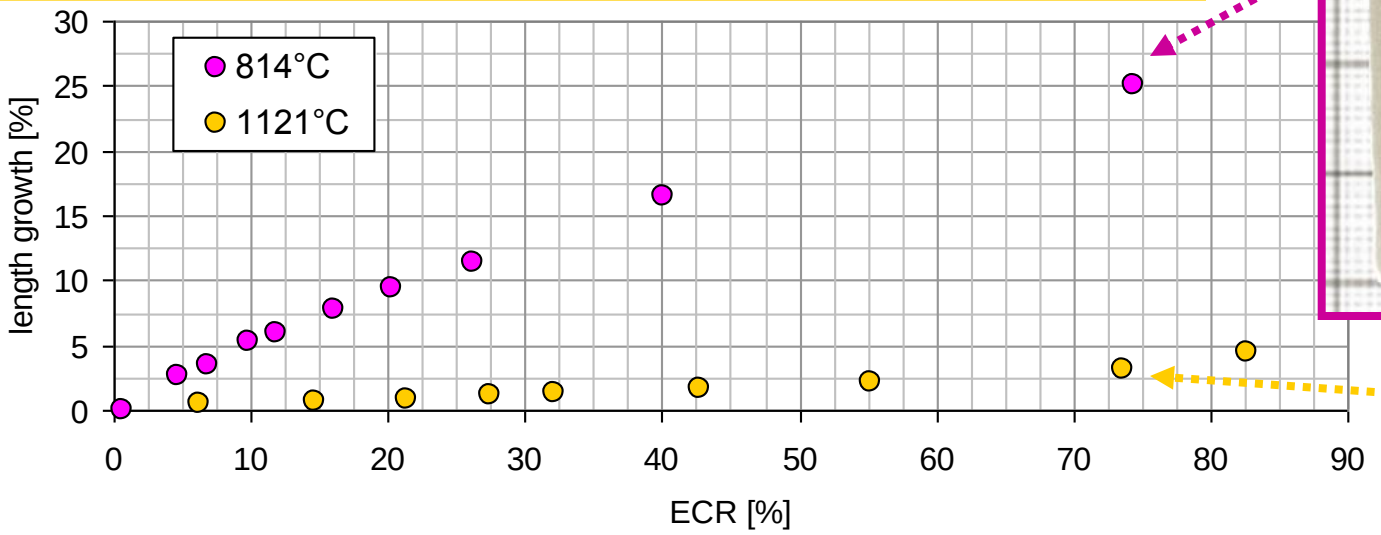


Visual inspection





- Significant length growth for lower temperatures

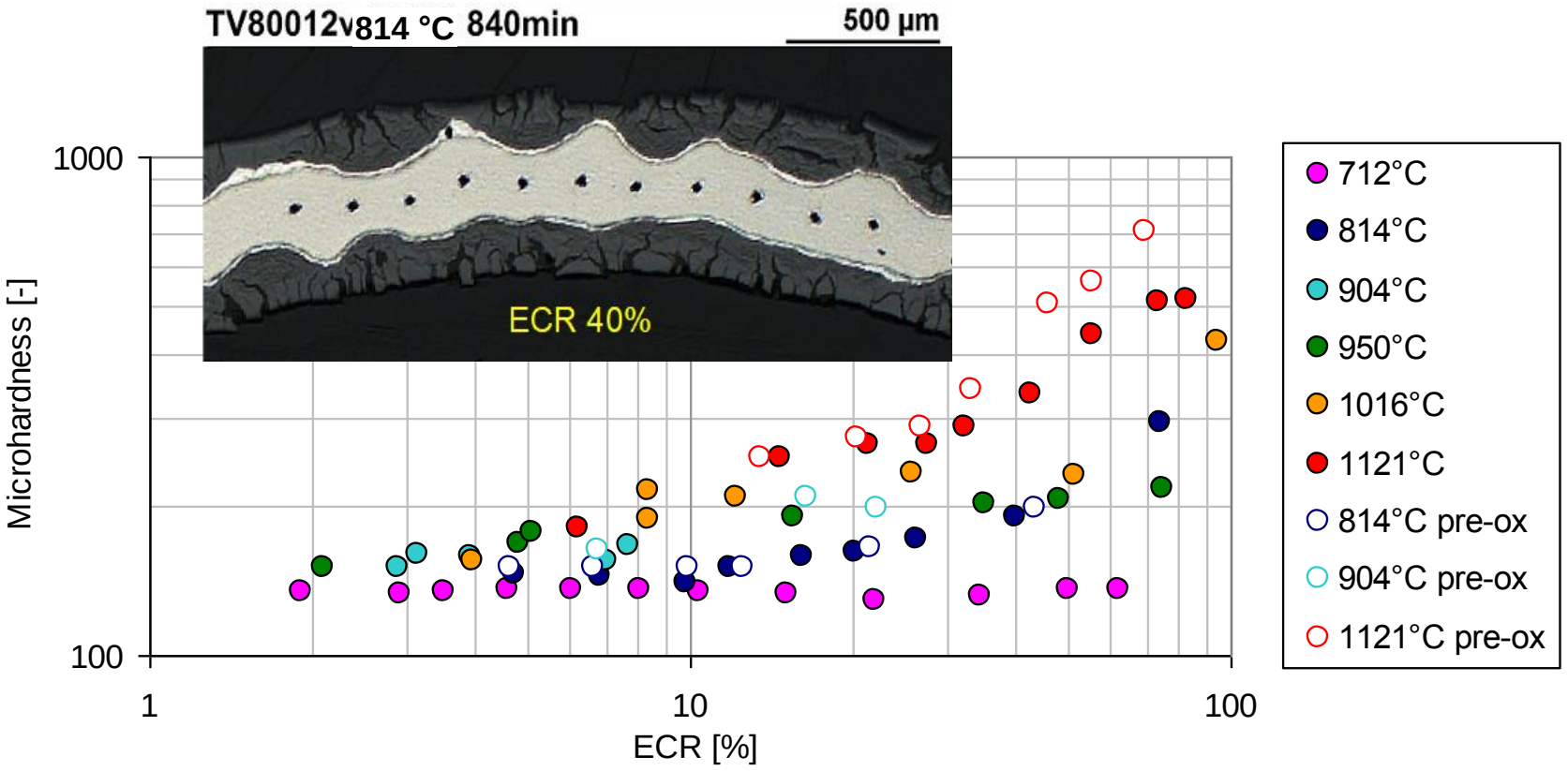




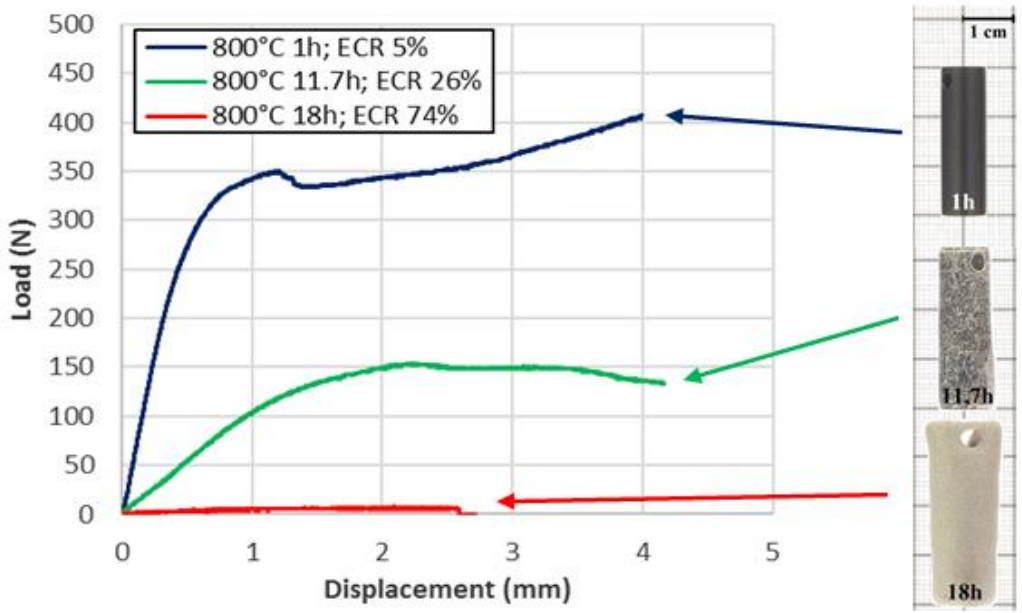


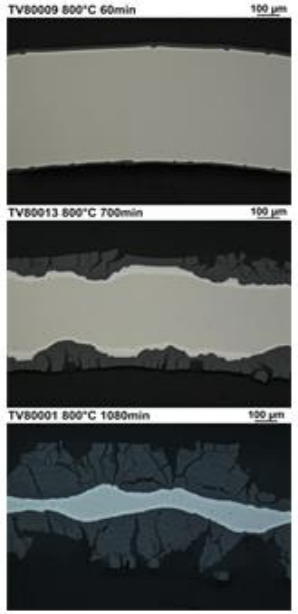
Microhardness evaluation

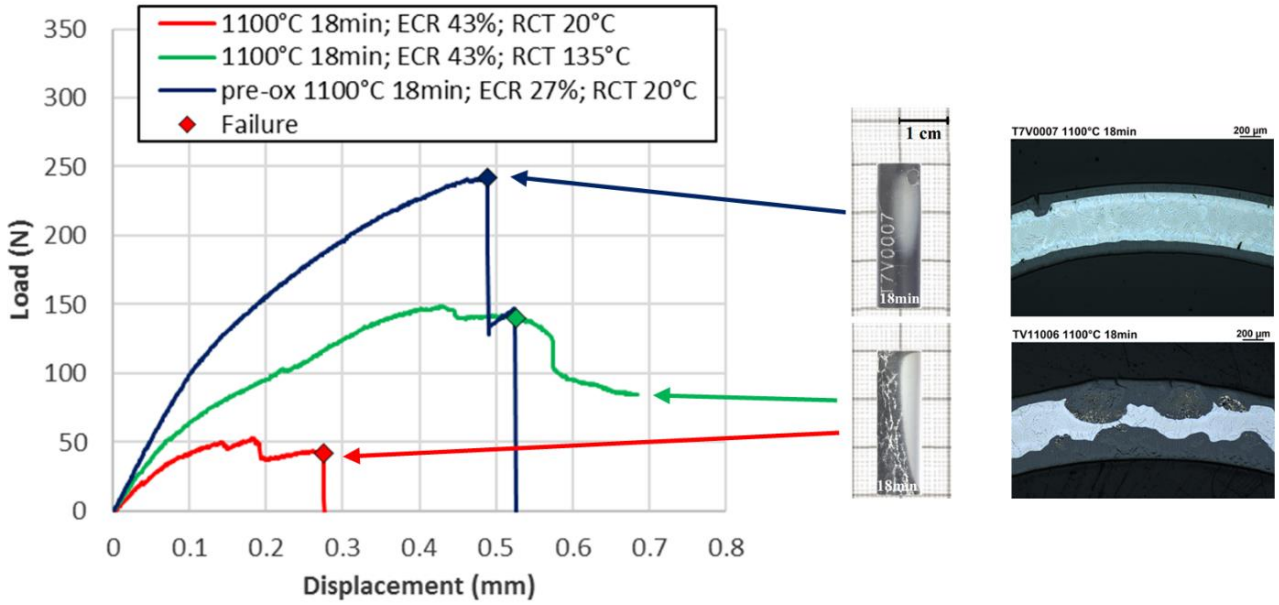
- Vickers hardness testing by load 100g in 2x10 points in the middle of the sample wall
- Microhardness increases with increasing temterature



Ring compression test

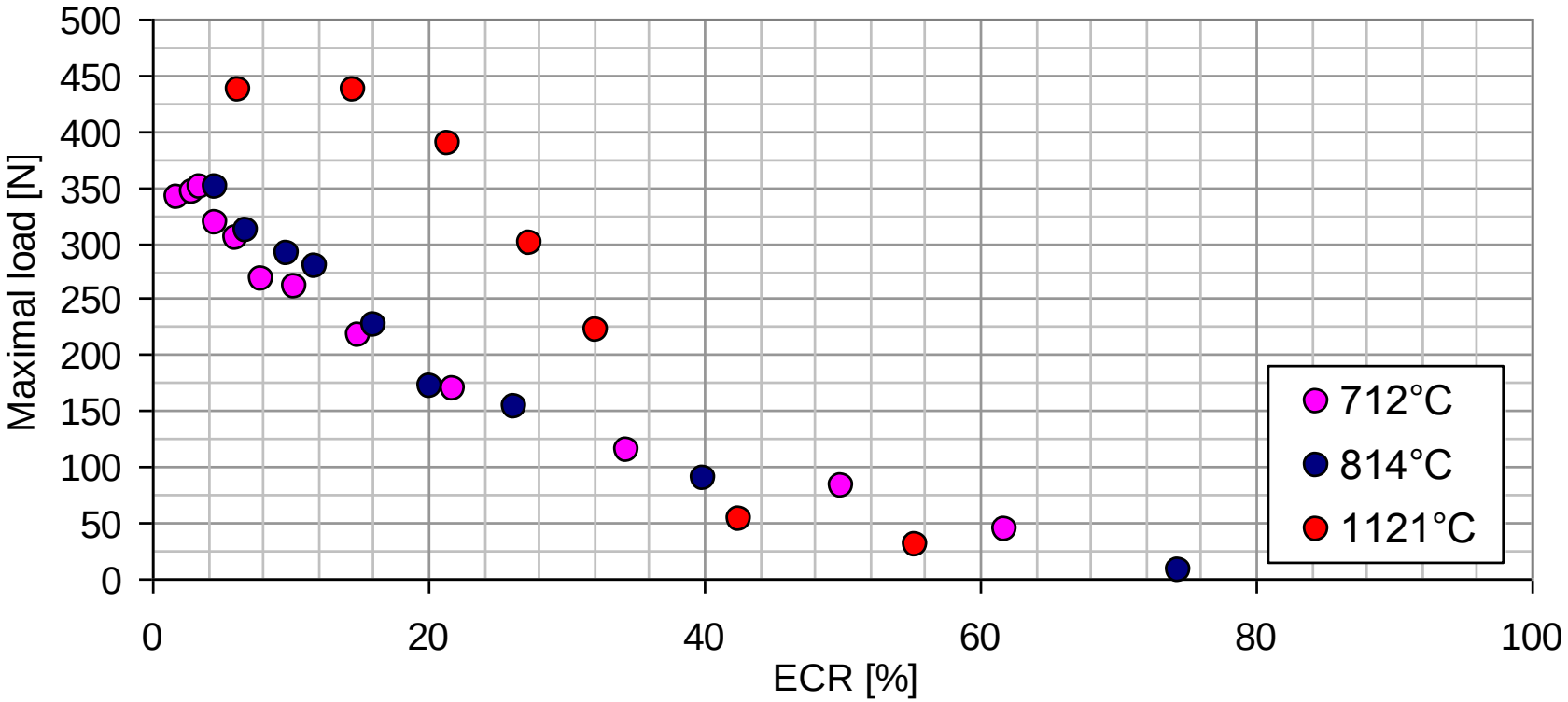






Ring compression test

- Maximal load decreases with increasing ECR
- Ductility decreases with increasing temperature



Conclusion

- Local attack of nitrogen
- Needle-shaped grain of α-Zr(O) across of wall for longer exposure time
- Pre-oxidized samples have lower weight gain than as-received ones
- E110 has the lowest weight gain for 700, 800 °C and for 1100, 1200 °C
- Disagreement E110 with codes ASTEC and MELCOR 2.1
- Axial length growth depending on temperature
- Microhardness increases with temperature and exposure time → lower ductility
- Cladding strength decreases

Future work

- Other variants of pre-oxidation:
  - **steam:**
    - 425 °C/10,7 MPa
    - (**5**;10;20) μm
  - **water (WWER chemistry):**
    - 360 °C/19,7 MPa
    - (5;10;…) μm
  - **High temperature pre-oxidation** („LOCA + air ingess“):
    - 800–1200 °C/0,1 MPa
- Develop correlations for E110
- High temperature oxidation in steam + air environment

Thanks for your attention!





UJP PRAHA

23th International QUENCH-workshop 2017

Variation in time of the high temperature oxidation of zirconium alloys in various atmospheres

Abstract

This presentation is about the oxidation of zirconium alloys at high temperatures in various atmospheres with special focus on the time dependence of the oxide/nitride formation to show which reactions could happen during an air ingress scenario in reactors or in spent fuel pools.

Therefore test series were made with 2 cm Zircaloy-4 cladding tube segments at three different temperatures, 900 °C, 1000 °C and 1100 °C and in four different atmospheres: Steam, synthetic air, nitrogen with steam and synthetic air with steam. These tests were made in a tube furnace coupled to a mass spectrometer.

The mass gain of the samples is shown in three diagrams. Nitrogen is significantly accelerating the oxidation of zirconium.

For the temperature 1000 °C macro- and micrographs of the samples in all atmospheres are shown. They prove that the nitrogen attack starts locally on the oxide scale surface.

Another diagram shows the gas release during the reaction of Zircaloy-4 with a nitrogen and steam atmosphere. It can be observed that there is a significant hydrogen formation after the transition time, which is caused by the nitrogen effect.

All results are finally summarized in the 19th slide.



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Variation in time of the high-temperature oxidation of zirconium alloys in various atmospheres

P. van Appeldorn. M. Steinbrück. U. Stegmaier

23th Int. QUENCH Workshop. Karlsruhe Institute of Technology. 17-19 October 2017

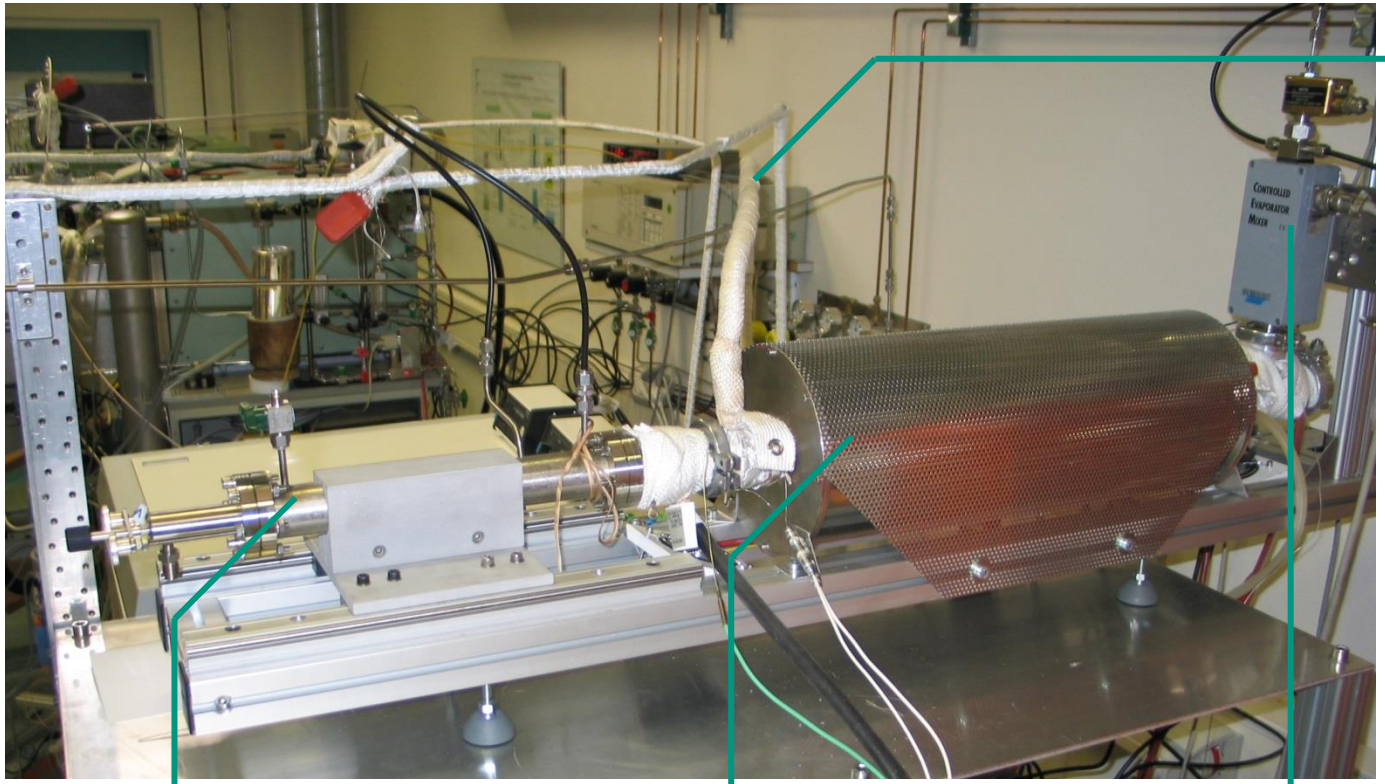
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Motivation

- Air ingress scenarios in reactors and spent fuel pools are worldwide under discussion
- Strong effect of nitrogen on oxidation mechanism and kinetics is known
- Investigation of the dependence in time of the oxide scale growth during oxidation of Zircaloy-4 in various atmospheres containing nitrogen and, for comparison, in steam.

Experimental setup



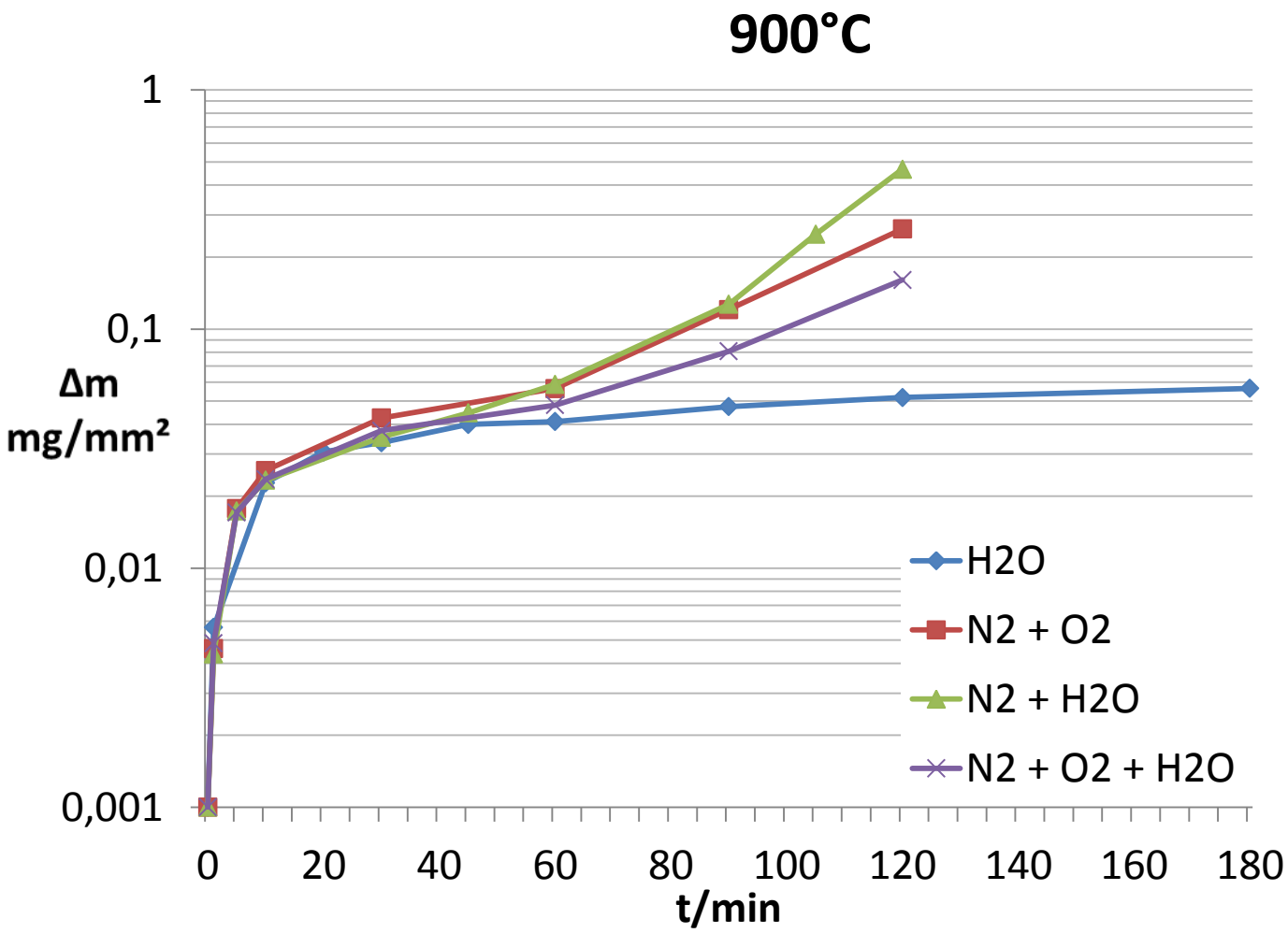
Samples, test matrix and test conduct

- 2 cm cladding tube segments made of Zircaloy-4



- Temperatures: 900°C;1000°C;1100°C
- 8-10 tests for each condition with different test time
- Reaction gases with flow rate:
    1. **H_2O**: 16.1 g/h
    2. **Synth. air:** 50 l/h (N_2: 80% + O_2: 20%)
    3. **N_2**: 40 l/h **+ H_2O**: 16.1 g/h
    4. **Synth. air**: 40 l/h (N_2: 80% + O_2: 20%) **+ H_2O**: 16.1 g/h
- Protective gas: argon 20 l/h

Mass change in different atmospheres





H_2O



$N_2 + O_2$

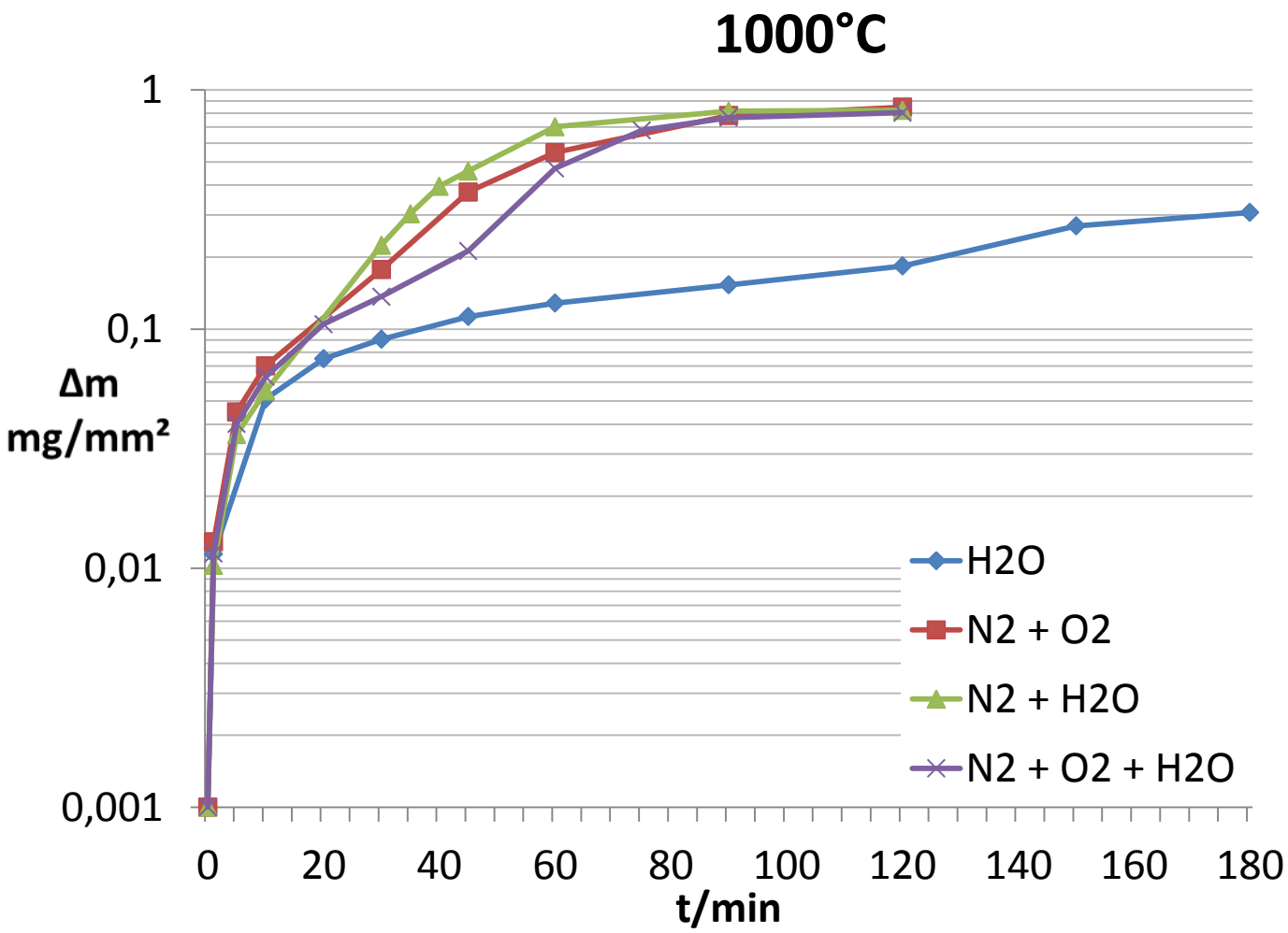


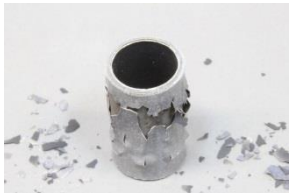
$N_2 + H_2O$



$N_2 + O_2 + H_2O$

Mass change in different atmospheres





H_2O



$N_2 + O_2$

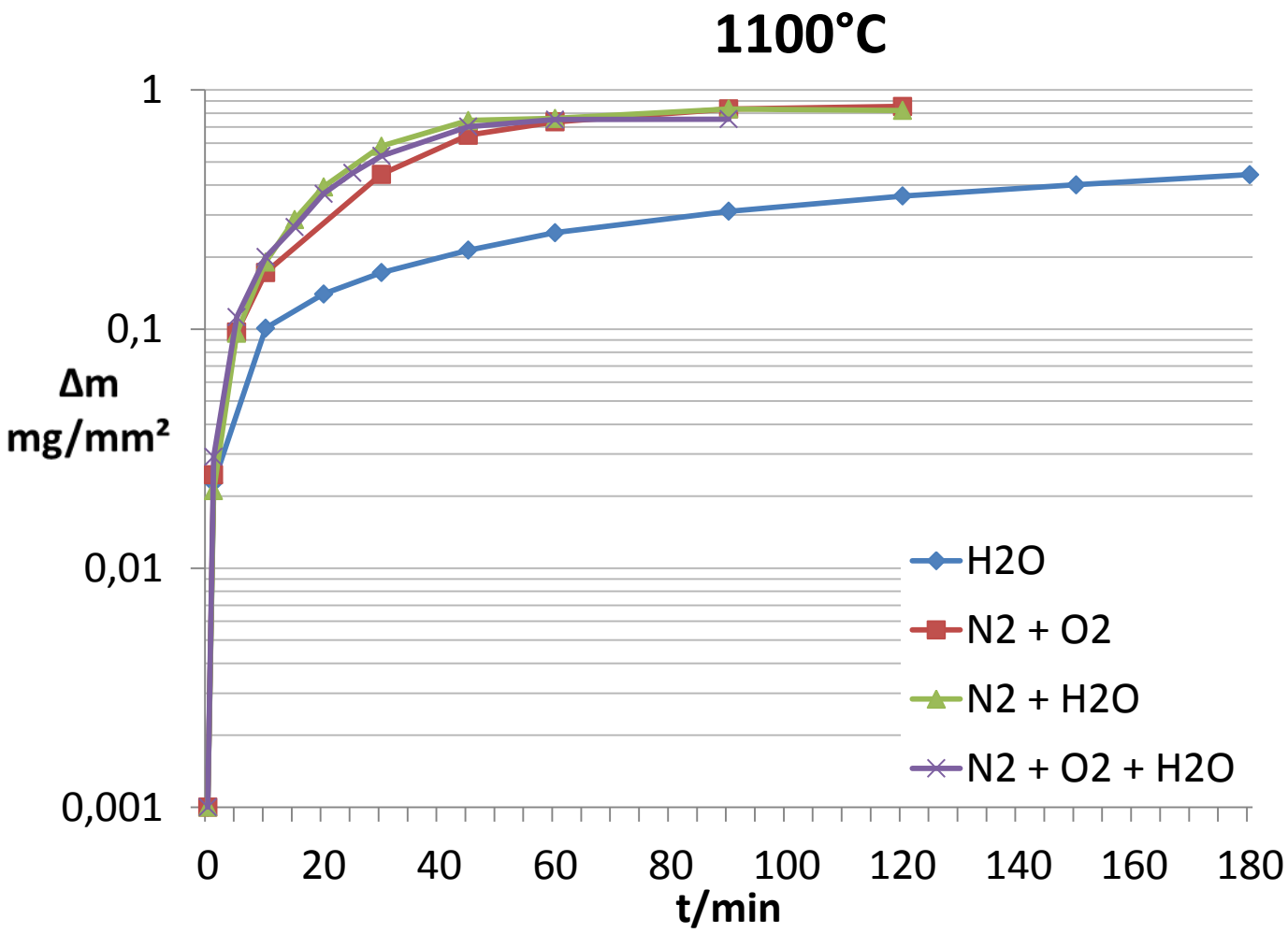


$N_2 + H_2O$



$N_2 + O_2 + H_2O$

Mass change in different atmospheres





H_2O



$N_2 + O_2$

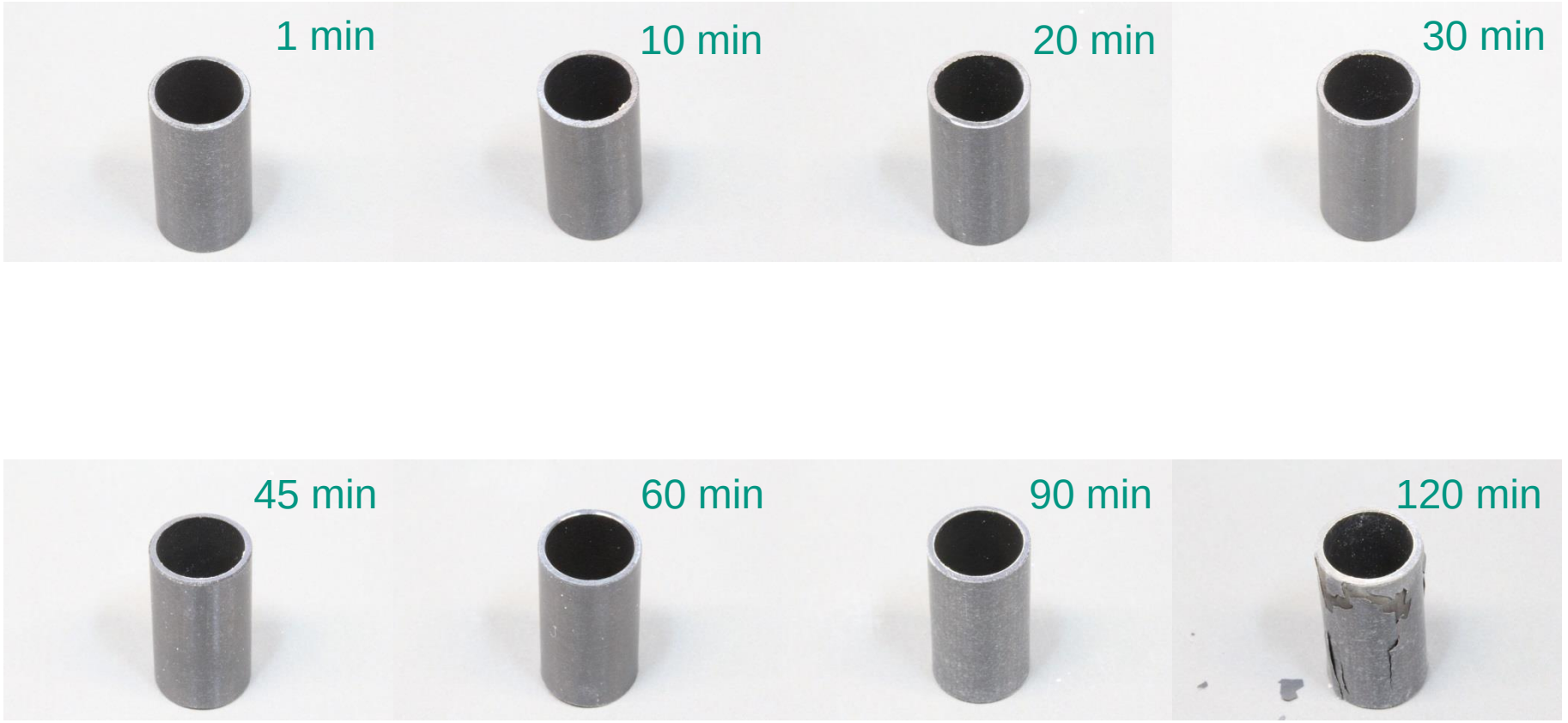


$N_2 + H_2O$

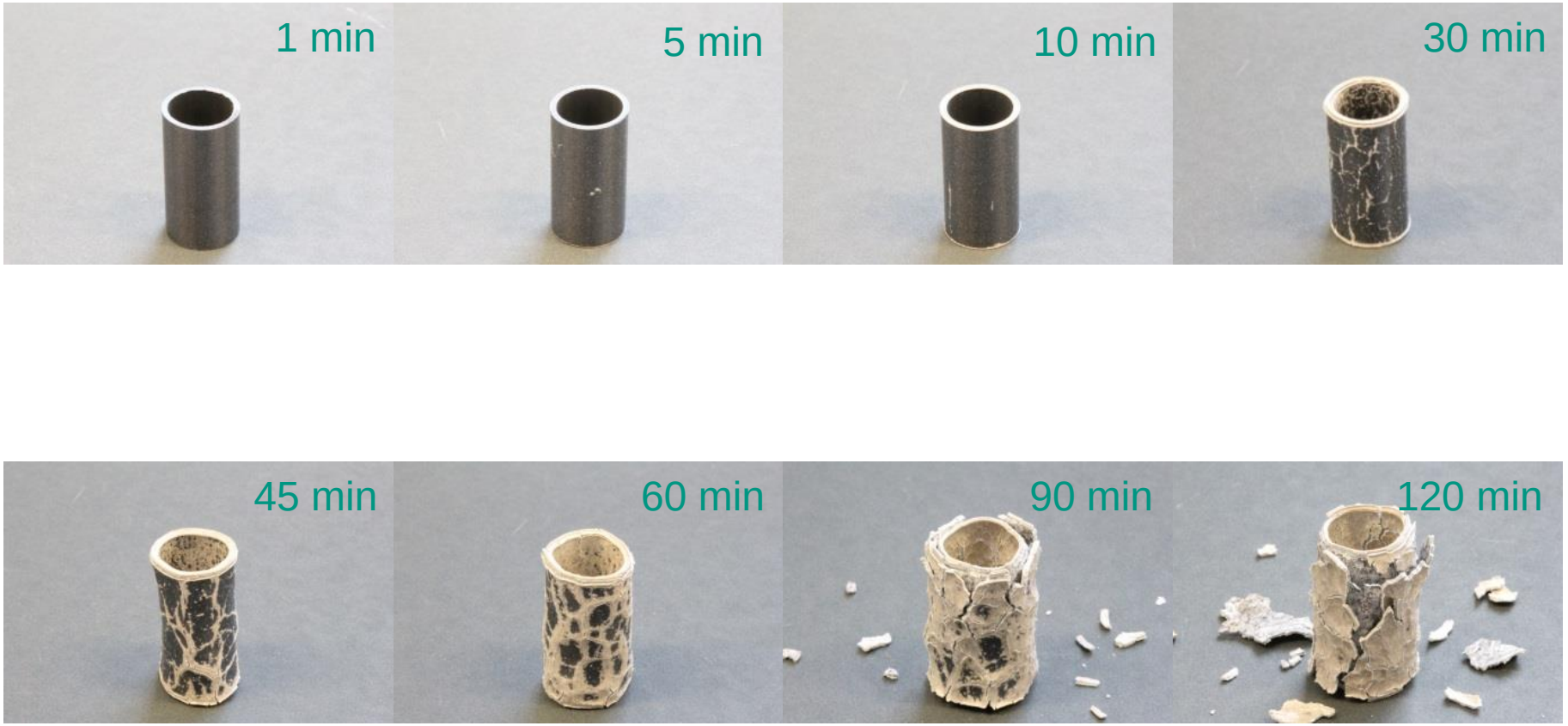


$N_2 + O_2 + H_2O$

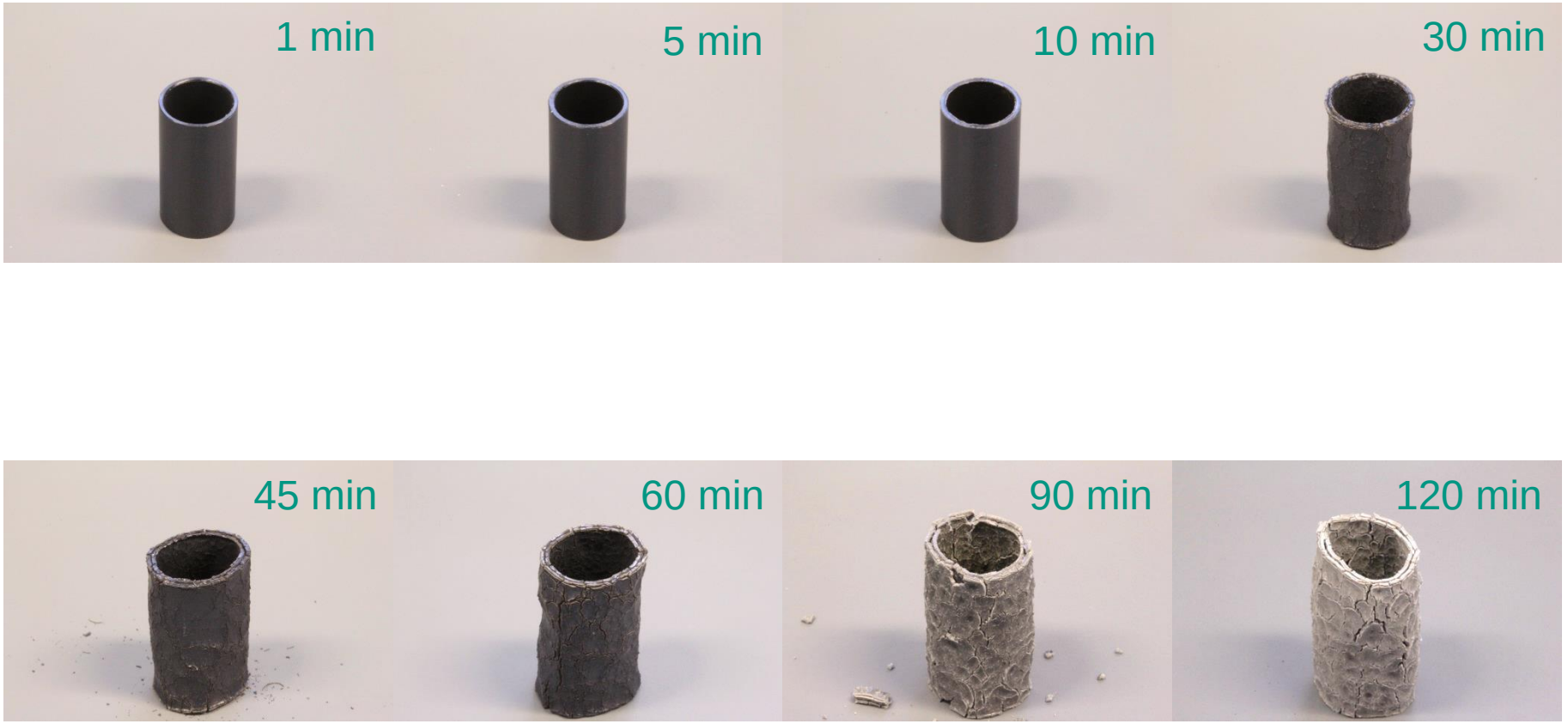
Time dependent reaction with steam at 1000°C



Time dependent reaction with synth. air at 1000°C



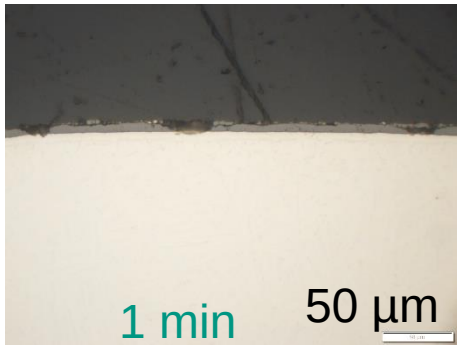
Time dependent reaction with nitrogen-steam at 1000°C

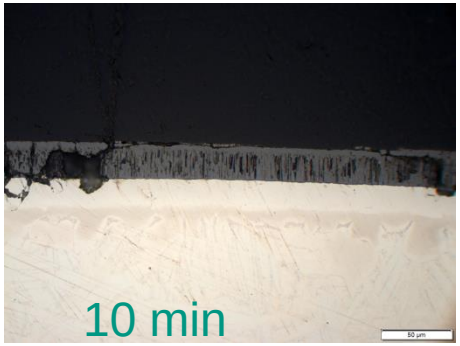


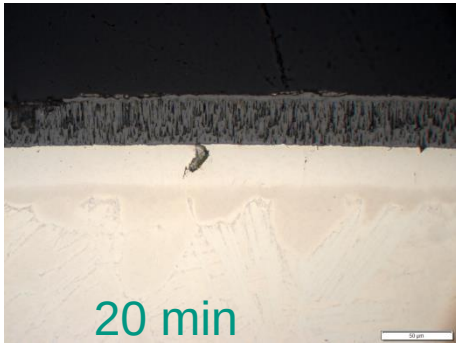
Time dependent reaction with synth. air-steam at 1000°C

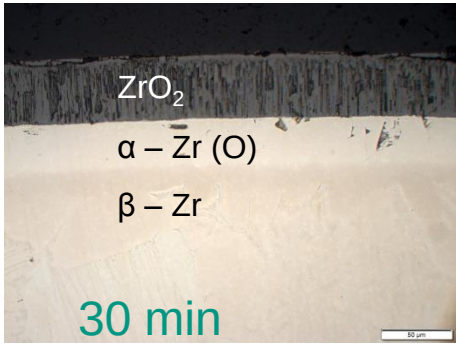


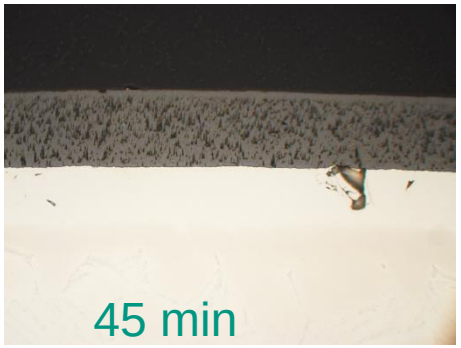
Micrographs of 1000°C samples in steam

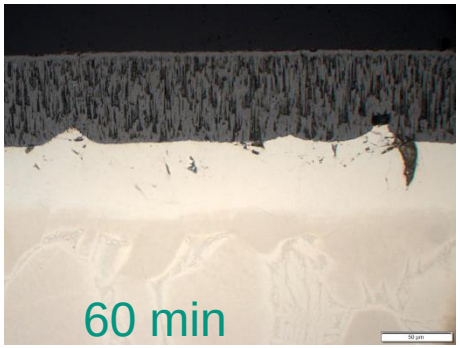


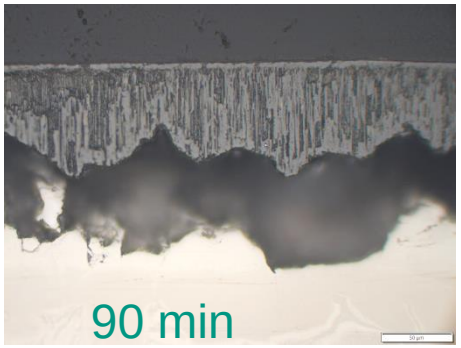


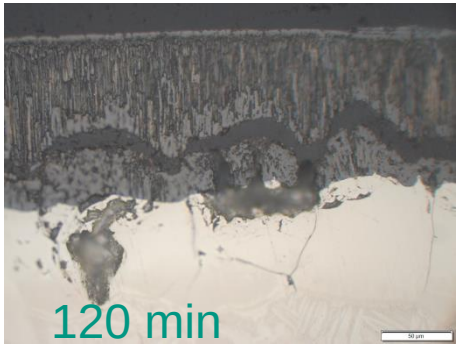




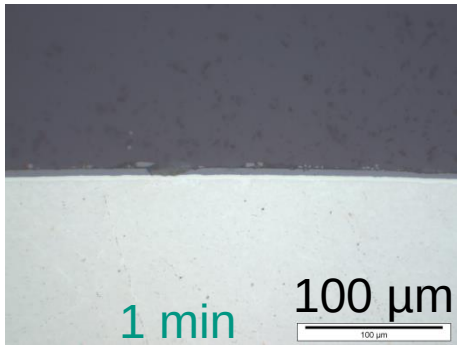


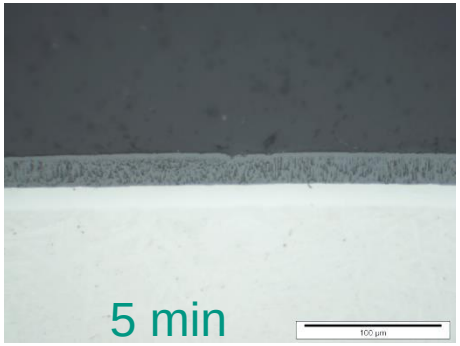


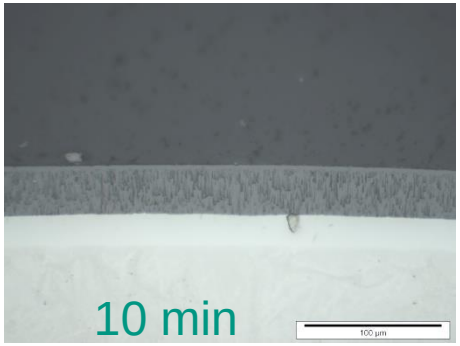


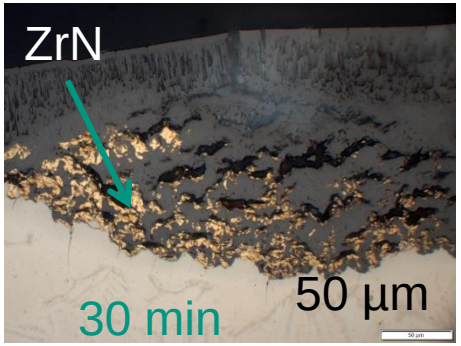


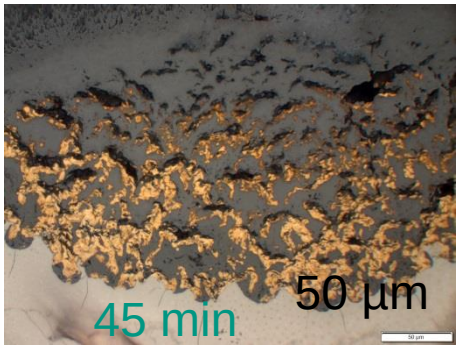
Micrographs of 1000°C samples in synth. air

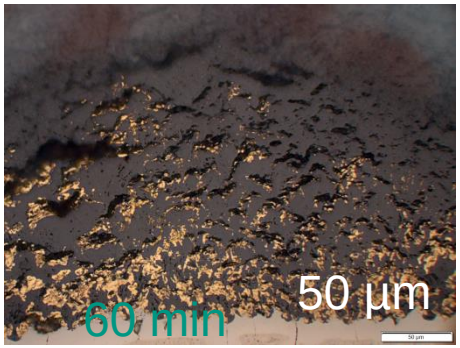


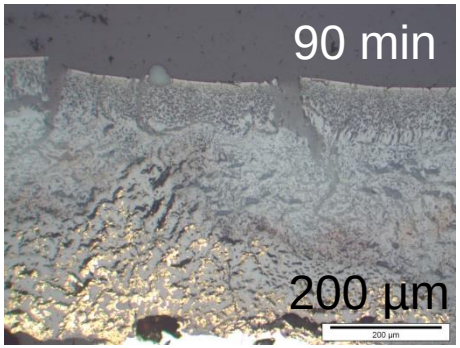


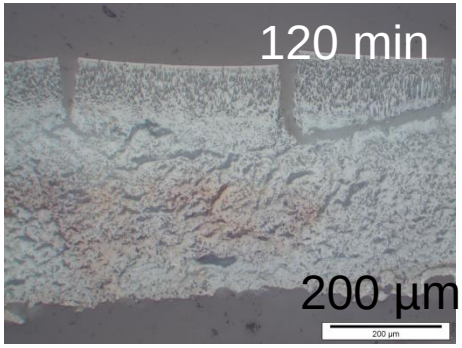




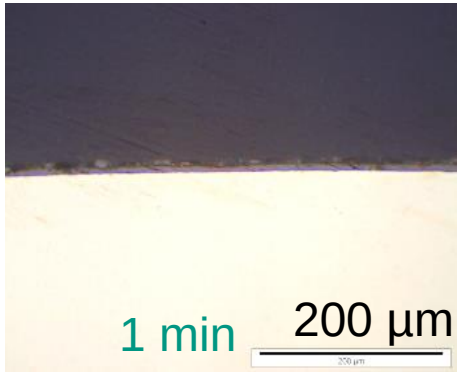


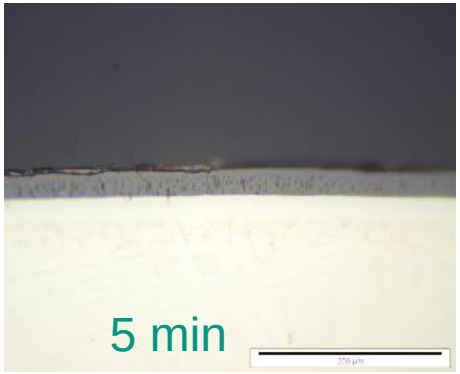


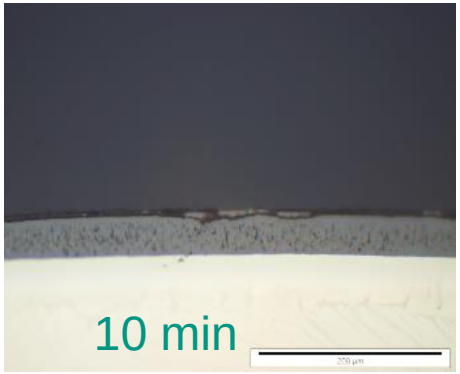


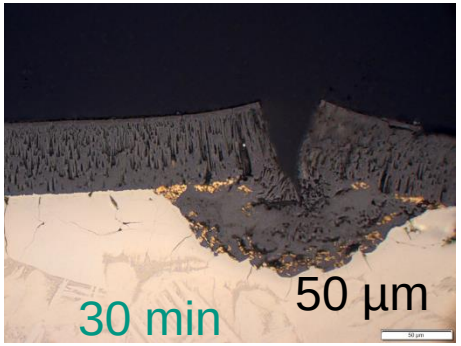


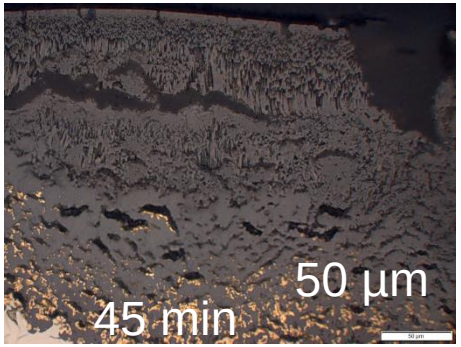
Micrographs of 1000°C samples in nitrogen - steam

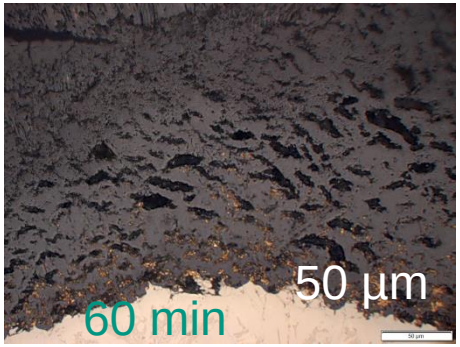


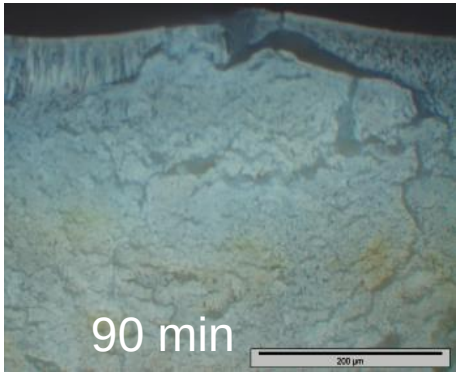


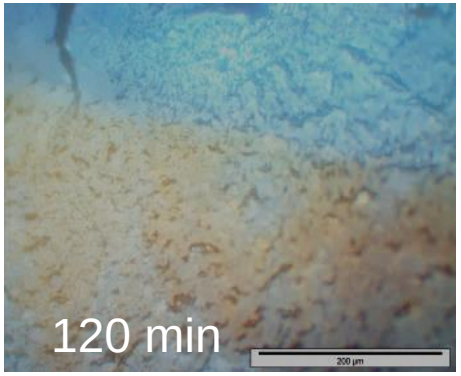




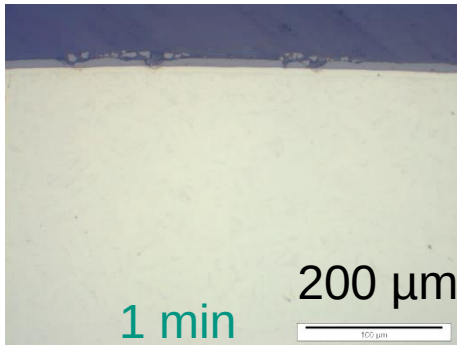


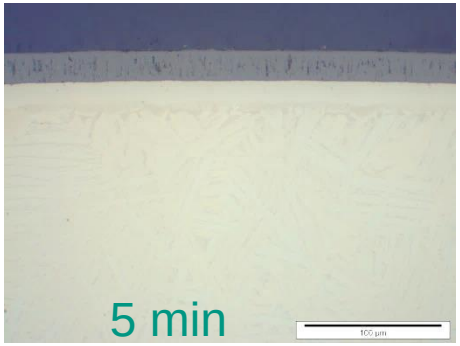


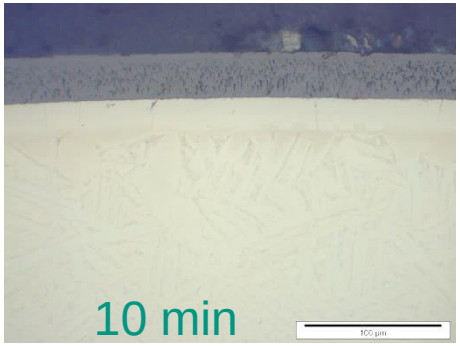


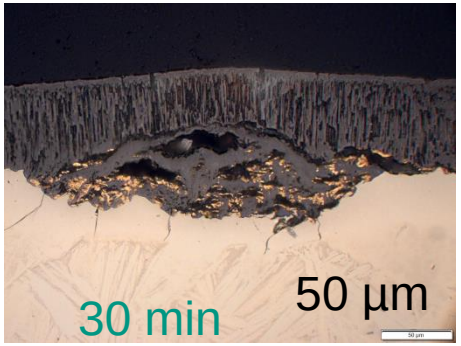


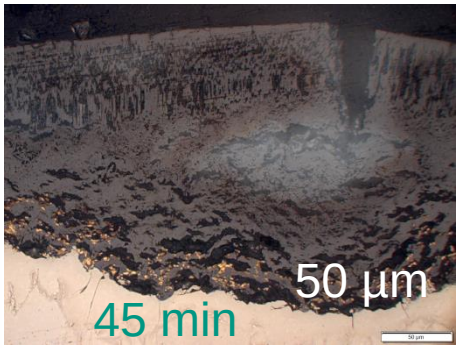
Micrographs of 1000°C samples in synth. air - steam

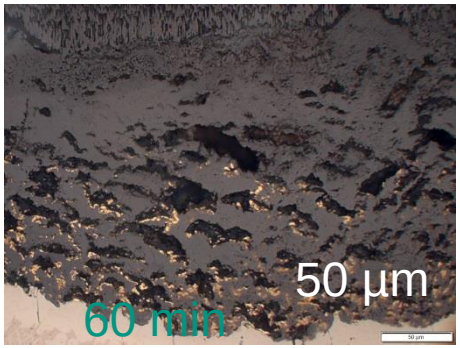


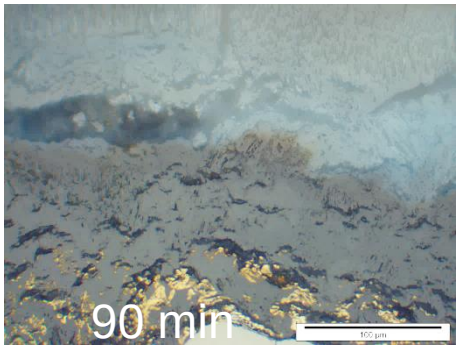


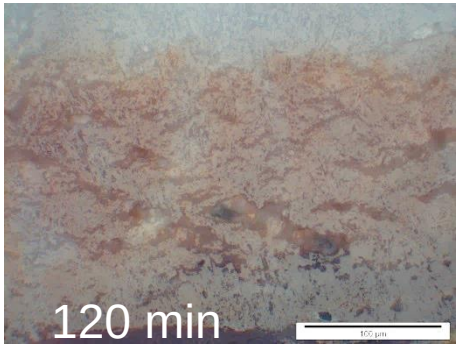




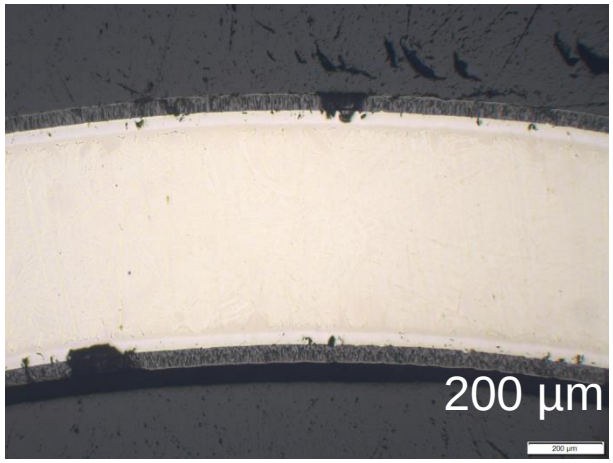




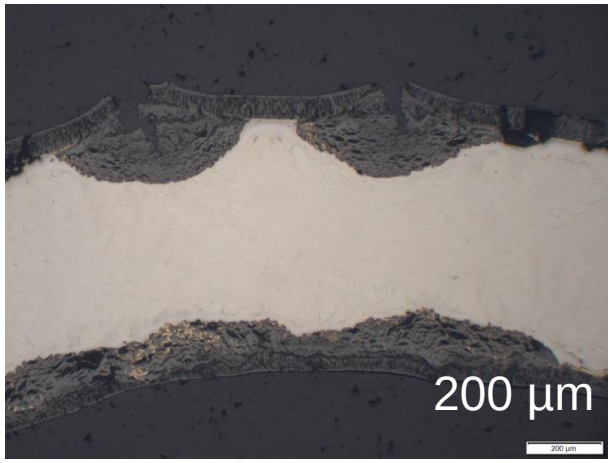




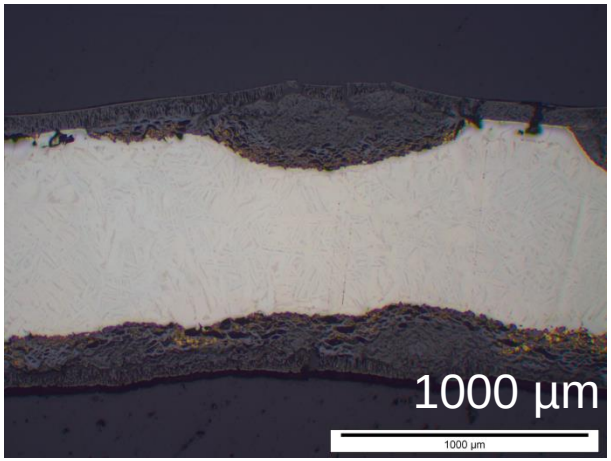
Micrographs of the 30 min samples at 1000°C



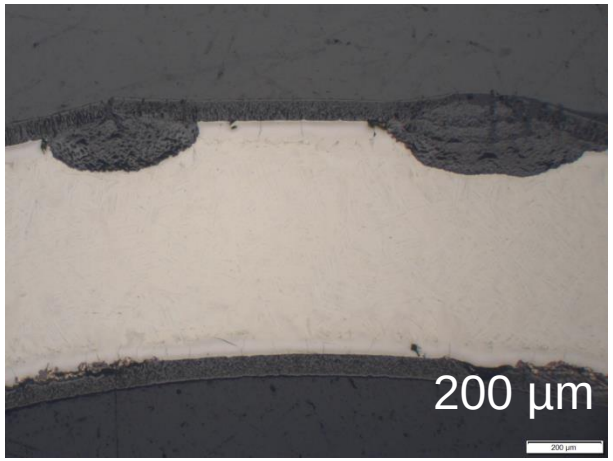
H_2O



$N_2 + O_2$

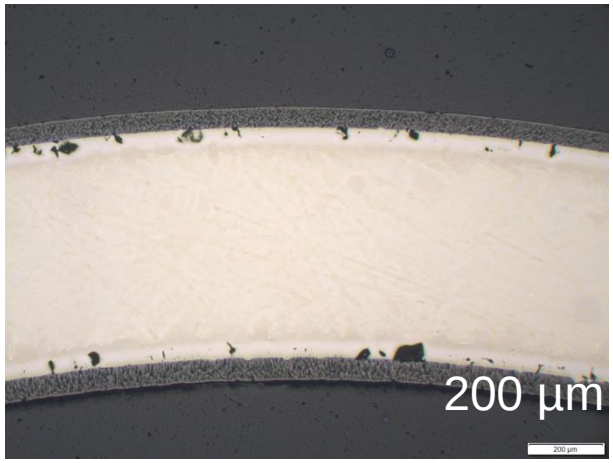


$N_2 + H_2O$

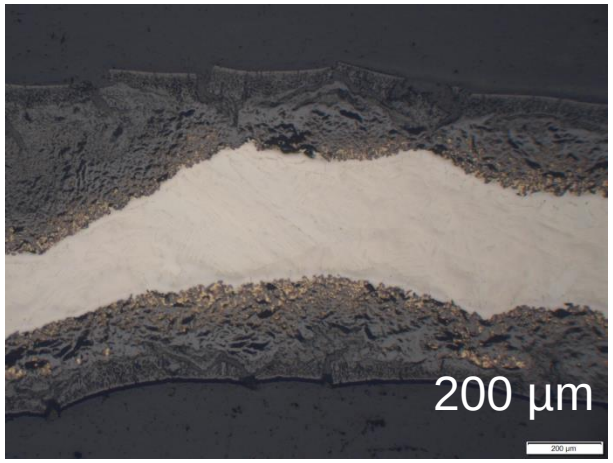


$N_2 + O_2 + H_2O$

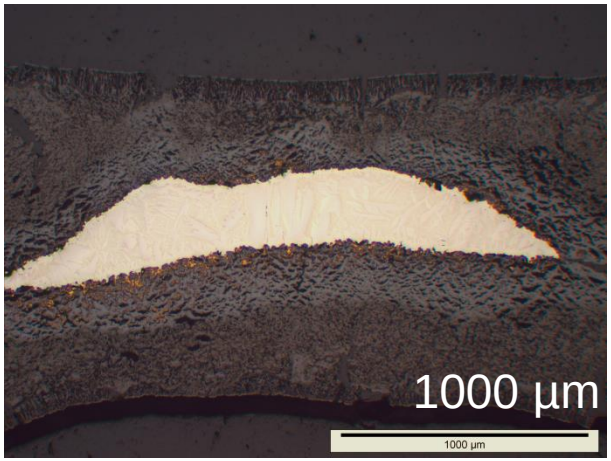
Micrographs of the 45 min samples at 1000°C



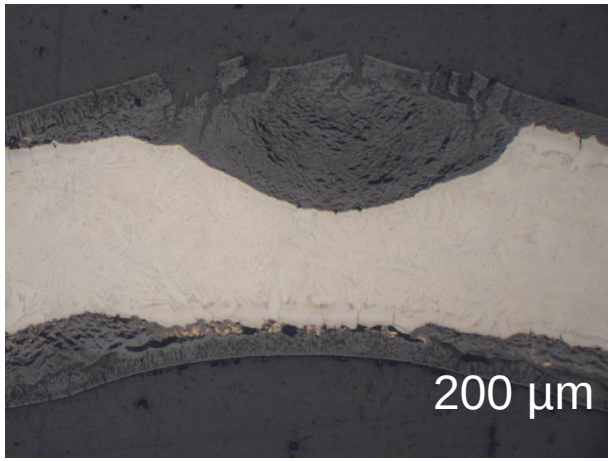
H_2O



$N_2 + O_2$

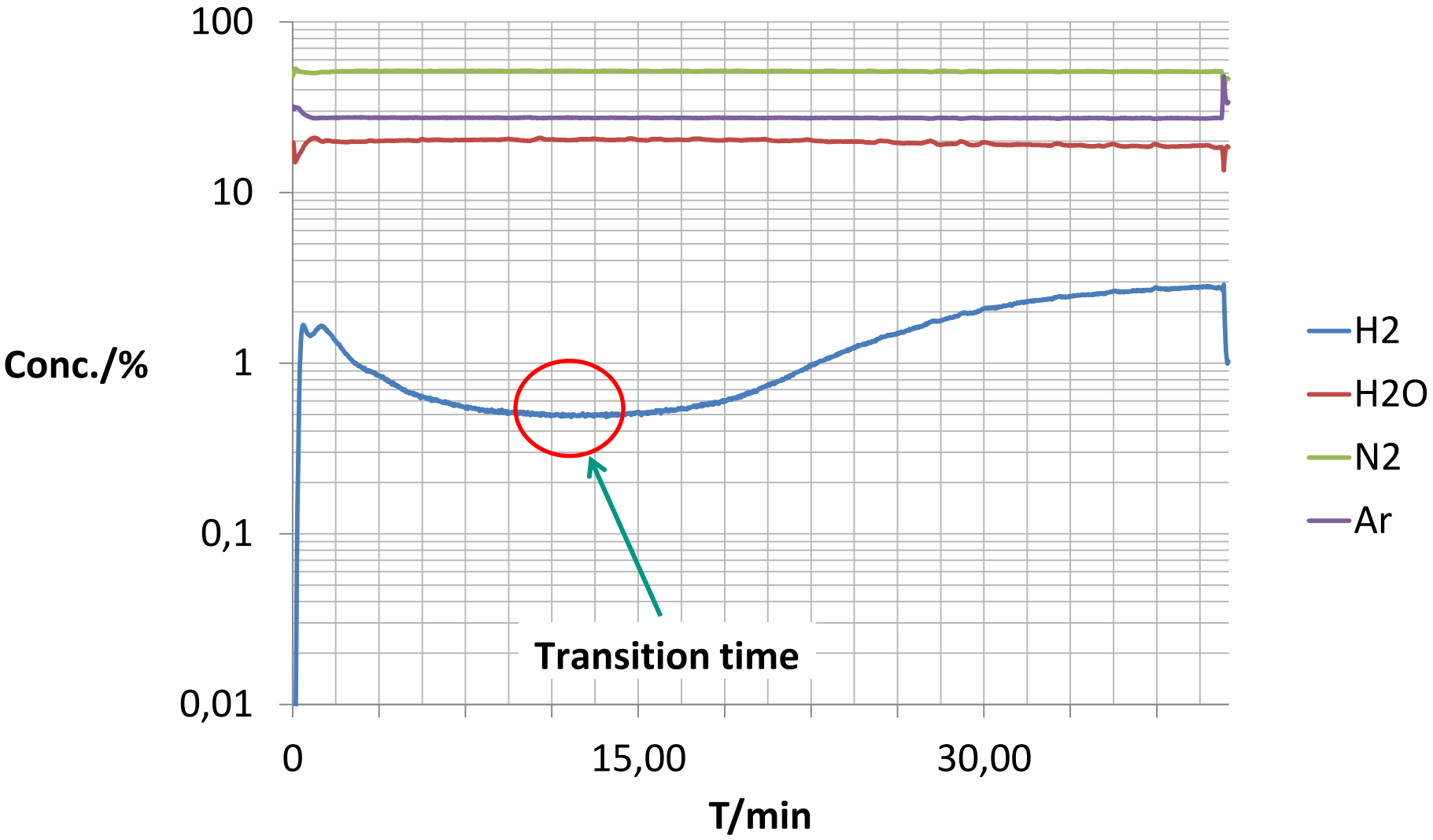


$N_2 + H_2O$



$N_2 + O_2 + H_2O$

Hydrogen formation at 1000°C in nitrogen - steam



Summary

- 124 samples have been produced, photographed and metallographically prepared in this test series
- The oxidation kinetics of Zry-4 in atmospheres containing nitrogen is faster compared to the oxidation in pure steam
- Nitrogen attack starts locally
- The oxidation behavior in air, steam - air and steam - nitrogen is similar
- Completely oxidized zirconium alloys at the temperature 1100°C are more stable than samples which were oxidized in 900°C and 1000°C

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Variation in time of the high-temperature oxidation of zirconium alloys in various atmospheres

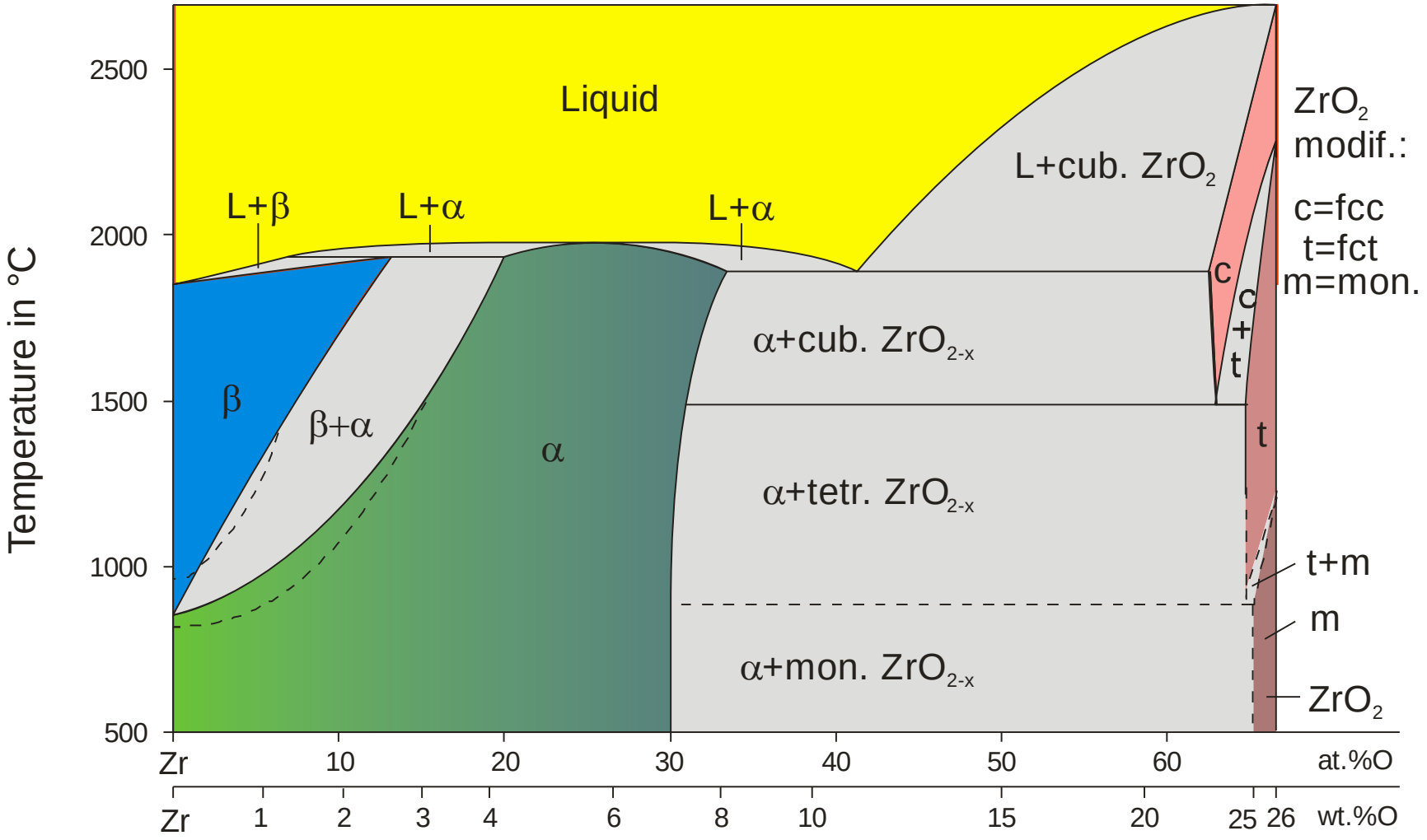
P. van Appeldorn. M. Steinbrück. U. Stegmaier

23th Int. QUENCH Workshop. Karlsruhe Institute of Technology. 17-19 October 2017

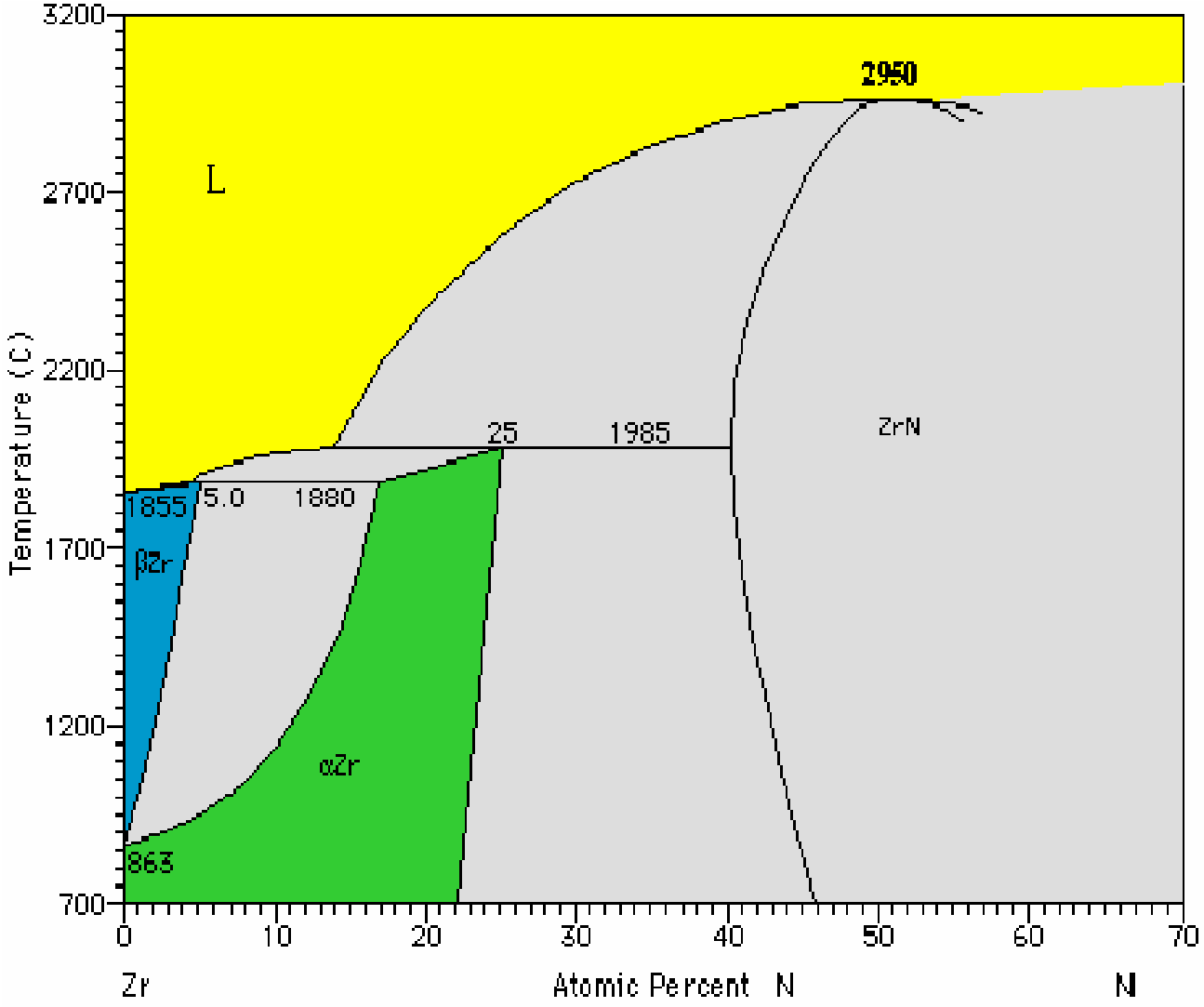
Institute for Applied Materials IAM-AWP



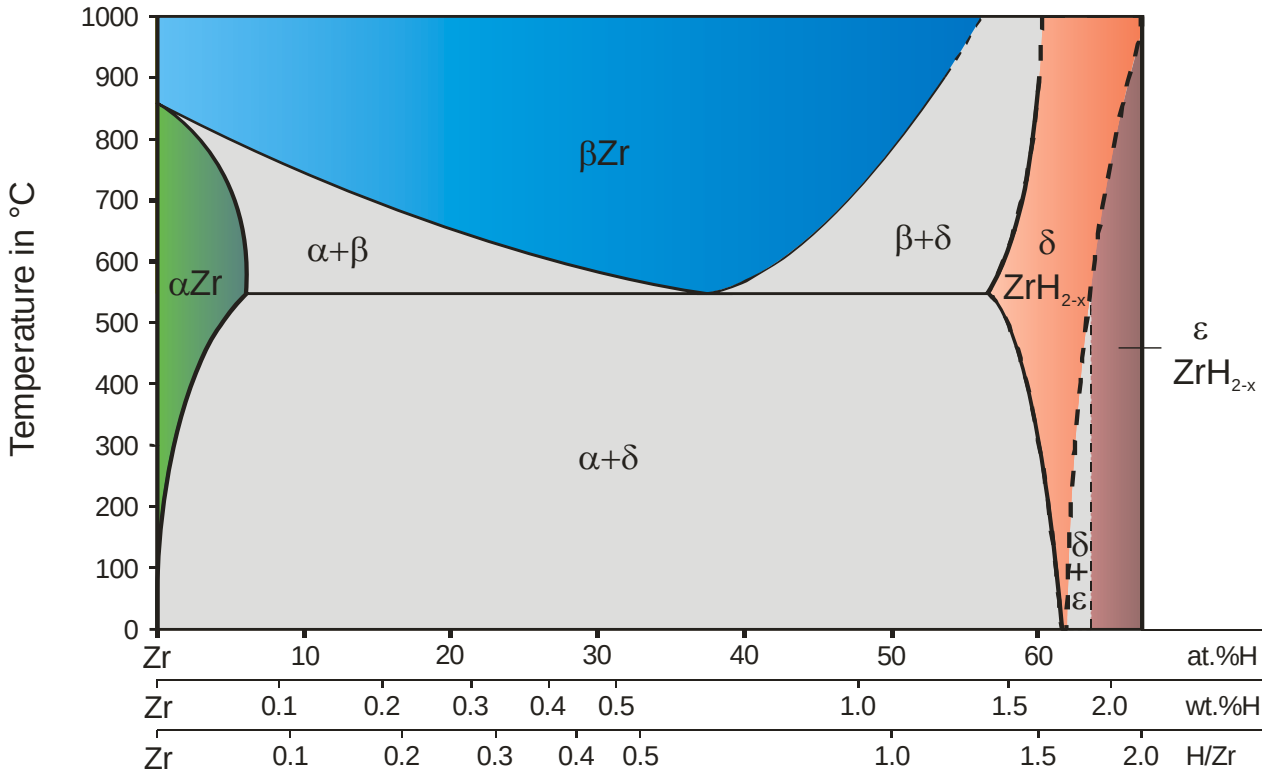
Phase diagram Zr - O



Phase diagram Zr-N



Phase diagram Zr - H



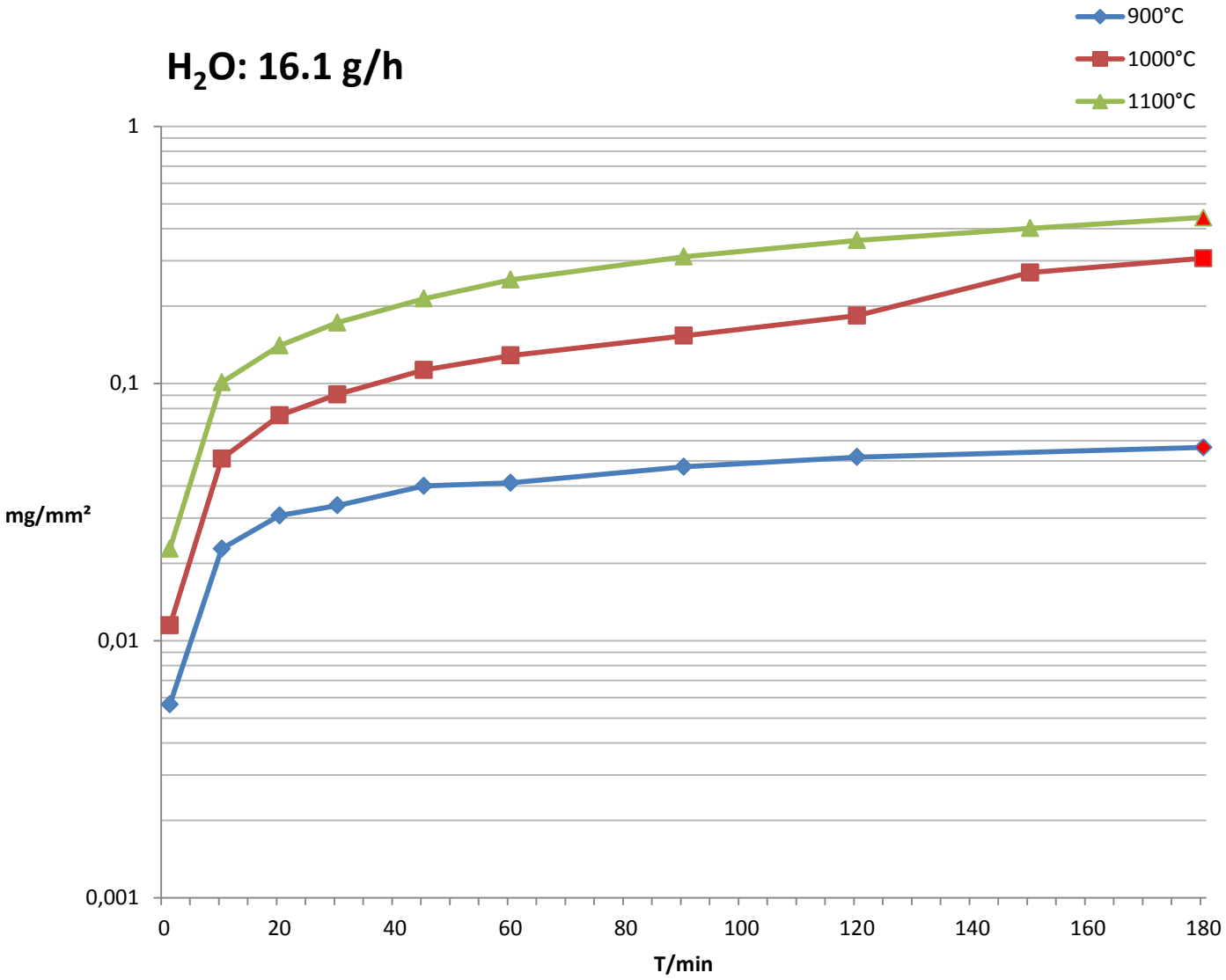
Sieverts' law:

$$\frac{H}{Zr} = k_S \cdot \sqrt{p_{H_2}}$$

with

$$k_S = A \cdot e^{\frac{-B}{RT}}$$

Mass change in different atmospheres





900°C 180min

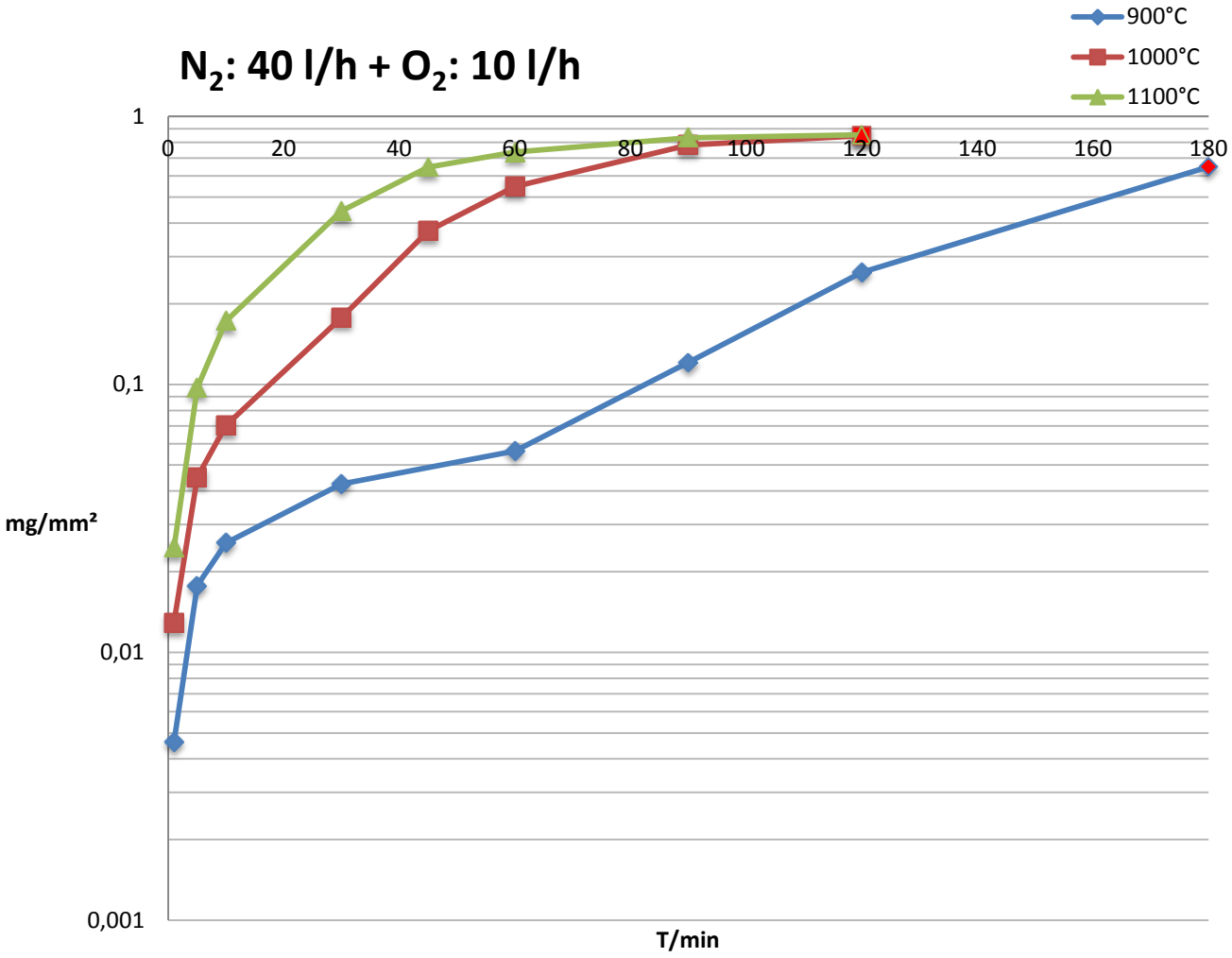


1000°C 180min



1100°C 180min

Mass change in different atmospheres





900°C 180min

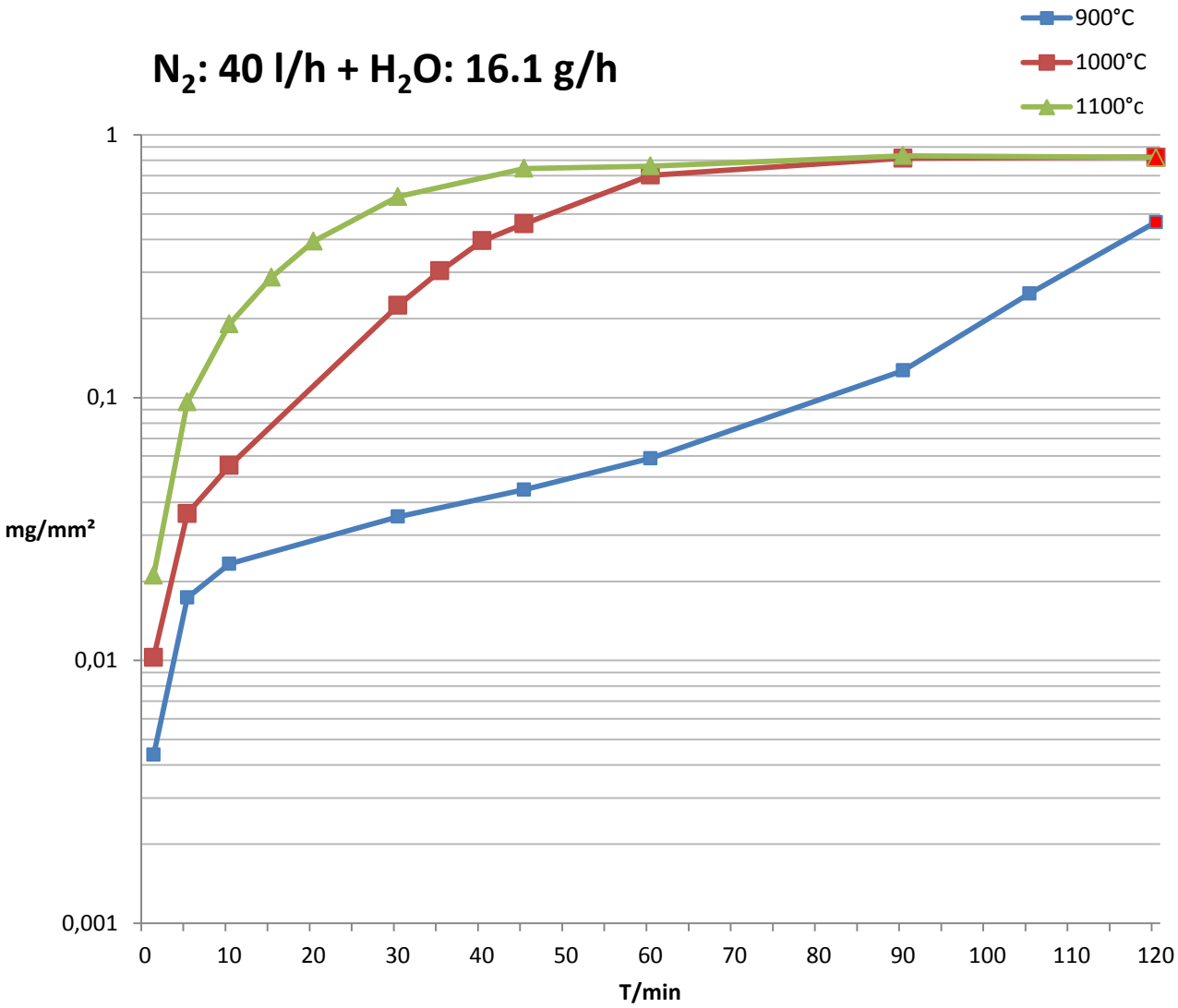


1000°C 120min



1100°C 120min

Mass change in different atmospheres





900°C 120min

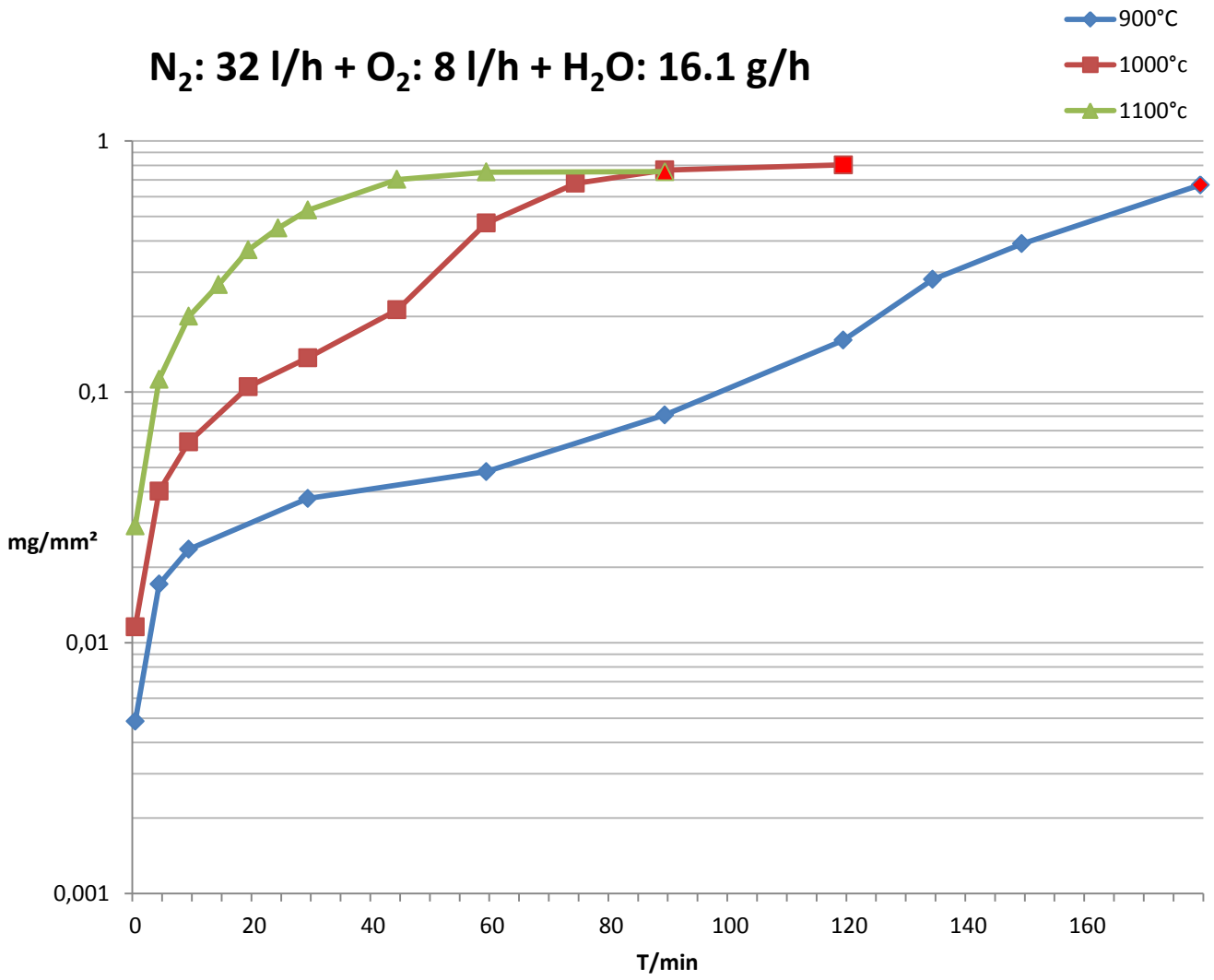


1000°C 120min



1100°C 120min

Mass change in different atmospheres





900°C 180min



1000°C 120min



1100°C 90min

Experimental setup



Experimental setup



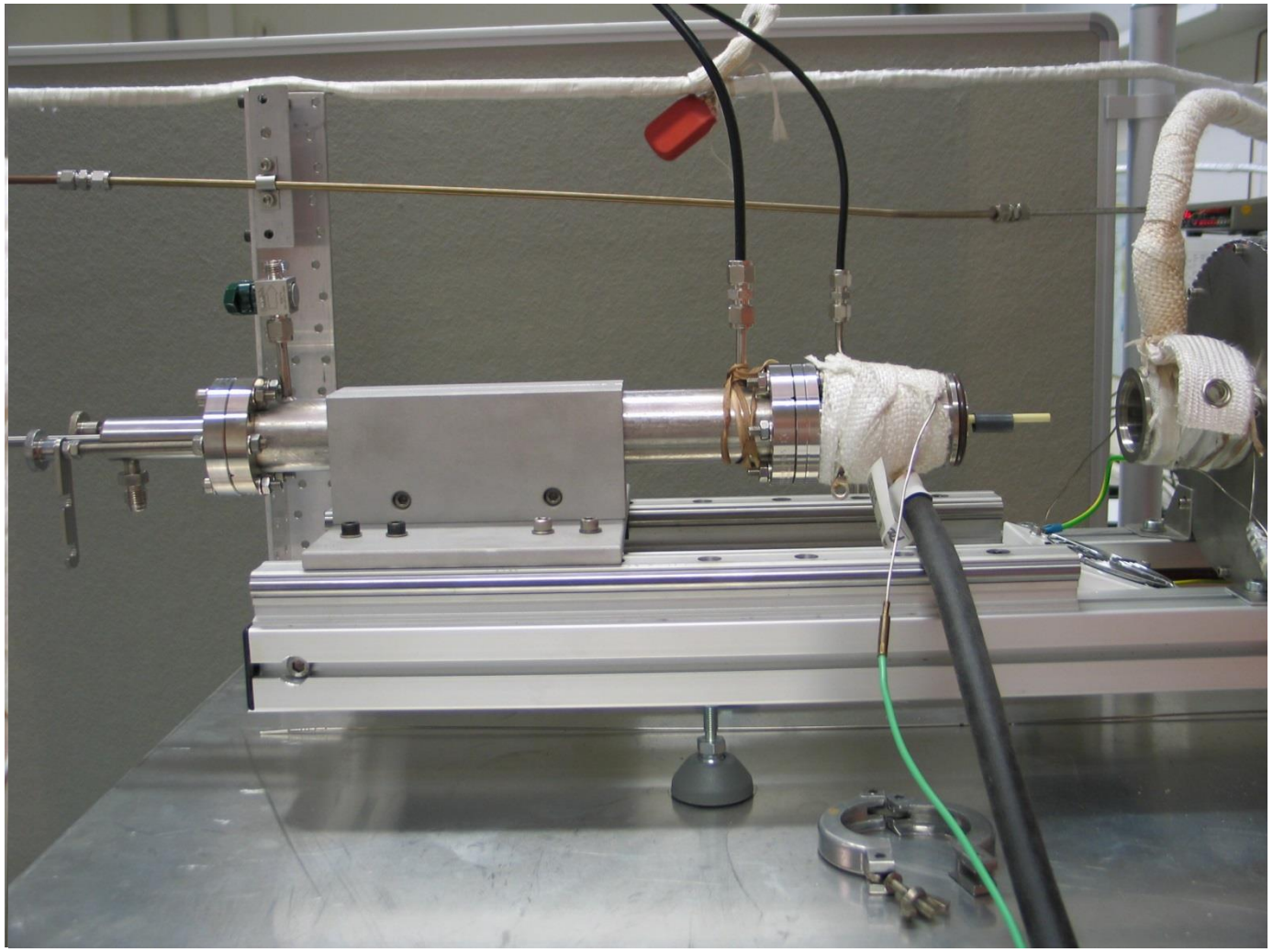
Sample gas inlet

Mass spectrometer

Water tank for steam



Experimental process



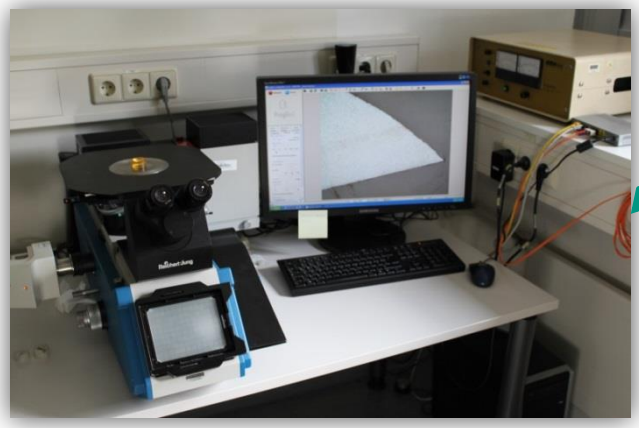
Preparation of the samples



Epoxy imbedding



Grinding/polishing



Light microscopy

A quantification of effect of nitrogen during air ingress scenarios

Sanggil Park

HGU-ACT Research Cooperation Center

act-park@handong.edu

In this study, we investigated the oxidation behaviors of Zry-4 cladding in oxygen and air at high temperature. The main difference in oxygen and air is an inclusion of nitrogen. It is well known that the nitrogen affects the reaction kinetics in two ways. One is the nitriding which is the zirconium nitride (ZrN) formation. By forming the ZrN in the oxide scale, it leads to the incoherent oxide/nitride scale which includes a micro porosity due to a volume mismatch of ZrO_2 and ZrN. The other is the re-oxidation which is a phase transformation from the ZrN to ZrO2 when the ZrN is re-oxidized by an incoming oxygen in air. In the air oxidation, both nitriding and re-oxidation phenomena occur simultaneously and hence it is quite difficult to analyze the effect of nitrogen very precisely and quantitatively.

In addition, the nitrogen becomes active where the oxygen gas partial pressure is sufficiently lower than the nitrogen gas partial pressure. Firstly the oxygen is dissolved in the zirconium metal and forms the oxygen-stabilized alpha zirconium phase. At this point and location, the nitrogen incorporates into the oxygen-stabilized alpha zirconium where the oxygen gas pressure is locally very low. For this reason, it would be critical to know the active condition for the nitrogen reaction in the nitriding and re-oxidation. When it comes to the nitriding, the low oxygen content would be preferable for the ZrN formation. However, for the re-oxidation, a certain degree of oxygen content should be available for the efficient re-oxidation of previously formed ZrN. Therefore, there could be a competition between total contents of oxygen and nitrogen in air. Although the ratio between oxygen and nitrogen is constant as a 2:8, the absolute amount of oxygen and nitrogen content can be different by a various total flow rate.

In order to identify the active condition for the effect of nitrogen, we planned a series of oxidation tests with only oxygen atmosphere and only air atmosphere at 1000ºC by varying the flow rate from 20 to 100 ml/min with an increment of 20 ml/min. By performing this test plan, we observed the global trend of reaction kinetics of air oxidation and oxygen oxidation.



A quantification of effect of nitrogen during air ingress scenarios

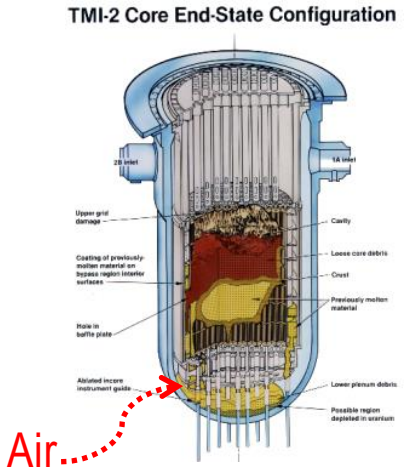
S. Park (HGU/ACT, Previously PSI)
G. Kim (HGU), J. Lee (HGU)

19 October 2017

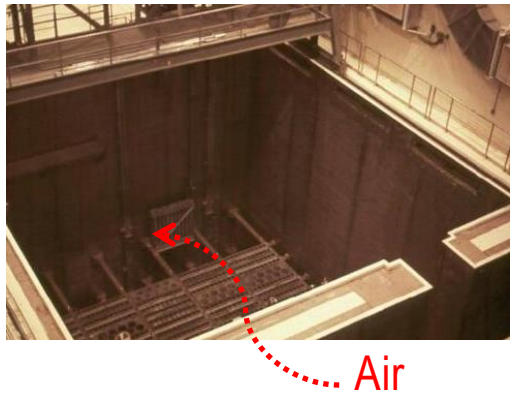
Air ingress scenarios

Air ingress scenarios: Zr oxidation in atmosphere containing N_2

- into the reactor core at the late phase of SAs after RPV failure
- into the reactor core during mid-loop operation to do refueling
- into the spent fuel pool during LOCA
- into the dry storage cask by the mis-handling

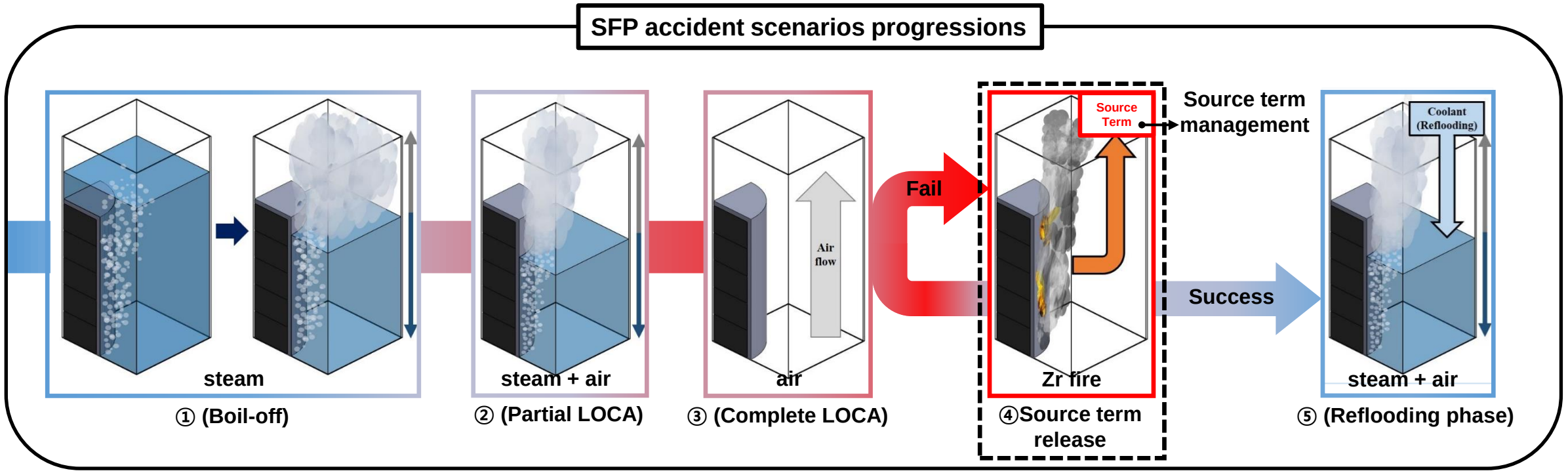


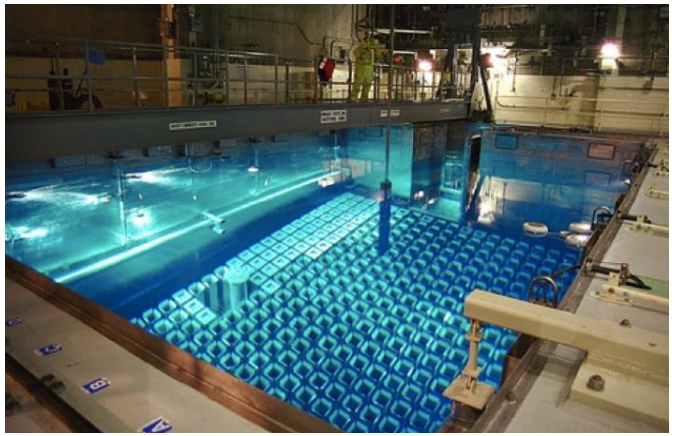






SFP accident scenarios





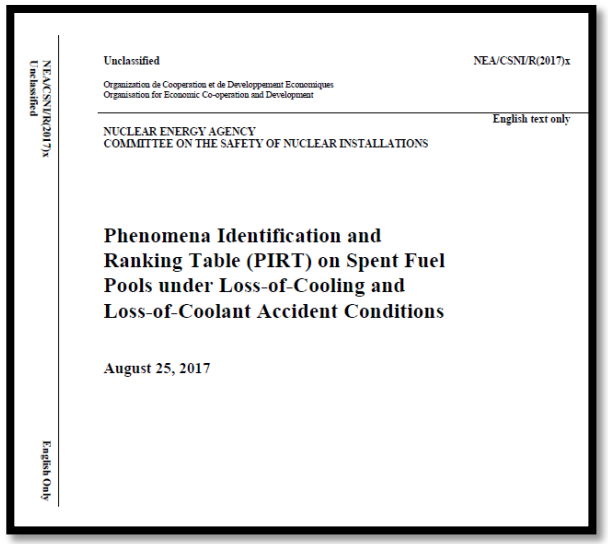
Loss of cooling

Loss of coolant



Complex oxidation mechanisms

Knowledge status on the role of nitrogen



NEA/CSNI/R(2017)x
Unclassified

Unclassified NEA/CSNI/R(2017)x

Organization de Cooperation et de Developpement Economiques
Organisation for Economic Co-operation and Development

English text only

NUCLEAR ENERGY AGENCY
COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

Phenomena Identification and Ranking Table (PIRT) on Spent Fuel Pools under Loss-of-Cooling and Loss-of-Coolant Accident Conditions

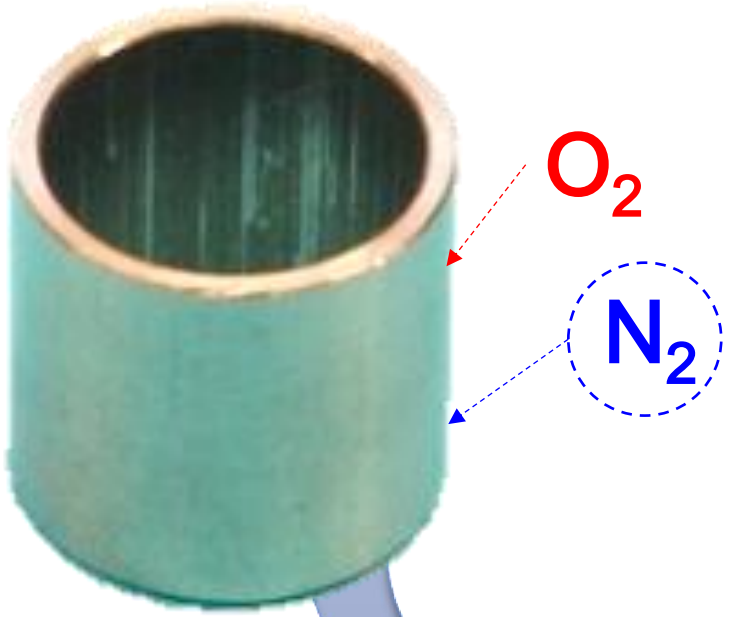
August 25, 2017

English Only

OECD Report on the SFP PIRT

Phenomena by category	#	Sce-nar-io	Importance ranking L	M	H	$I\!L$	State of knowledge: Availability of data N	S	A	KL^D	Availability of models N	S	A	KL^M
Radiative heat transfer from uncovered fuel assemblies to other FAs, racks and SFP structure.	III.12	SU	0	6	13	0.842	7	11	0	0.306	2	15	1	0.472
		FD	0	3	16	0.921	7	11	0	0.306	2	15	1	0.472
Fuel/cladding heat-up.	III.13	SU	1	1	16	0.917	3	5	10	0.694	1	8	9	0.722
		FD	1	1	16	0.917	3	5	10	0.694	1	8	9	0.722
Heat conduction in different components. e.g. storage racks, SFP walls and floor.	III.14	SU	8	10	1	0.316	3	8	7	0.611	2	9	7	0.639
		FD	7	10	2	0.368	3	8	7	0.611	2	9	7	0.639
Outlet fluid sensible heat. Influences the buoyancy of the bulk steam/gas flow in the SFP, and in turn, the transport of released fission products to the environment.	III.15	SU	8	9	2	0.342	5	9	4	0.472	1	12	5	0.611
		FD	8	8	3	0.368	5	9	4	0.472	2	11	5	0.583
4. Fuel behaviour														
Effect of sea water injection on the fuel degradation and coolability.	III.16	SU	8	9	0	0.265	9	5	1	0.233	9	6	0	0.200
		FD	8	9	0	0.265	9	5	1	0.233	9	6	0	0.200
Water radiolysis and hydrogen production.	III.17	SU	13	4	1	0.167	5	7	6	0.528	5	7	6	0.528
		FD	16	1	1	0.083	4	8	6	0.556	5	7	6	0.528
Fuel cladding embrittlement due to hydriding and oxidation.	III.18	SU	4	4	11	0.684	1	12	6	0.632	2	12	5	0.579
		FD	5	4	10	0.632	1	12	6	0.632	2	12	5	0.579
Zr oxidation in steam. Includes heat/hydrogen generation and kinetics (oxide breakaway).	III.19	SU	1	1	17	0.921	2	2	15	0.842	0	6	13	0.842
		FD	1	3	15	0.868	2	2	15	0.842	0	6	13	0.842
Zr oxidation in air by oxygen. Includes heat generation and kinetics (oxide breakaway).	III.20	SU	1	3	15	0.868	4	7	8	0.605	2	9	7	0.639
		FD	0	1	18	0.974	3	8	8	0.632	1	10	7	0.667
Nitride formation in air.	III.21	SU	0	6	13	0.842	4	12	3	0.474	5	12	2	0.421
		FD	0	4	15	0.895	3	13	3	0.500	4	13	2	0.447
Re-oxidation of ZrN by steam/oxygen.	III.22	SU	0	9	9	0.750	6	11	1	0.361	11	7	0	0.194
		FD	0	8	9	0.765	5	12	1	0.389	10	8	0	0.222
Zr oxidation in steam/air mixtures. Includes oxygen starvation during oxidation in steam/air and oxide breakaway.	III.23	SU	0	1	18	0.974	4	9	6	0.553	2	12	5	0.579
		FD	0	1	18	0.974	4	9	6	0.553	2	12	5	0.579
Cladding ignition.	III.24	SU	1	3	14	0.861	3	9	6	0.583	2	12	4	0.556
		FD	0	0	18	1.000	2	10	6	0.611	1	13	4	0.583
Oxidation of debris.	III.25	SU	4	5	8	0.618	4	11	2	0.441	2	14	1	0.471
		FD	4	5	8	0.618	4	11	2	0.441	2	14	1	0.471

Question ?





Effect of N_2



Answer !



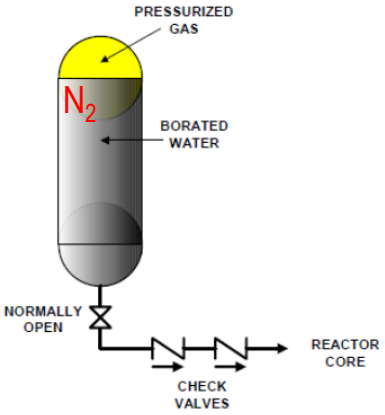
- Qualitative Answer
- Quantitative Answer

Qualitative Answer on the Effect of N_2 [1/4]

N_2 filling gas for the safety purpose

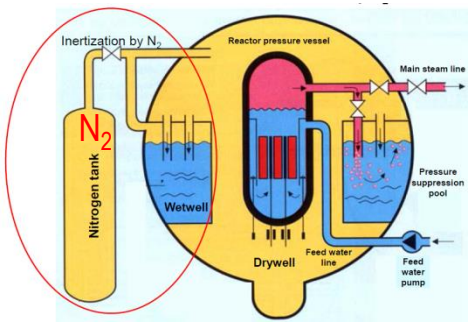


► N_2 gas has been used as a filling gas due to its stability



N_2 filling gas for the safety purpose in NPP

- PWR: Hydroaccumulator (passive HP injection)
- BWR: N_2 inerted containment (to prevent hydrogen combustion)



Qualitative Answer on the Effect of N_2 [2/4]

Is nitrogen a stable gas?



Oxygen (O_2)

- Dissociation of double bond = +497 kJ/mol
- Gibbs energy of formation ΔG_{ZrO_2} = -896 kJ/mol (1073.15K)



Nitrogen (N_2)

- Dissociation of triple bond = +945 kJ/mol
- Gibbs energy of formation ΔG_{ZrN} = -264 kJ/mol (1073.15K)

O_2 reacts first

↓

After consuming O_2

↓

N_2 reacts actively!

- Steam oxidation: $2H_2O+Zr \rightarrow ZrO_2+2H_2$ $\Delta H_{rxn}=-616$ kJ/mol Zr
- Oxygen oxidation: $O_2+Zr \rightarrow ZrO_2$ $\Delta H_{rxn}=-1094.8$ kJ/mol Zr

ca. 1.8 times heat release

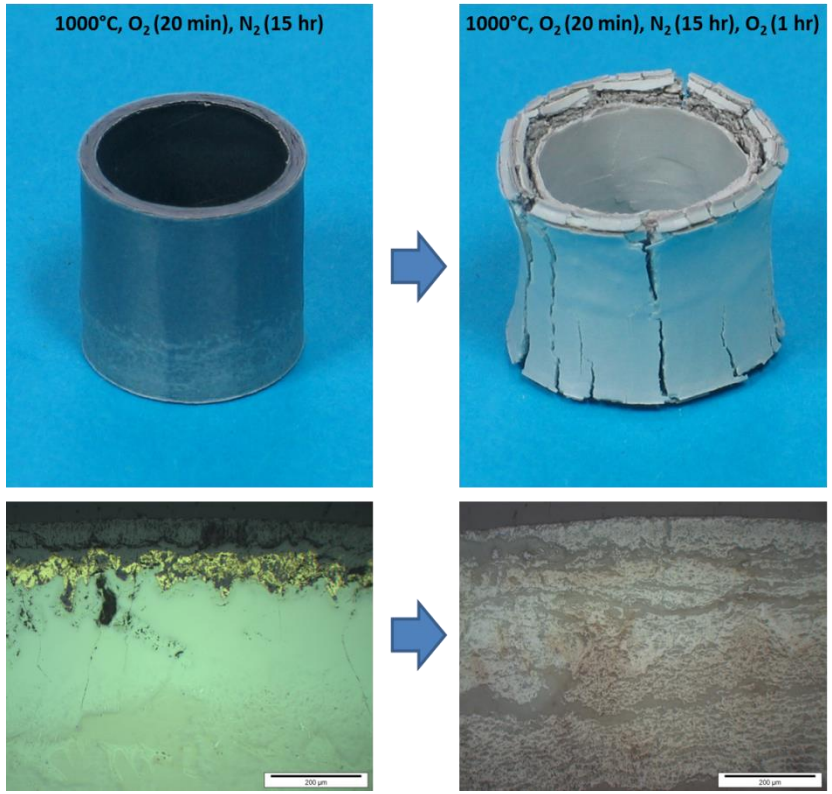
- Nitriding: $1/2N_2+Zr \rightarrow ZrN$ $\Delta H_{rxn}=-365.4$ kJ/mol Zr
- Re-oxidation: $ZrN+O_2 \rightarrow ZrO_2+1/2N_2$ $\Delta H_{rxn}=-729.4$ kJ/mol Zr

Cladding degradation by nitriding and re-oxidation

Qualitative Answer on the Effect of N_2 [3/4]

Cladding degradation under influence of N_2

Nitriding | **Re-oxidation**



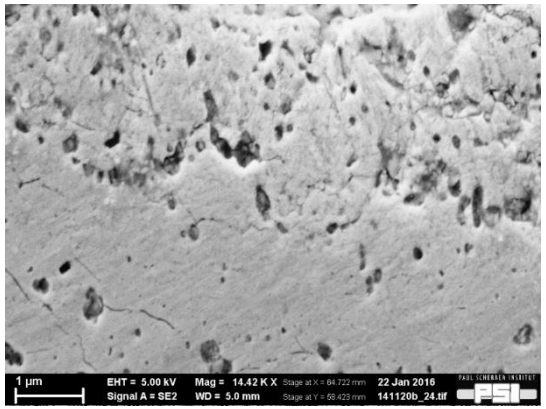
- **Nitriding** of pre-oxidized Zr-based cladding leads to micro porous and less coherent oxide scale.

$$\overline{V}_{ZrN}/\overline{V}_{ZrO_2} = 0.68$$

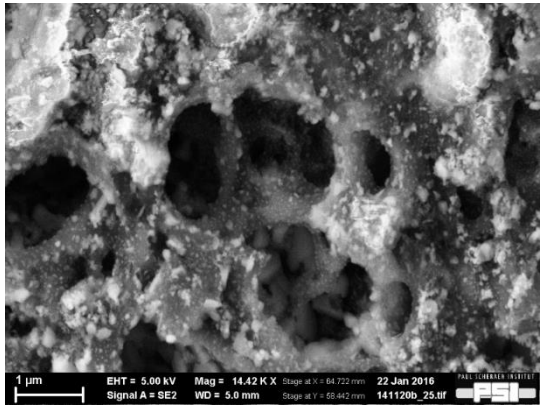
- **Re-oxidation** of nitrided pre-oxidized Zr-based cladding leads to macro cracked oxide scale.

$$\overline{V}_{ZrO_2}/\overline{V}_{ZrN} = 1.47$$

Oxide 14420x



Nitride 14420x



Qualitative Answer on the Effect of N_2 [4/4]



- Is it stable like inert gas? → No.
- Cladding degradation by Nitriding & Re-oxidation
- Nitriding when?
  1. O_2 is sufficiently consumed.
  2. O_2 is dissolved in the Zr-metal.
  3. Oxygen-stabilized α-Zr(O) is formed.
- Re-oxidation when?
  1. ZrN is exposed by the incoming O_2.

Quantitative Answer on the Effect of N_2 [1/6]



- TGA: Setaram Setsys
- Cladding: Zry-4
- Test matrix (1 hour test at 1000°C)

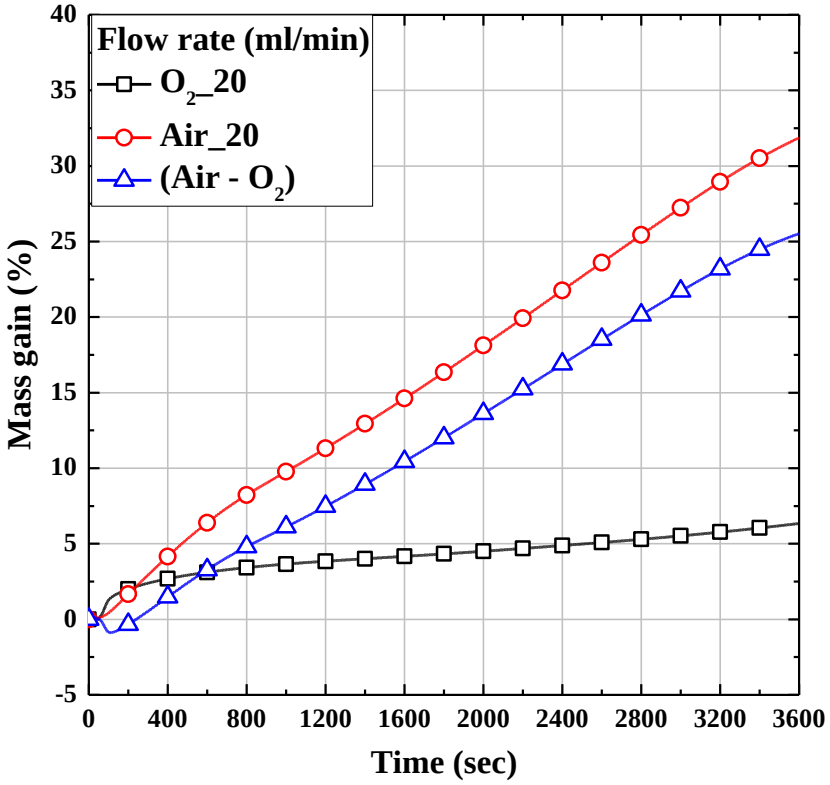
Flow rate (ml/min)	20	40	60	80	100
O_2/Ar	o	o	o	o	o
Air/Ar	o	o	o	o	o

- Every test was performed twice to make sure the reproducibility.

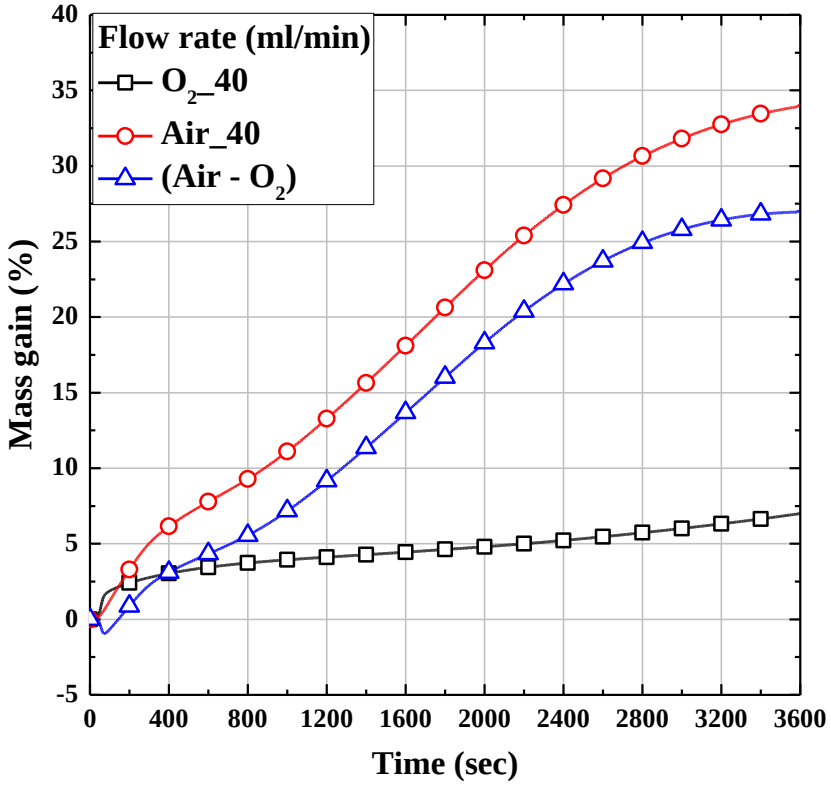
Quantitative Answer on the Effect of N_2 [2/6]

Results for 20 and 40 ml/min tests

20 ml/min



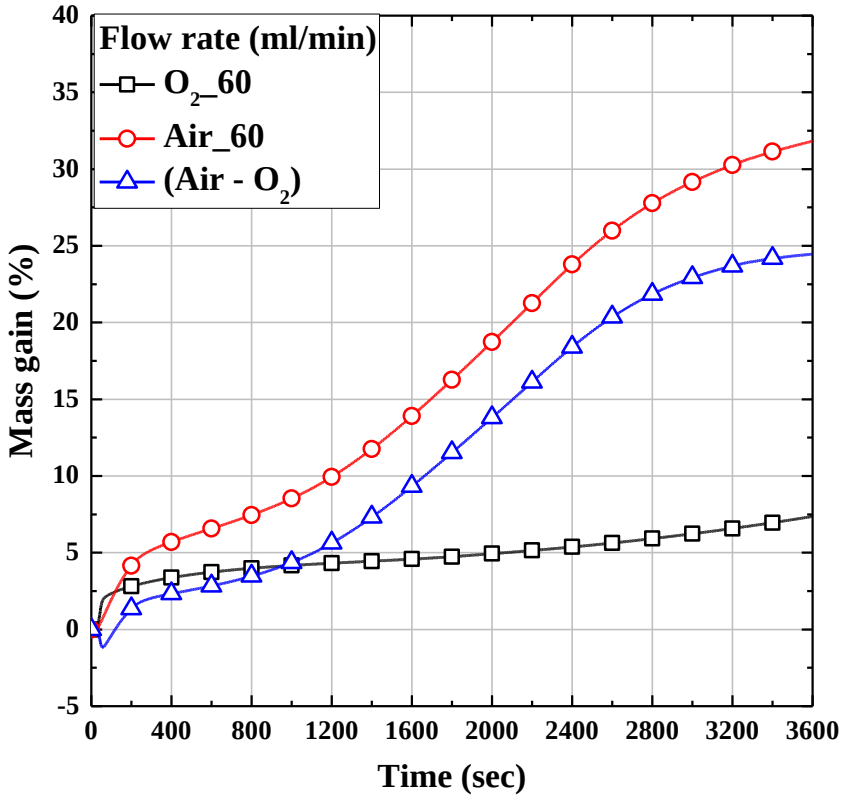
40 ml/min



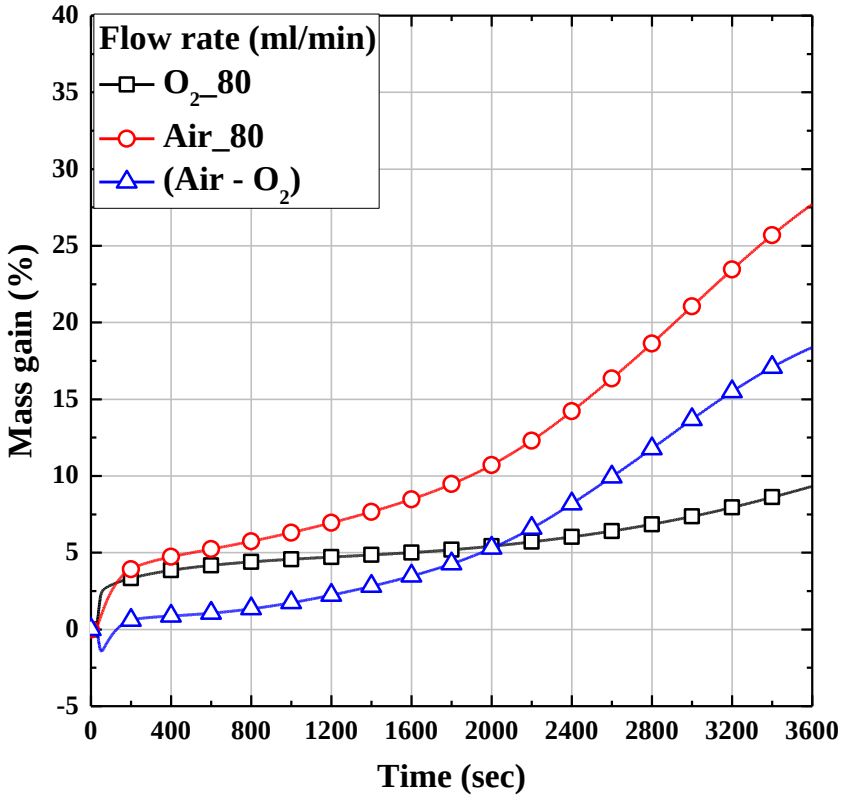
Quantitative Answer on the Effect of N_2 [3/6]

Results for 60 and 80 ml/min tests

60 ml/min



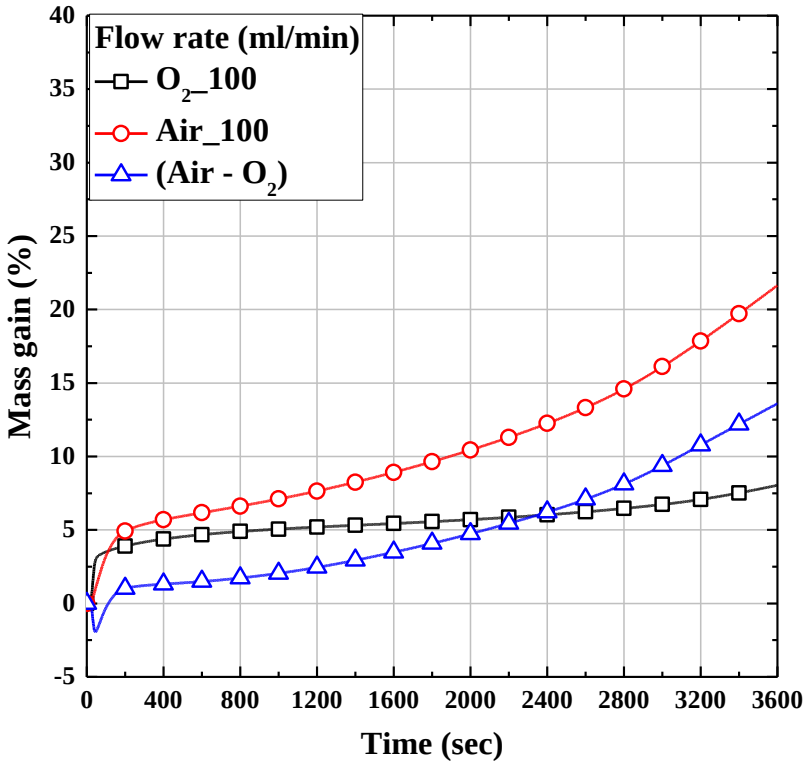
80 ml/min



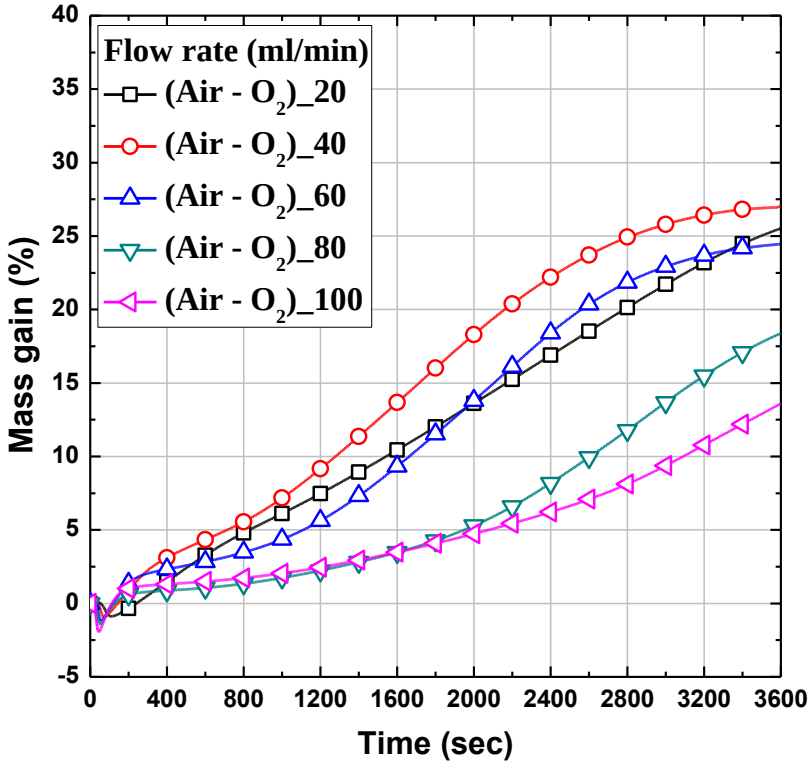
Quantitative Answer on the Effect of N_2 [4/6]

Results for 100 ml/min test and Summary

100 ml/min

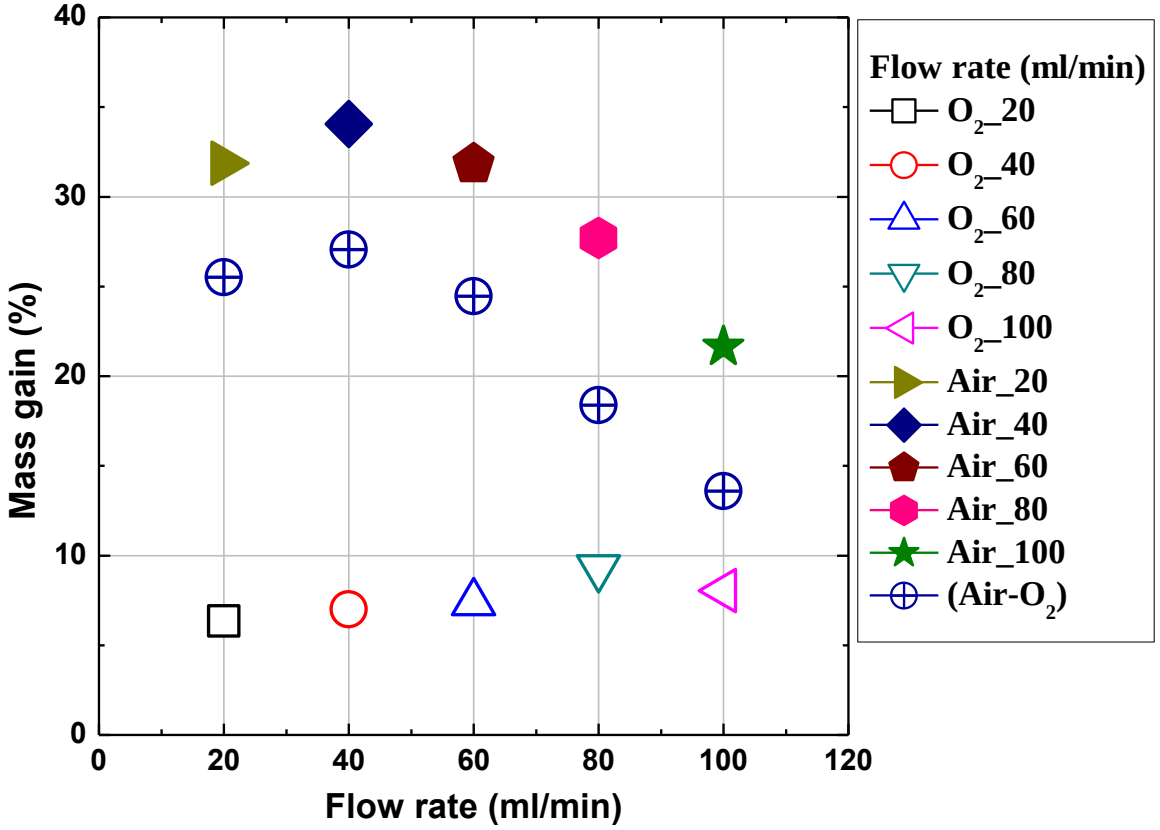


Total



Quantitative Answer on the Effect of N_2 [5/6]

Discussion

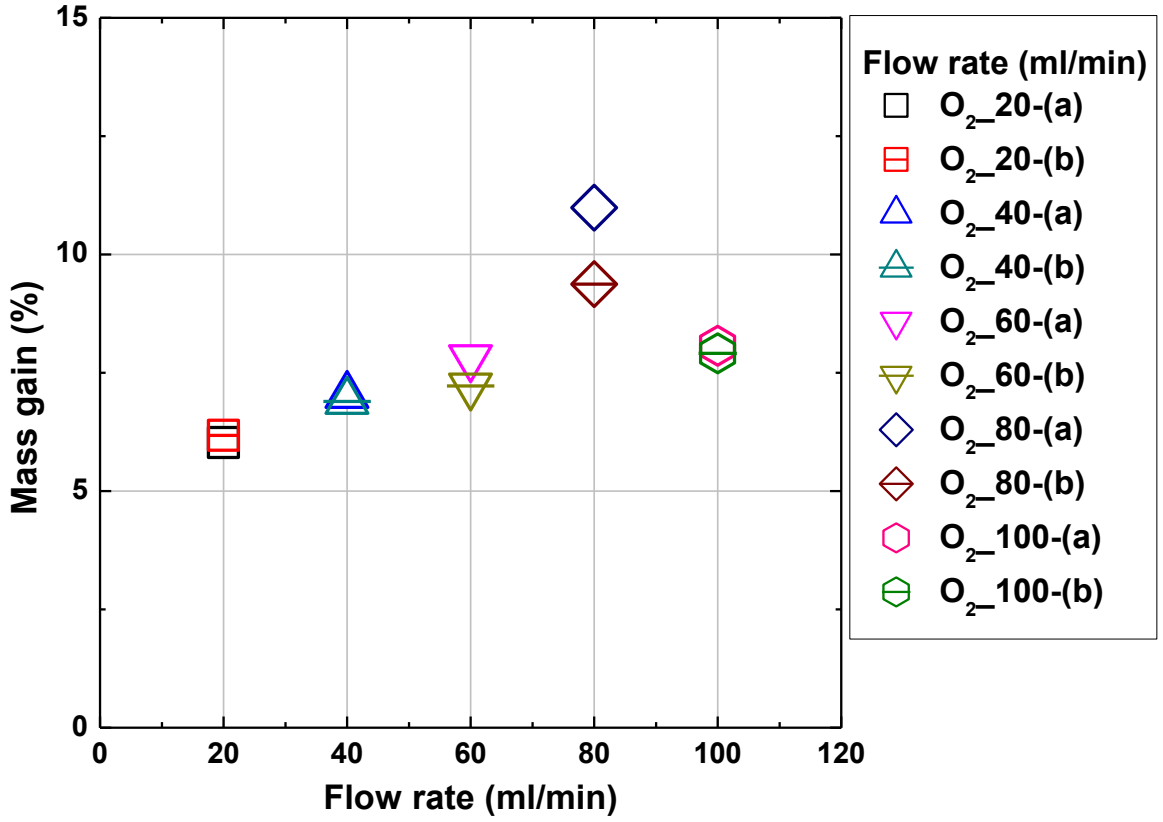


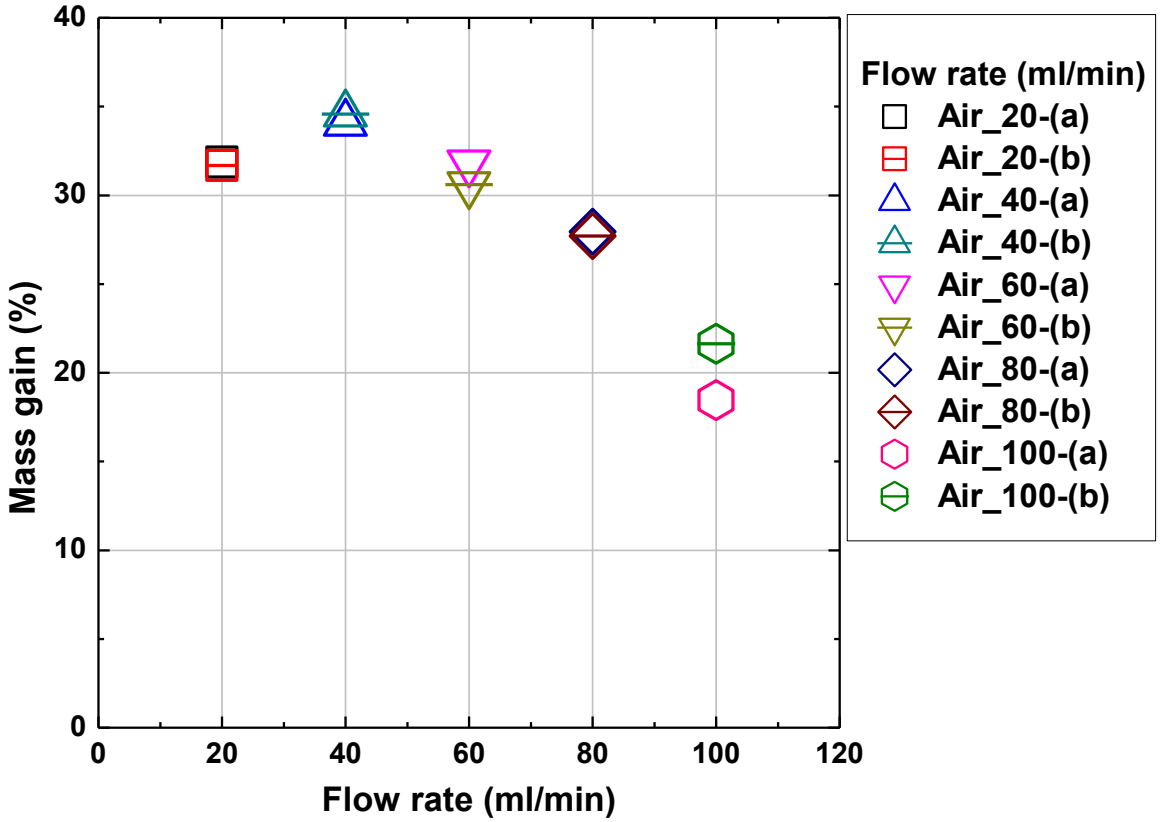
Flow rate (ml/min)	20	40	60	80	100
Δm (Air)	31.86	34.05	31.82	27.70	21.64
Δm (O_2)	6.34	7.00	7.37	9.32	8.03
Difference	25.52	27.05	24.45	18.38	13.61
Ratio Δm(Air/O_2)	5.03	4.86	4.31	2.97	2.69
Ratio Diff/Δm(O_2)	4.03	3.86	3.31	1.97	1.69

High chance of O_2 starvation

Quantitative Answer on the Effect of N_2 [6/6]

Reproducibility Check





Summary

Effect of N_2 is significantly high!

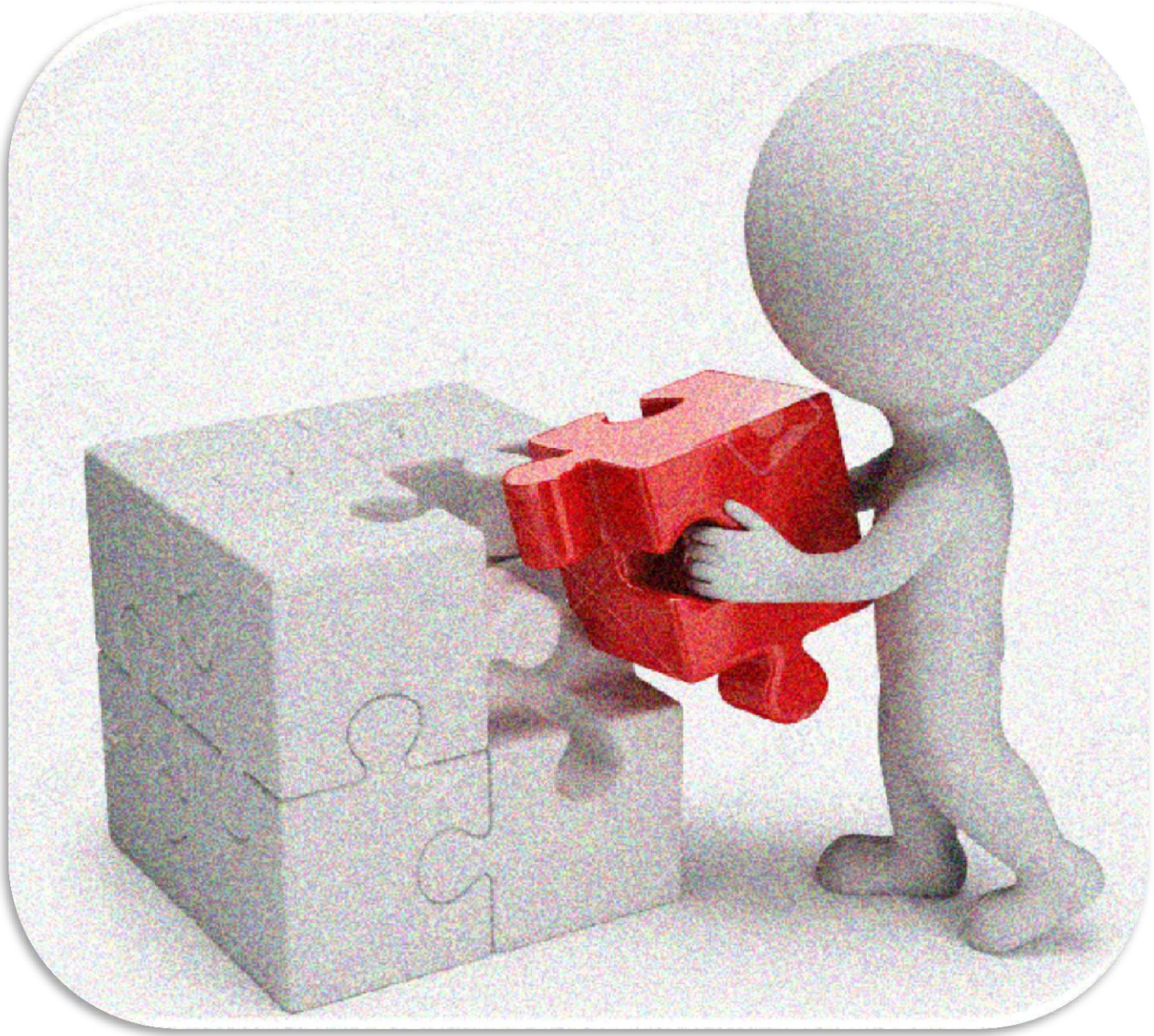
- Effect of N_2: nitriding and re-oxidation.
- Effect of O_2: oxidation.
- Effect of N_2 > Effect of O_2 (1.5 to 4 times high)

Effect of N_2 is dependent on the flow rate

- Nitriding is promoted in a low flow rate.
- Re-oxidation is promoted in a high flow rate.

Optimal flow rate for the effect of N_2

QnA



Close the missing puzzle blocks

To enhance the knowledge!

Status Report on Nitriding Model

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[b] ACT Co. Ltd., Techno 9 Ro, Yuseong-gu, Daejeon, 34027, Korea
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Abstract

The strongly exothermic reaction of nuclear cladding materials with overheated steam is well known. Since several years it has been recognized, that the influence of air and especially nitrogen is responsible for an increase of the oxidation rates of zirconium based cladding materials. Experiments with air have shown increased oxidation rates of the cladding due to breakaway of amorphous material and hence reduction of the protectivity of the oxide layer. Under steam or oxygen starvation it was recognized, that the cladding reacts with nitrogen to produce an amorphous zirconium nitride. This ZrN may reoxidize violently after steam or oxygen recovery as observed at the QUENCH 16 experiment at KIT due to the porous structure of ZrN and therefore enhanced diffusion rates. Those conditions easily may be realized for spent fuel pool accidents or for nuclear accidents under midloop operation.

A series of separate effect tests has been performed at KIT at different temperatures in the frame of a PhD to study the effect of nitriding of pre-oxidized cladding. More than 30 experiments have been performed under breakaway conditions at temperatures of 900°C and 1000°C and more than 40 under non breakaway conditions at temperatures between 900°C and 1200°C. The analysis of these experiments and the first steps of the development of a computer model for the implementation in severe accident codes will be presented.



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PSI



Bernd Jäckel, Sanggil Park, Leticia Fernandez-Moguel :: :: Paul Scherrer Institut

Status Report of Nitriding Model

Quench Workshop, 2017, Karlsruhe, Germany

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2. Separate Effect Tests

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Introduction

Historical overview

Cladding degradation in the past was mostly oxidation by overheated steam under reactor accident conditions.

Later on oxidation in air environment was investigated.

The air oxidation was then modeled (nitrogen as catalyst).

The next step is the nitriding under oxygen starvation.

Programs running to understand re-oxidation of ZrN.

Introduction



A fresh sample compared with a pre-oxidized and nitrided sample and finally a sample after re-oxidation with oxygen.

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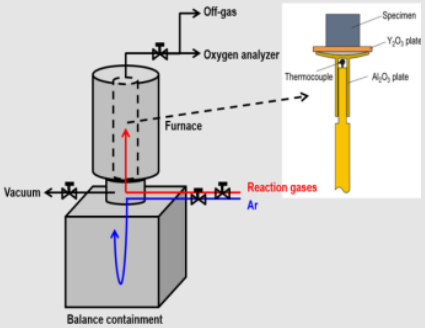
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Experimental Facilities

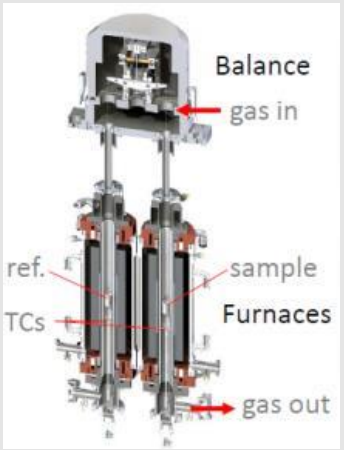
The experiments were conducted in two different facilities

- ❑ Netzsch STA 409, Gas flow 3 l/h Ar, 10 l/h N_2 or O_2
- ❑ Setaram TAG, Gas flow 0.9 l/h Ar, 0.9 l/h N_2 or O_2









Experimental Facilities

- **Mass gain measurements were done in both facilities**
- **Sample size is almost identical in both facilities**
- **The tube furnace in both facilities is slightly different**
- **The Setaram TAG facility has an additional reference tube**
- **A higher leakage rate of the Netzsch STA 409 facility can cause problems at very low reaction rates with N_2**

Test Matrix

Temperature	Pre-oxidation	Nitriding	Re-oxidation
900°C	2h, 6h	1h, 15h	2h
1000°C	20min, 40min, 60min	30min, 1h, 3h, 6h, 15h	1h
1100°C	30s, 10min	30min, 1h, 3h, 6h, 15h	20min
1200°C	10s, 60s	1h, 15h	10min

Not all combinations of the above shown matrix for pre-oxidation, nitriding and re-oxidation were used, but more than 70 tests could be conducted and analyzed. The tests at 900°C and 1200°C were only conducted with the Netzsch STA 409 facility, while the tests at 1000°C and 1100°C were conducted with both facilities. Every second test was without re-oxidation.

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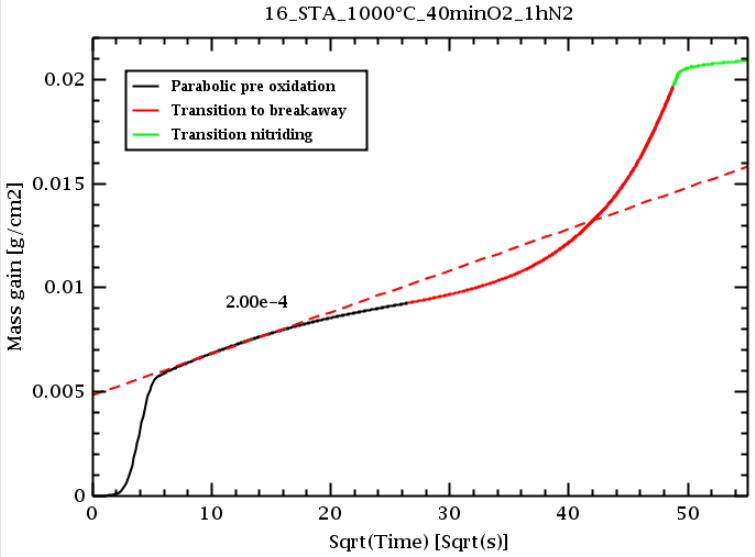
1. Introduction

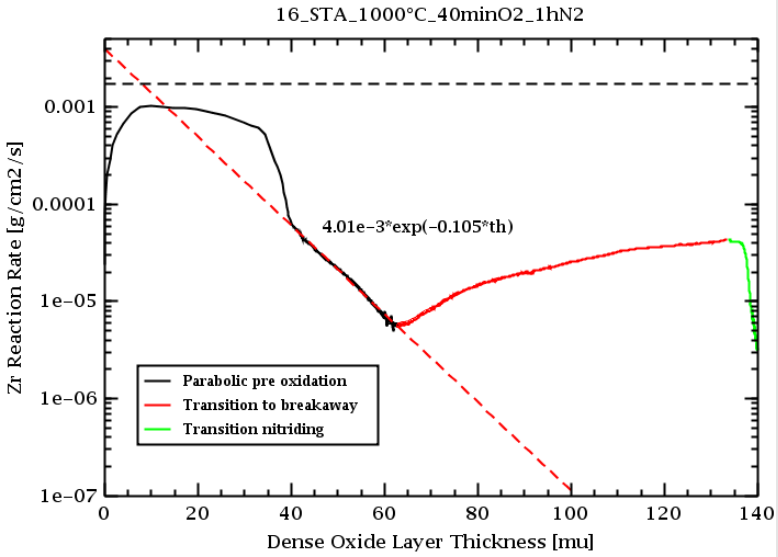
2. Separate Effect Tests

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Mass Gain Measurements and Analysis

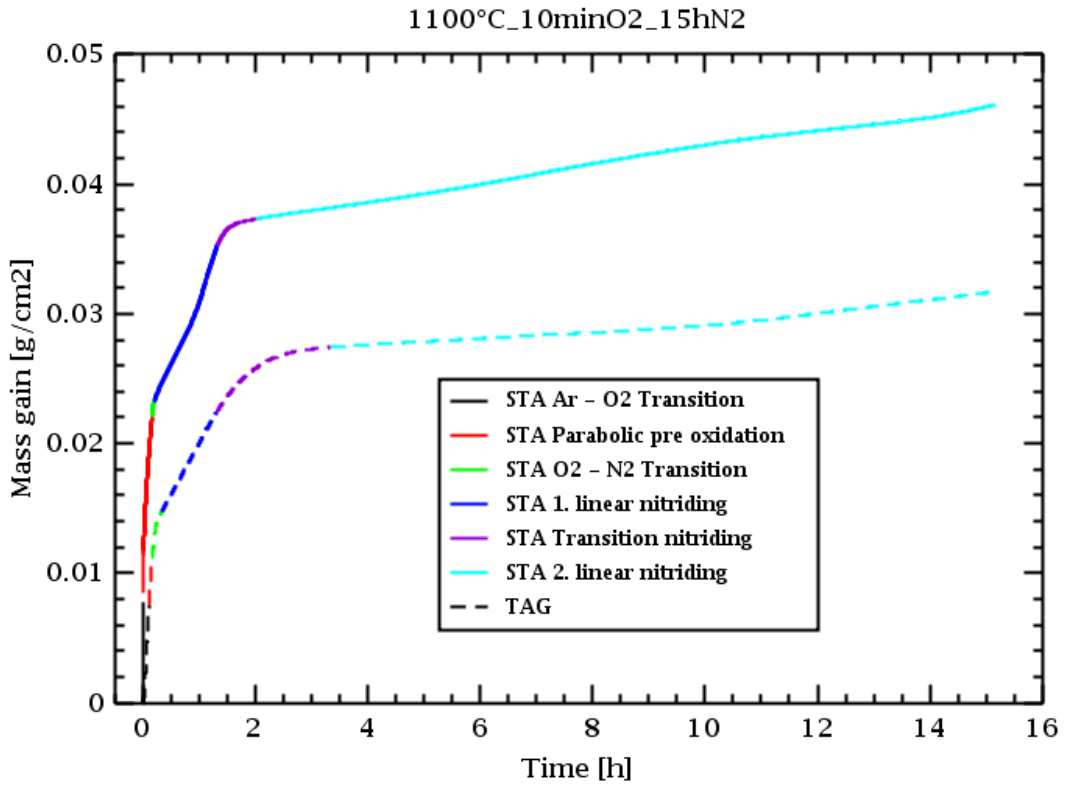




In the period of pre-oxidation two methods of analysis were used:

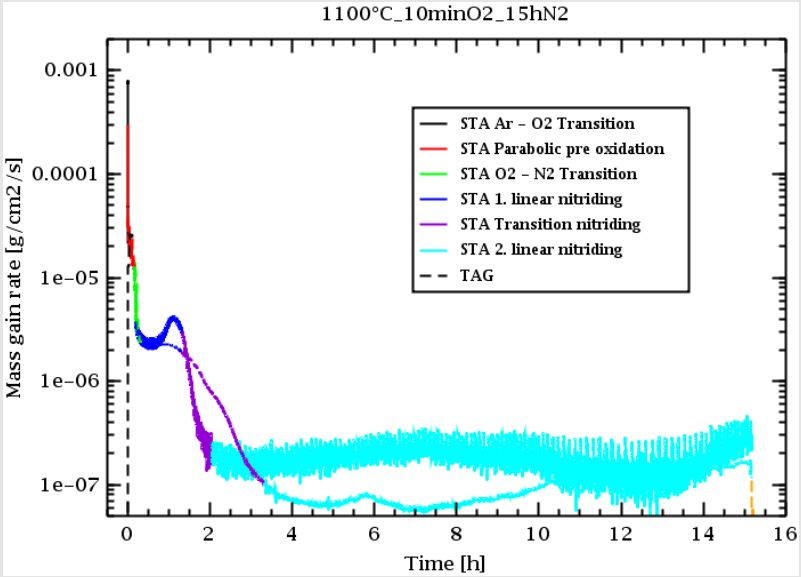
- ❑ Mass gain versus square root of time (parabolic protective oxidation model)
- ❑ Reaction rate versus oxide layer thickness (diffusion based model)

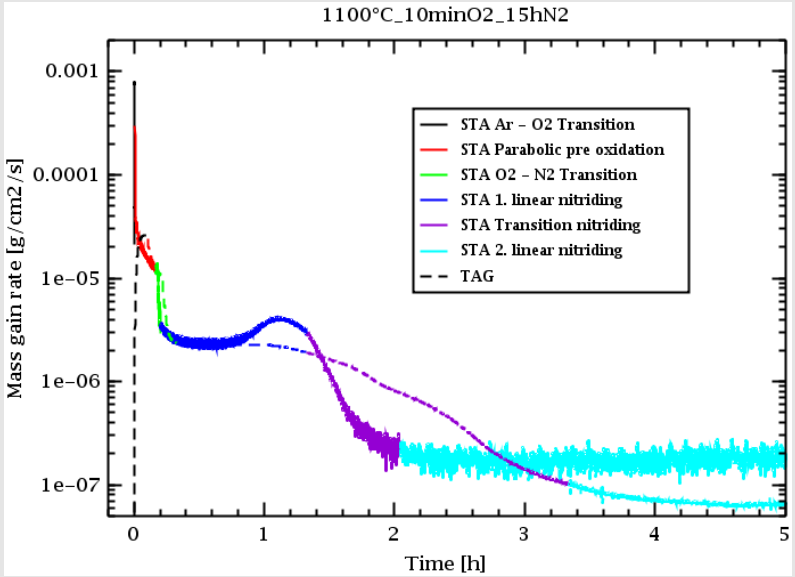
Influence of Facilities on Results



Two facilities are used for the same test setup and the results are compared regarding the differences of the facilities.

Influence of Facilities on Results

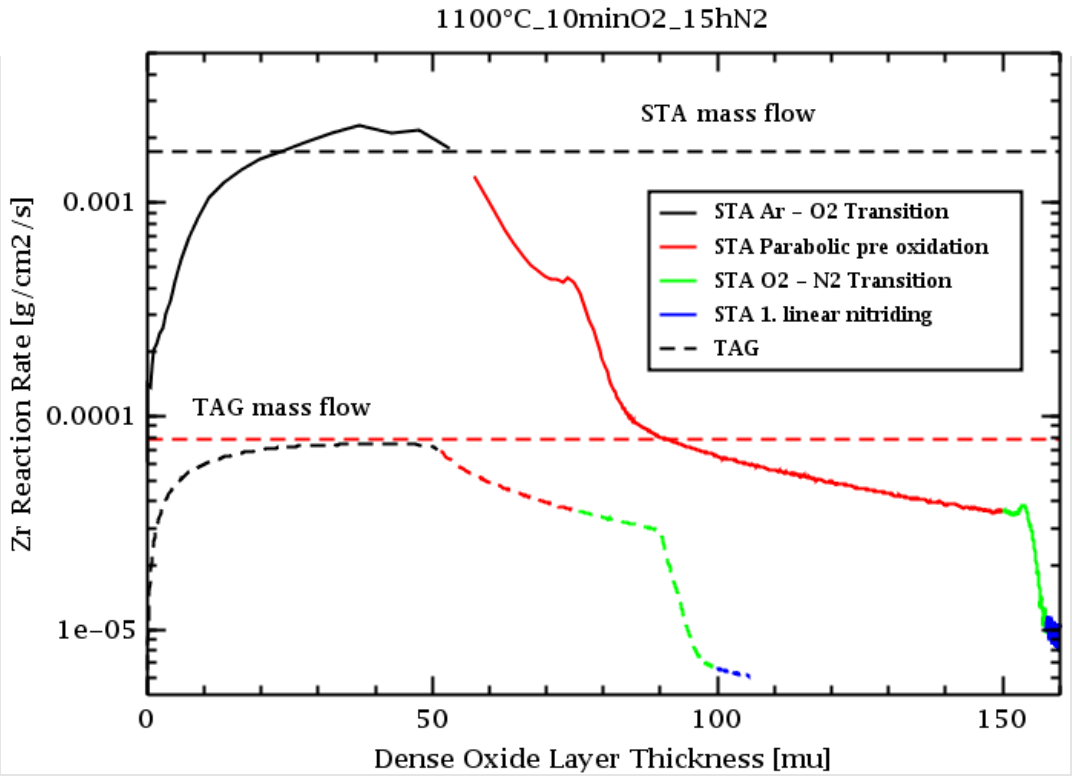




The nitriding process can clearly be divided into two areas.

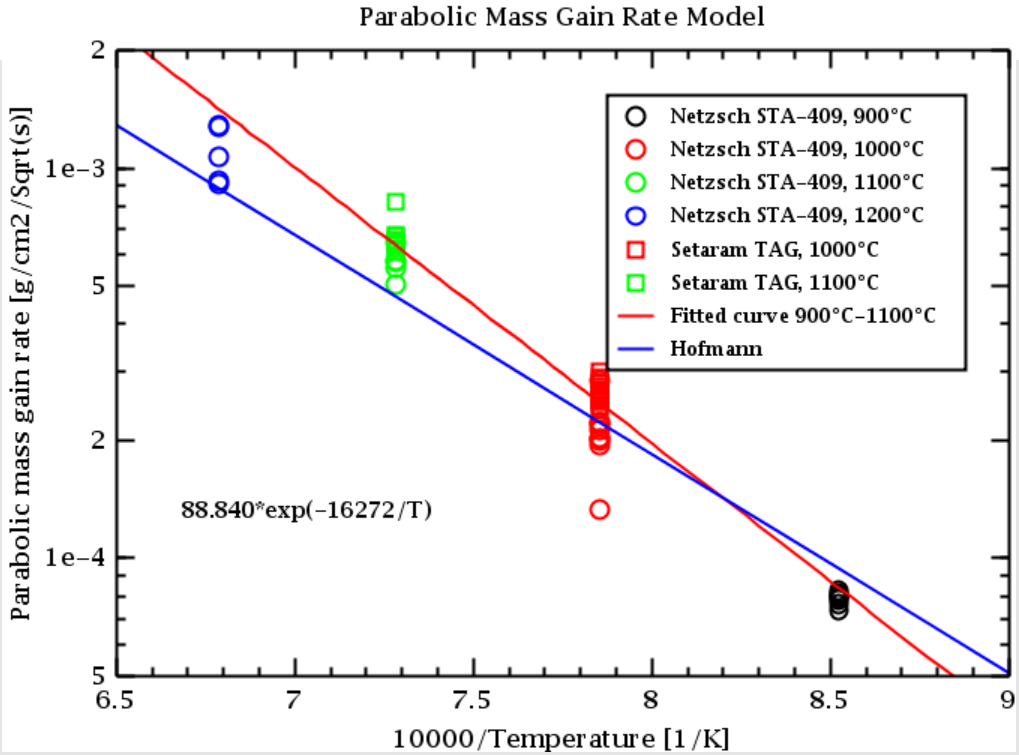
- Fast nitriding mechanism with 2-3*10^{-6} g/cm^2/s
- Slow nitriding mechanism with 1-2*10^{-7} g/cm^2/s

Influence of Facilities on Results



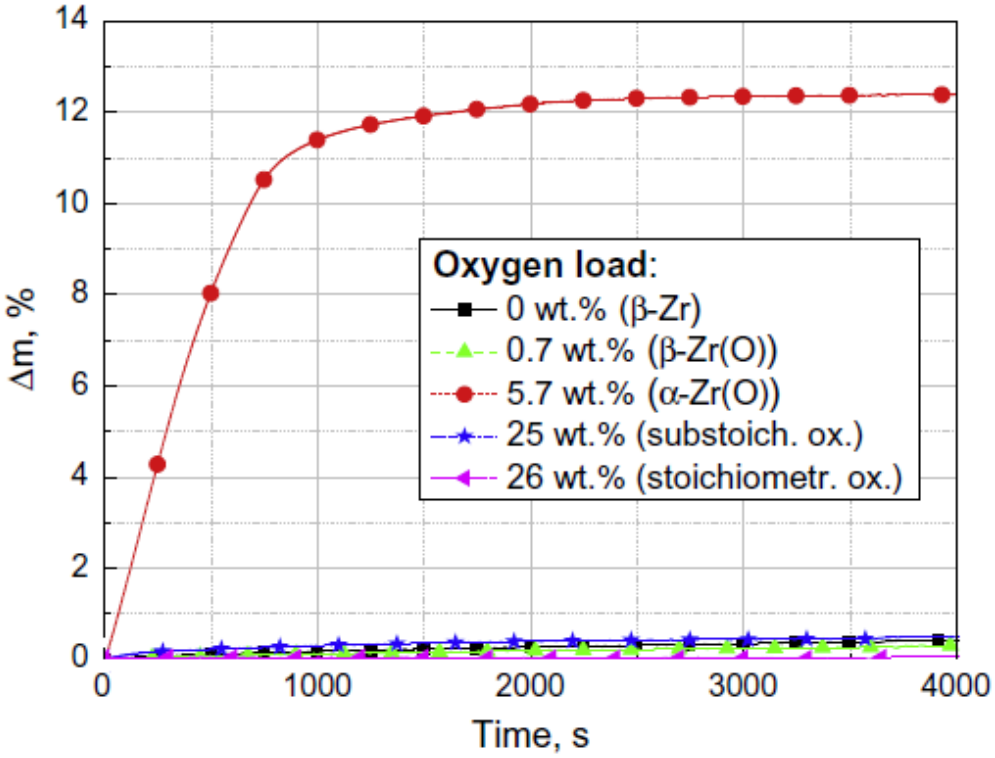
The pre-oxidation at this test setup is clearly influenced by starvation in both facilities. The slope in the diffusion limited oxidation is almost identical, but the multiplication value is different.

Nitriding model (I)



The pre-oxidation of the samples is following closely the oxidation correlation from Hofmann (blue line). The fitted red line is not using the data points of the 1200°C tests, because the oxidation period was to short to establish a constant oxygen concentration for getting reliable data.

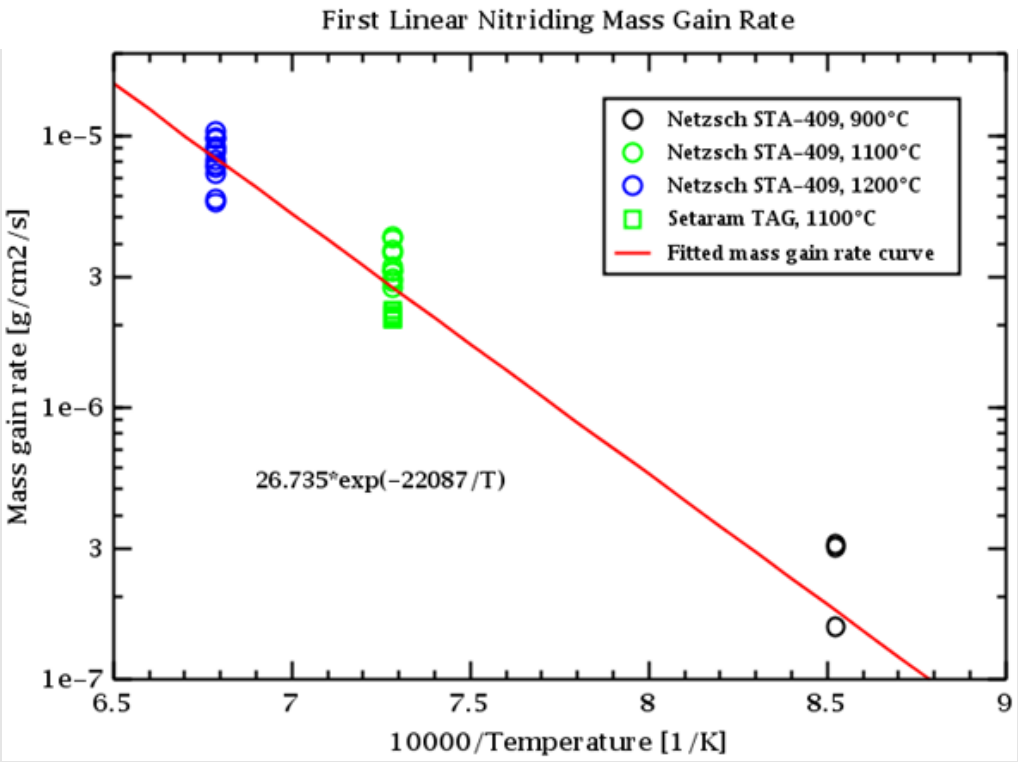
Nitriding model (II)



The fast nitriding phase is interpreted as nitriding of α-Zr(O).

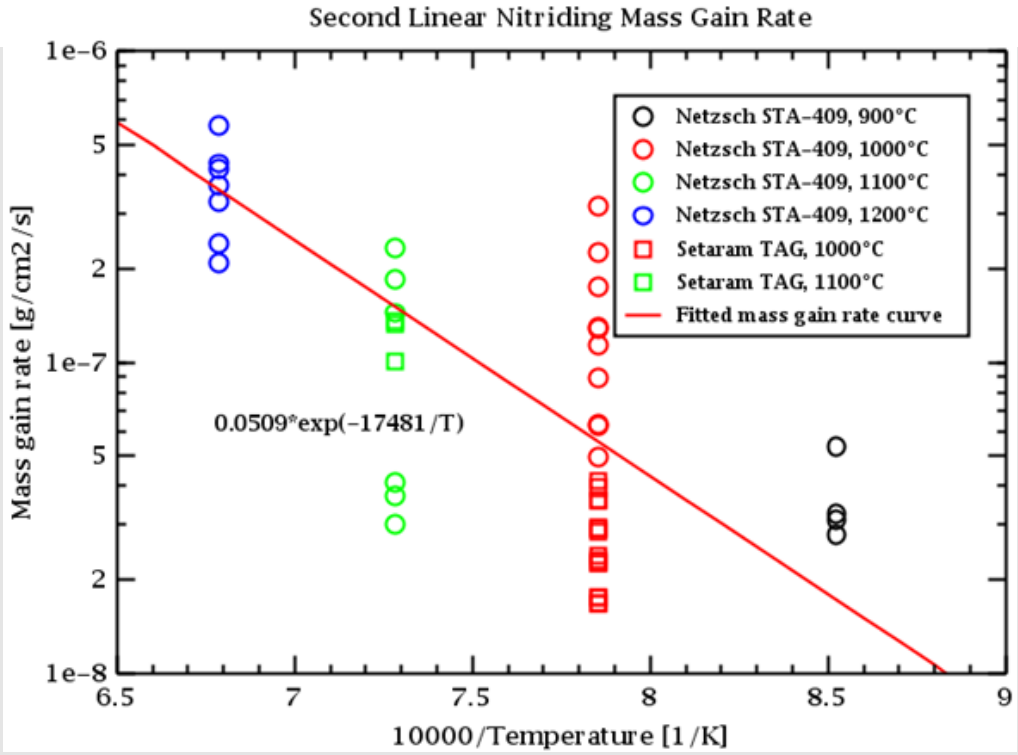
M.Steinbrück, Journal of Nuclear Materials 447, (2014) 46-55

Nitriding model (III)



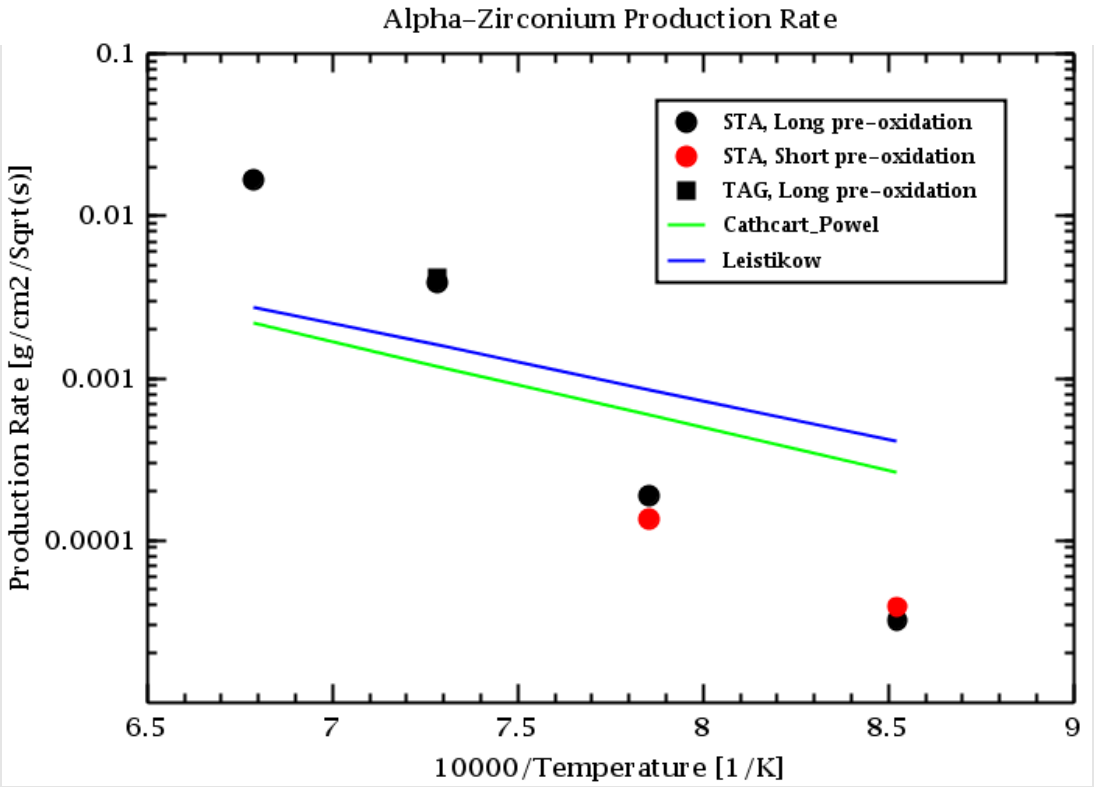
The fast nitriding reaction rates of the tests at different temperatures shows a similar behavior as the pre-oxidation by oxygen. The temperature dependency of the reaction rates can be described with an Arrhenius plot.

Nitriding model (IV)



The slow nitriding reaction rates of the tests at different temperatures are very close to the detection limit of the measurement device, especially if using the Netzsch STA-409 facility at low temperature (900°C). The other reason for the wide spreading of the data is the leakage of up to 200 ppm in this facility.

Nitriding model (V)



Integrating the mass gain during the fast nitriding phase inclusive the transition phase should result in the amount of α-Zr(O). A comparison with the curves of Cathcart-Powel and Leistikow shows a poor estimation.

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Outlook

In a close cooperation with KIT open questions will be solved and additional data will be implemented in this model to extend the temperature range and the credibility of the data.

A standalone code will be written to use the model for post test calculations (model assessment) and further implementation into severe accident codes.

The re-oxidation will be as well implemented using data of this PhD and additional data from KIT.


Questions?

23rd International QUENCH Workshop, 17 - 19 October 2017
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KIT











































Weissenburger
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