

Update of the QUENCH Program

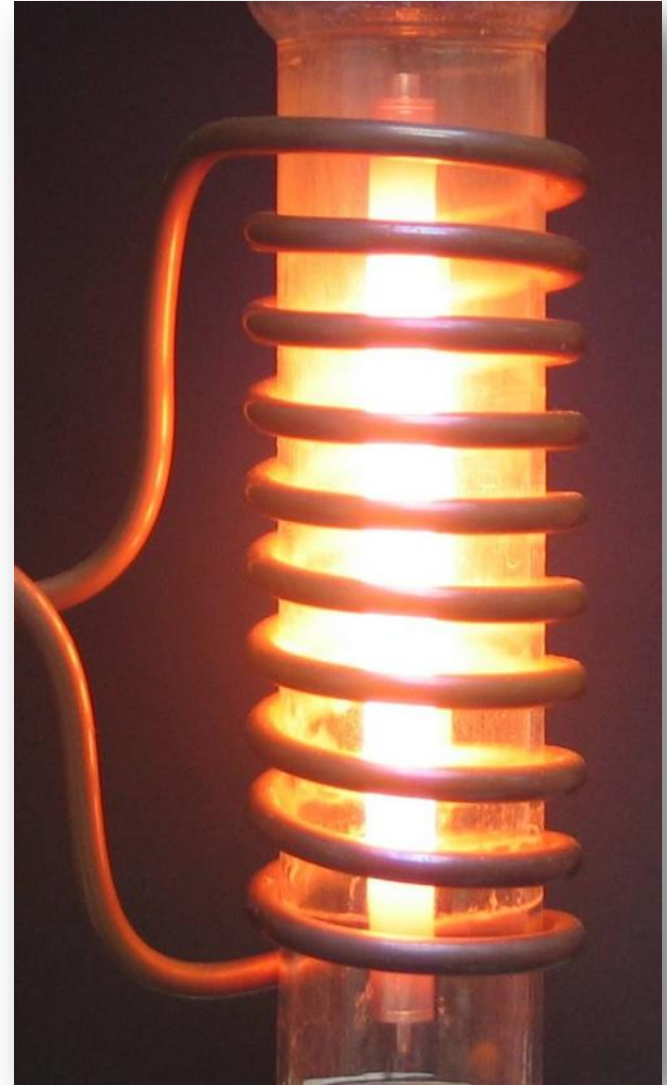
M. Steinbrück, J. Stuckert, M. Große et al.

24th International QUENCH Workshop, Karlsruhe, 13-15 November 2018

Institute for Applied Materials, Programme NUSAFE



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



- 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

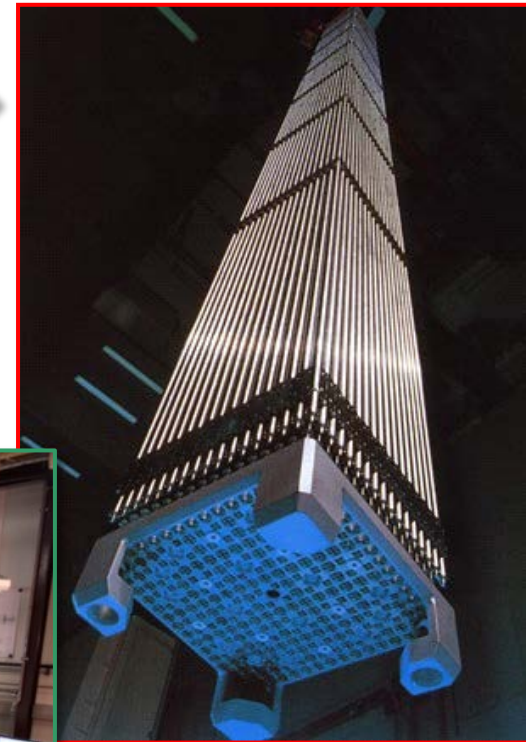
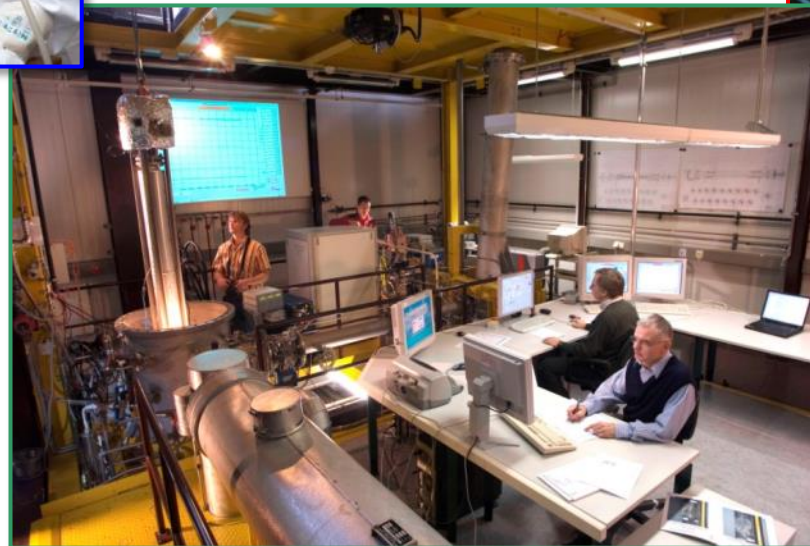
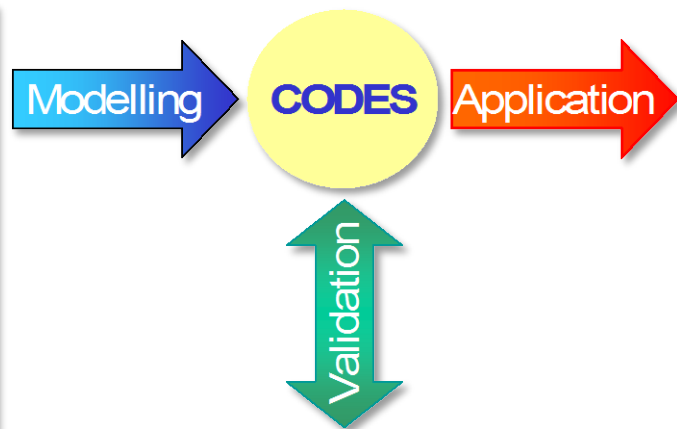
QUENCH Programme

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



Separate-effects tests

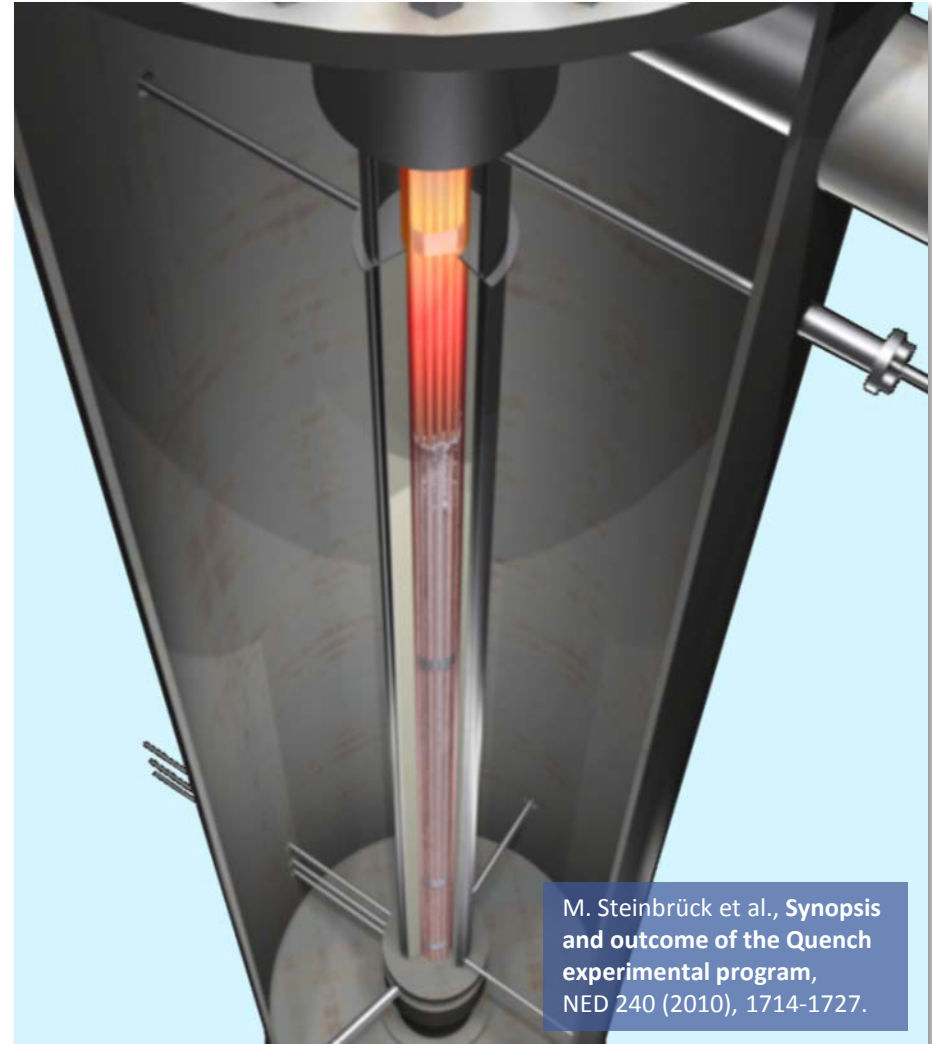
Bundle experiments



PWR fuel element

QUENCH facility

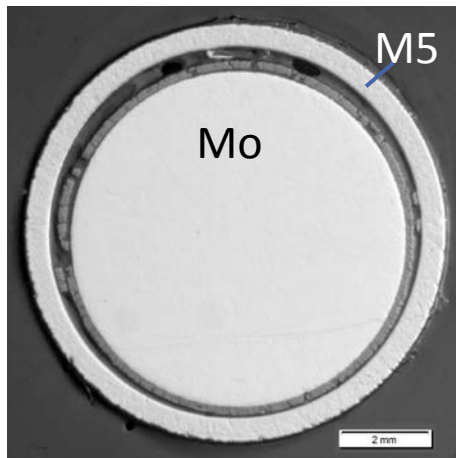
- Unique out-of-pile bundle facility to investigate reflood of an overheated reactor core
- 21-31 electrically heated fuel rod simulators; T up to $>2000^{\circ}\text{C}$
- Extensive instrumentation for T, p, flow rates, level, etc. + MS
- So far, 19 experiments on SA performed (1996-today)
 - Influence 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



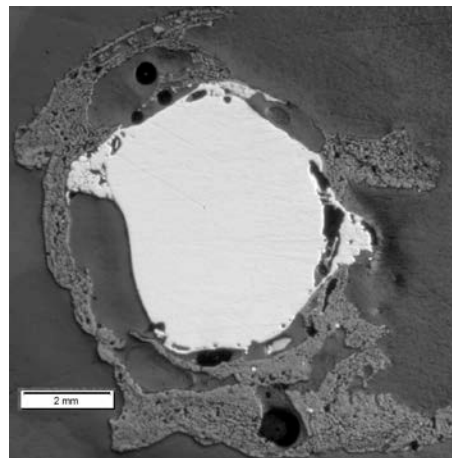
M. Steinbrück et al., *Synopsis and outcome of the Quench experimental program*, NED 240 (2010), 1714-1727.

QUENCH-18

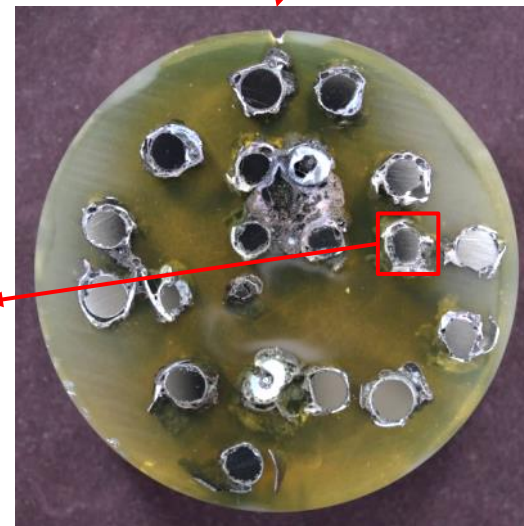
- Successfully conducted in Sept 2017
- In the framework of the EC-China ALISA project
- With M5[®] cladding, two pressurized rods, two Ag-In-Cd absorber rods, and air ingress
- Strongly degraded bundle
- PTE of the main bundle part still pending
- Issue with MS measurements of O₂ concentration in presence of H₂O



Intact rod at 1450 mm

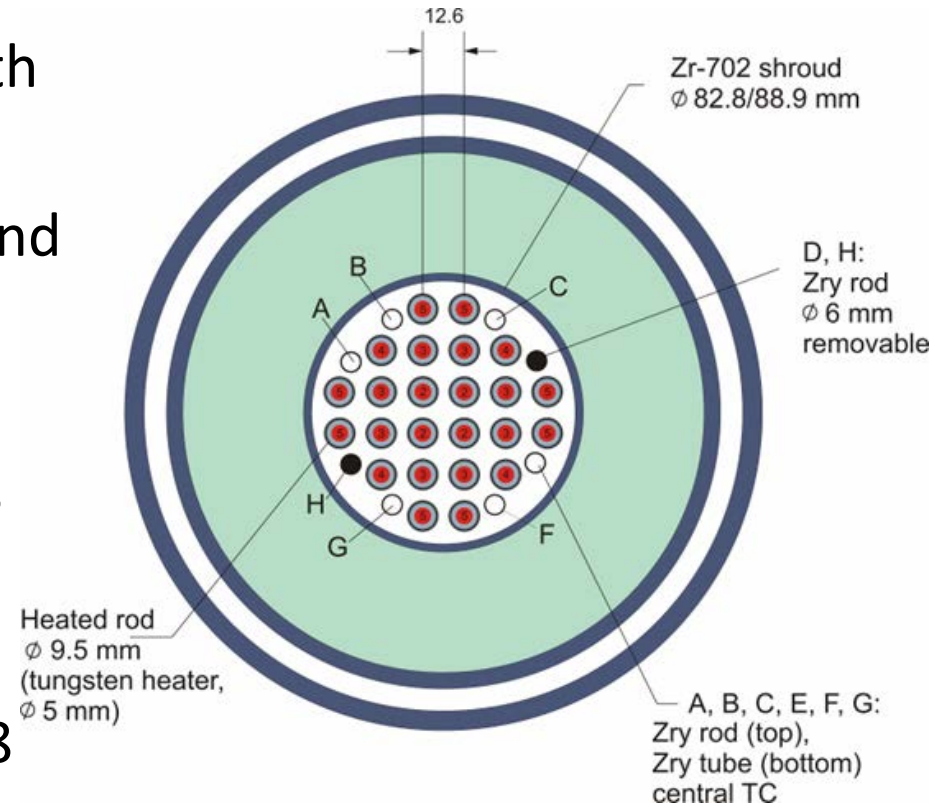


Strongly degraded bundle at 1250 mm



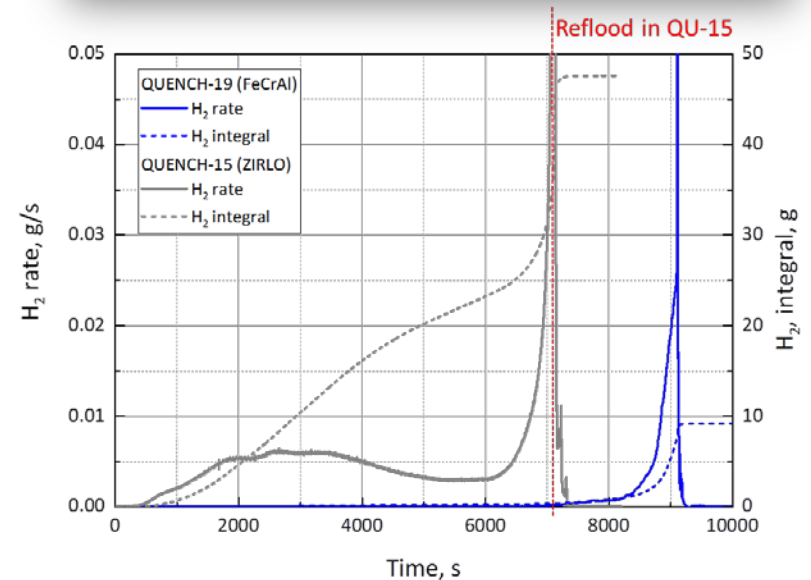
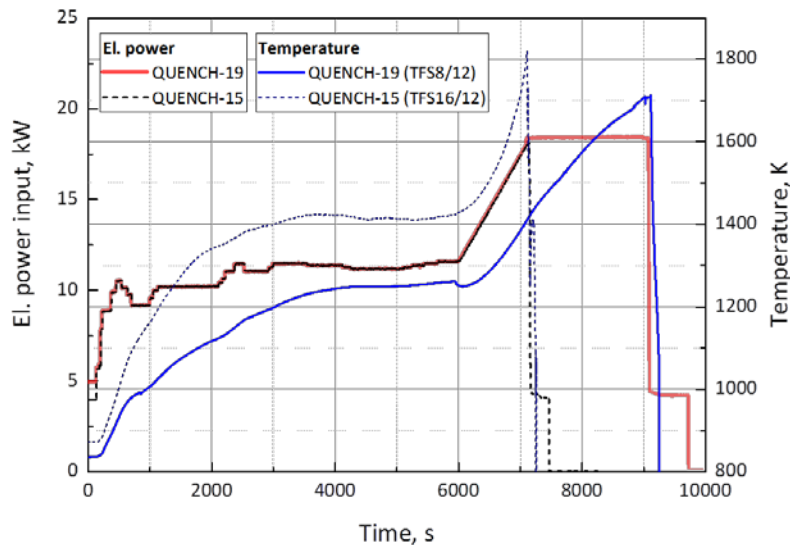
QUENCH-19

- Worldwide first bundle test with ATF cladding
- With FeCrAl cladding, shroud and spacer grids
- In cooperation with ORNL
- Scenario similar to QUENCH-15 (same bundle geometry, same electrical power input)
- Conducted on 29th August 2018



QUENCH-19

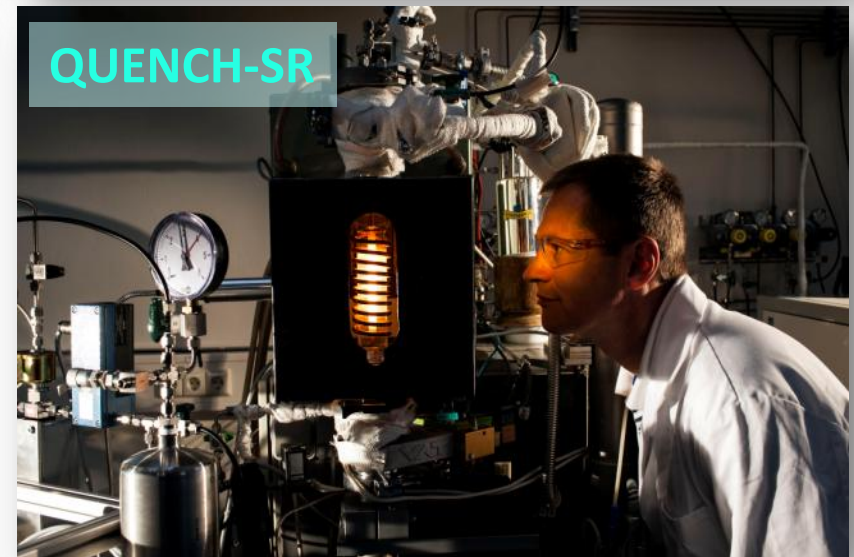
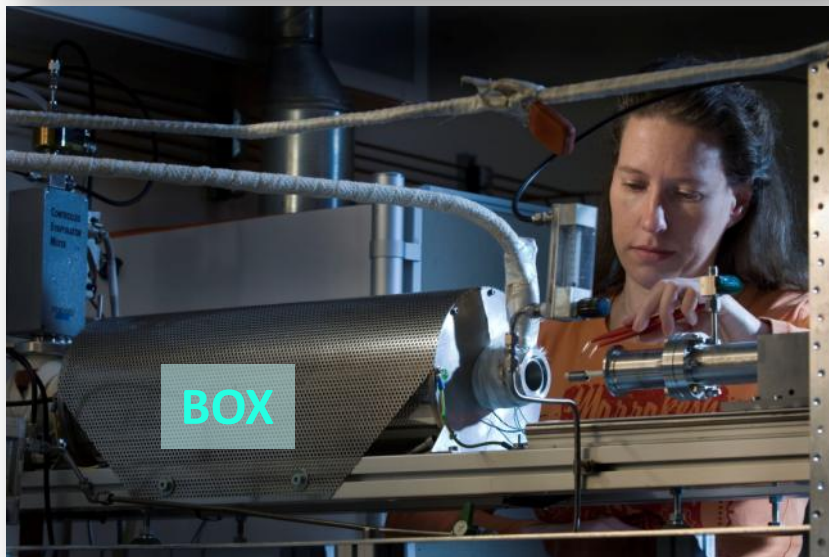
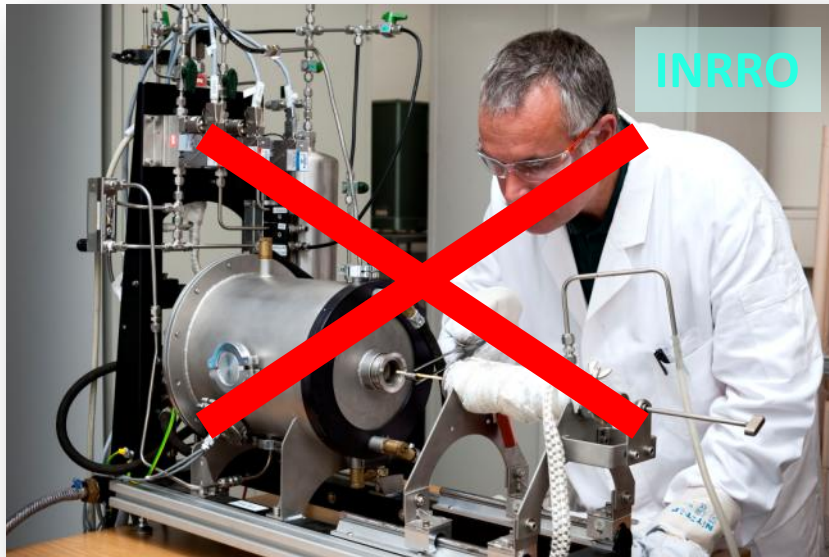
- 100x less hydrogen released up to quenching time of Q-15
- Melting temperature of FeCrAl locally reached
- Significant gain of coping time with FeCrAl compared to Zr alloy



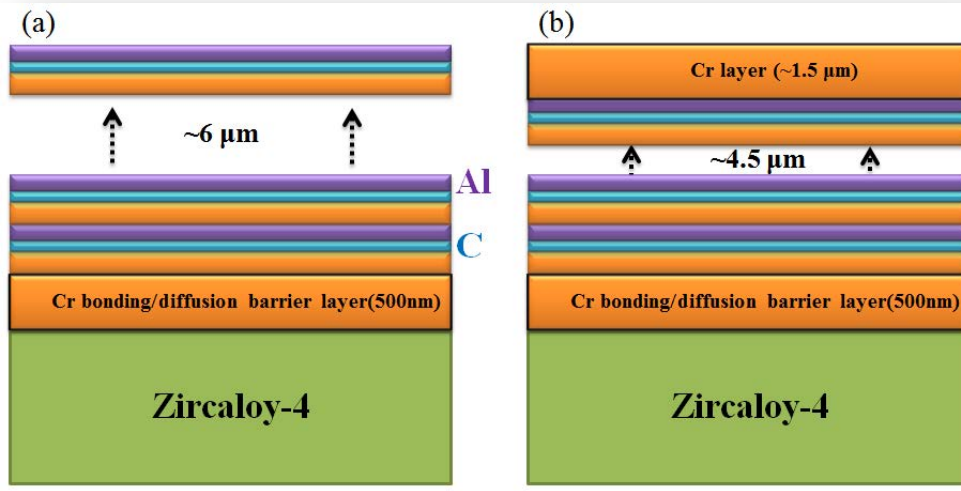
Separate-effects tests in 2018

- Experiments on high-temperature oxidation of silicon carbide ceramic-matrix-composites in various atmospheres for different applications
 - Optimization of Cr_2AlC MAX phase coatings on Zry
 - Autoclave tests with MAX phase coatings at Westinghouse, USA
 - High-temperature oxidation of various ATF cladding materials (Fe alloys, MAX phases, coated Zr alloys...) in the framework of international cooperations
 - Interaction between SiC and Zry
-
- CODEX-AIT3 at MTA Budapest proposed by KIT (SAFEST)

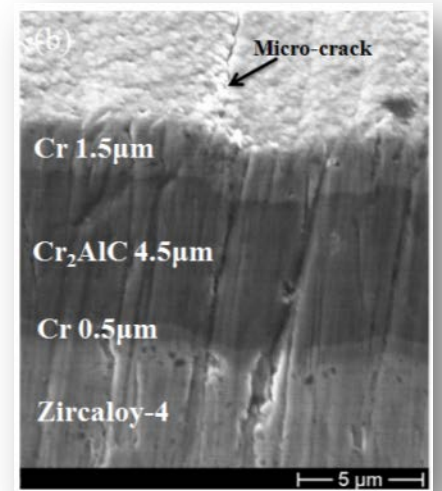
QUENCH Separate-effects tests: Main setups



Cr₂AlC MAX phase coating on Zry-4

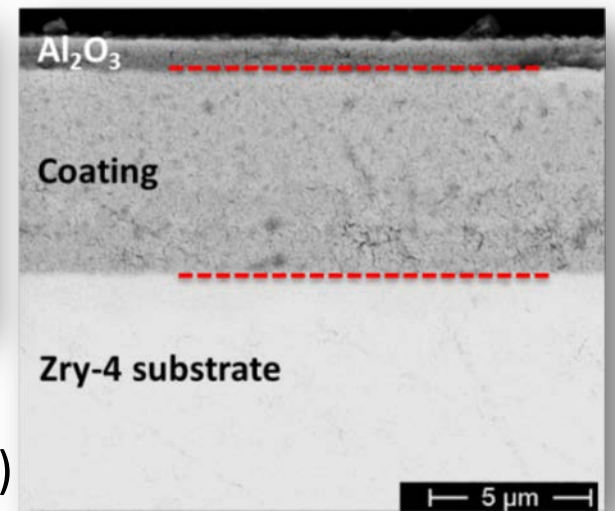
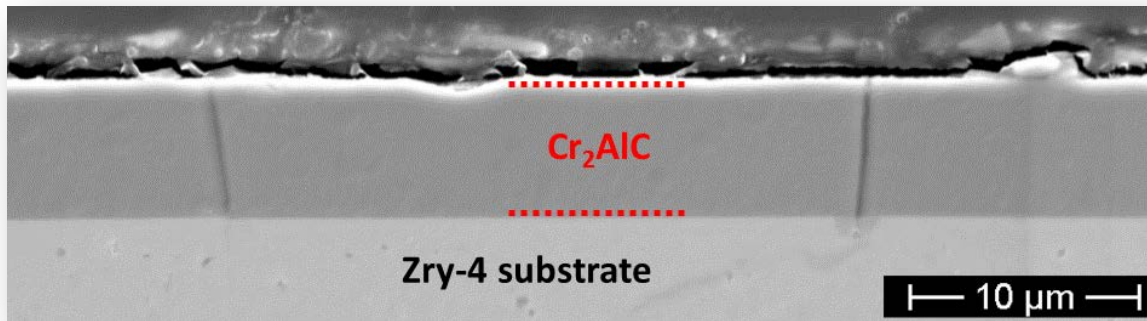


Annealing



Magnetron sputtered Cr/C/Al nano layers

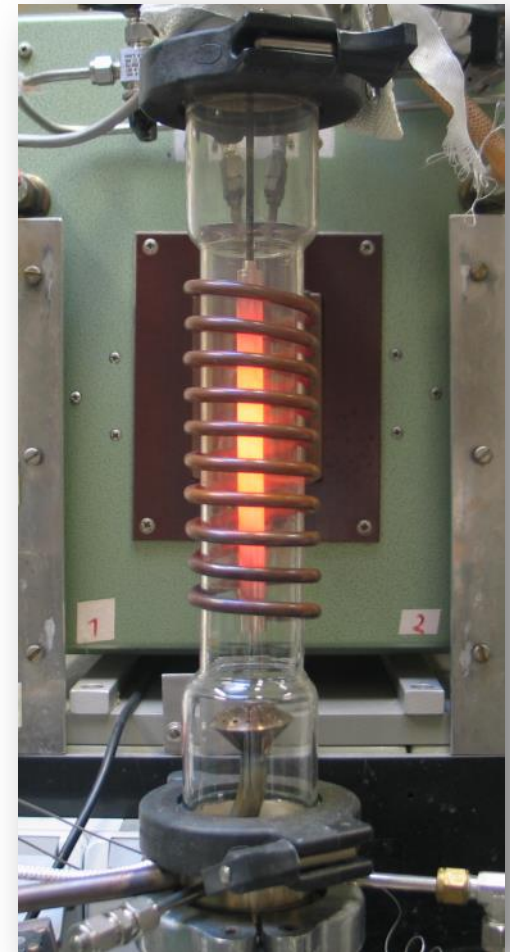
Pure MAX phase



➡ Excellent behavior of coatings during autoclave tests (↑) and HT oxidation in steam up to 1200°C (→)

HT oxidation in steam of SiC_f-SiC cladding

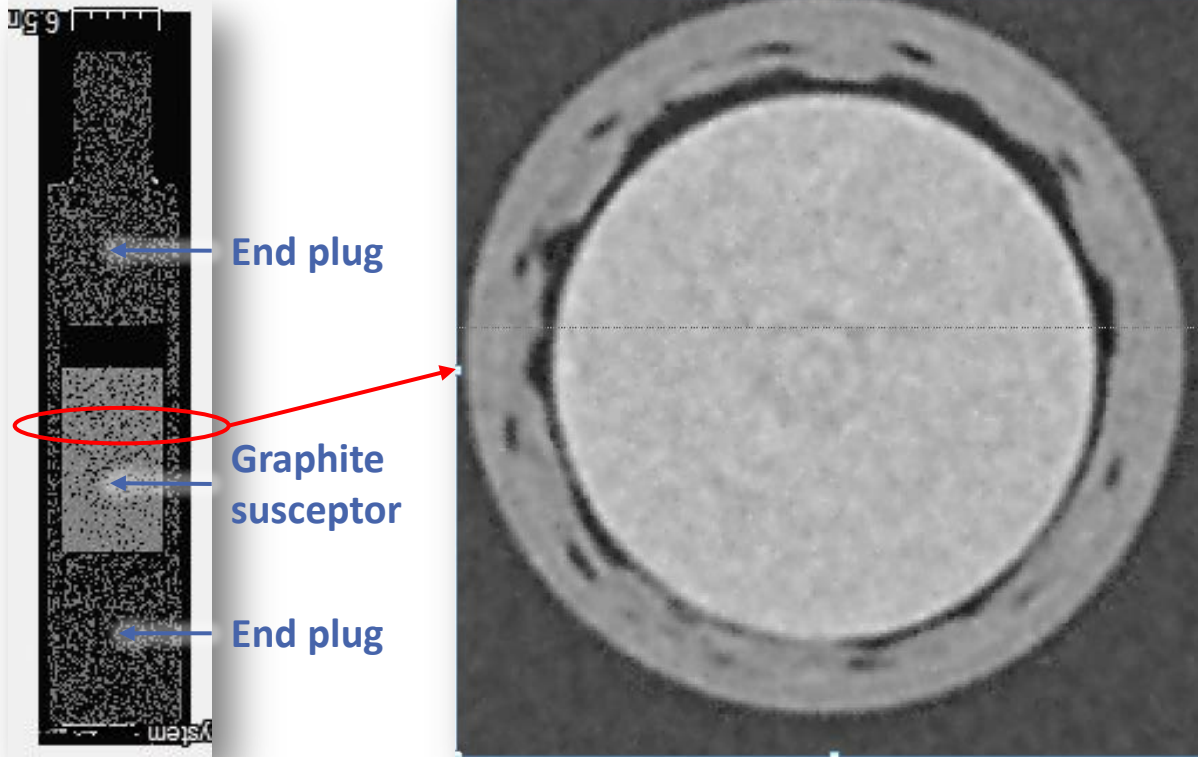
- QUENCH-SR rig with inductive heating of graphite as susceptor
- Two samples
- Four experiments
 - Transient test with target temperature 2200°C (sample 1)
 - Three subsequent isothermal tests at 1600, 1700, and 1750°C terminated by quenching with water (sample 2)



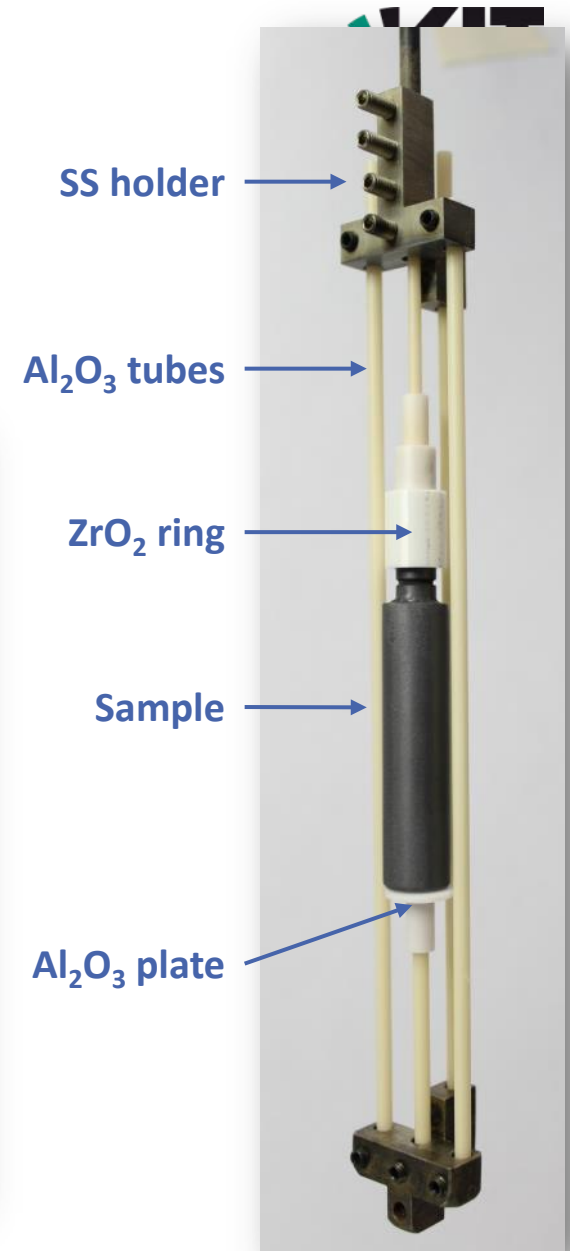
QUENCH-SR with
inductive heating

SiC_f-SiC samples

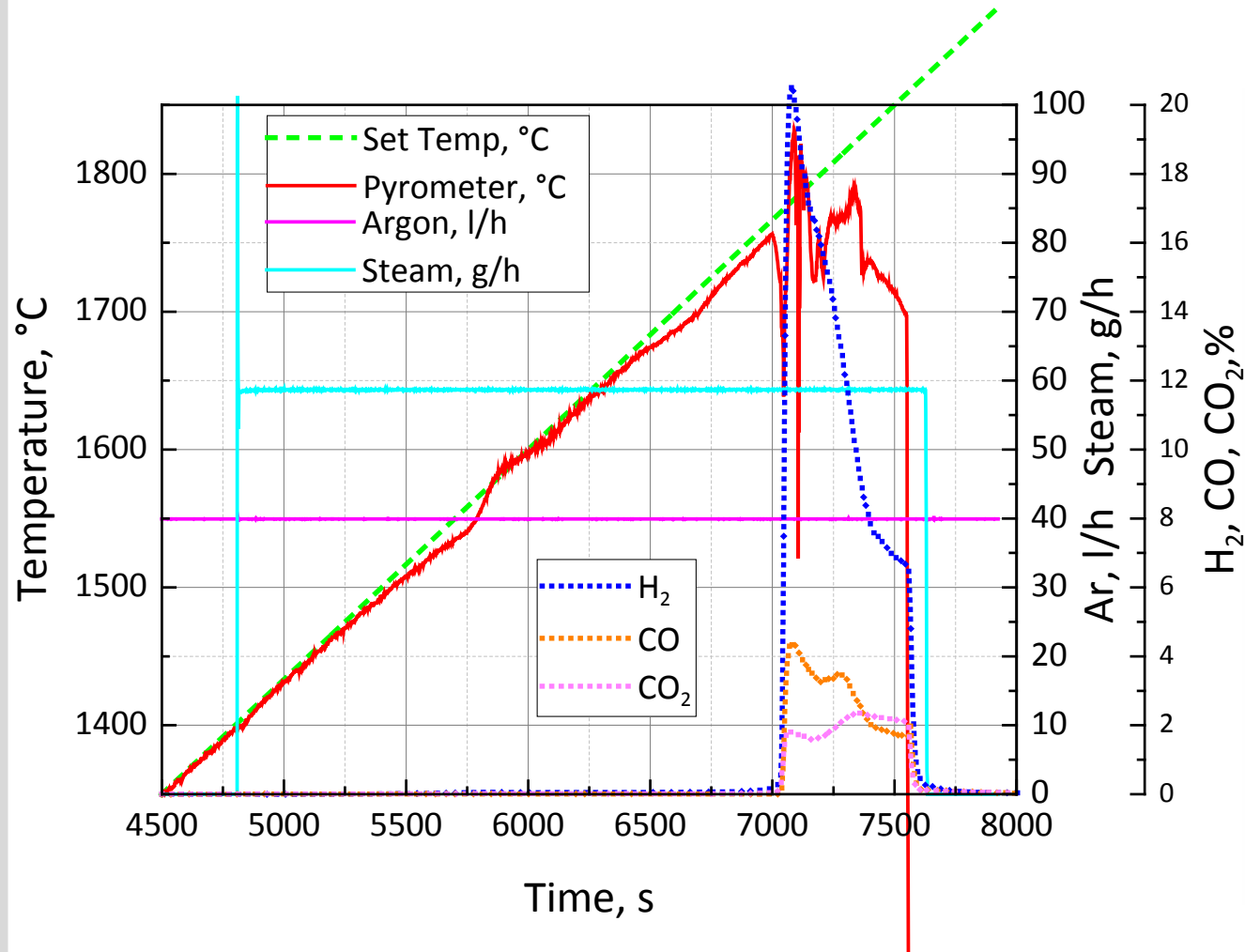
- Provided by General Atomics
- Leak tight with welded end plugs and filled with graphite



Neutron tomography of as-received sample

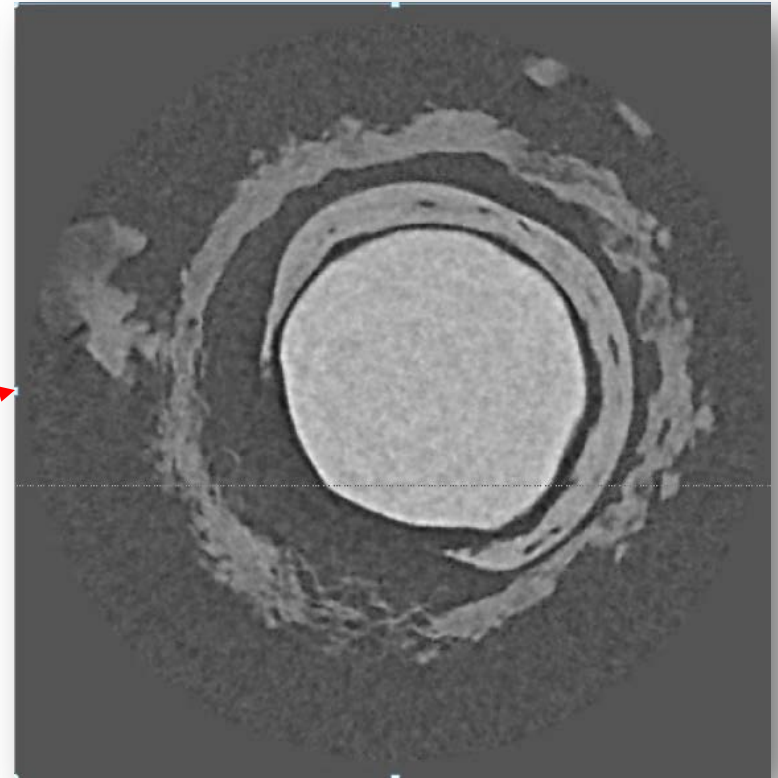
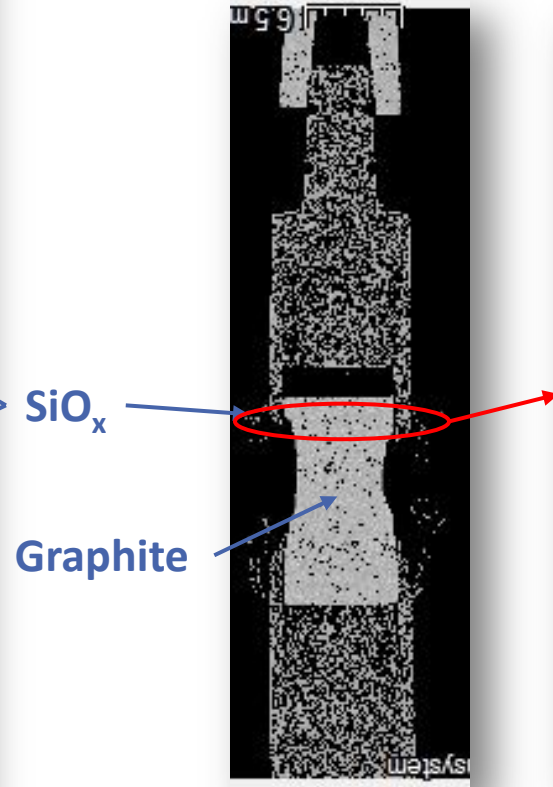
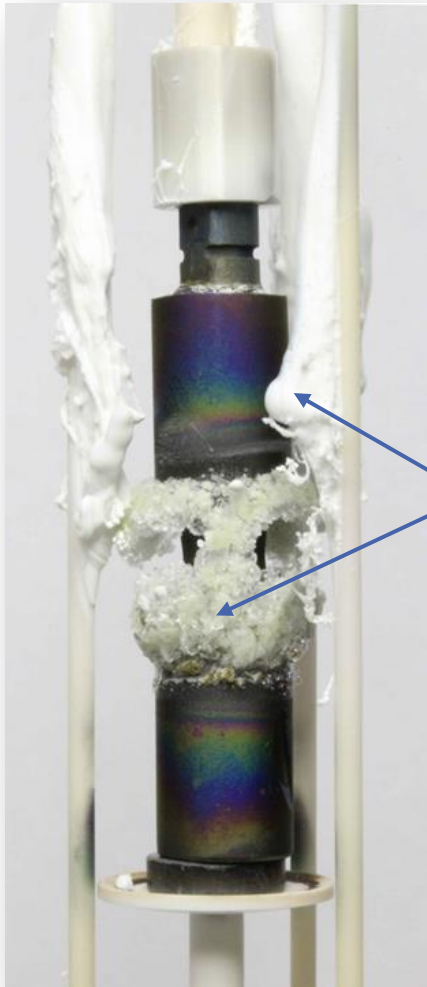


Transient test: Conduct and MS results



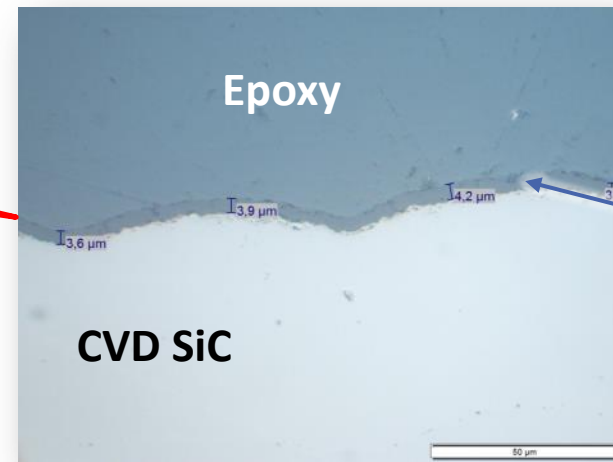
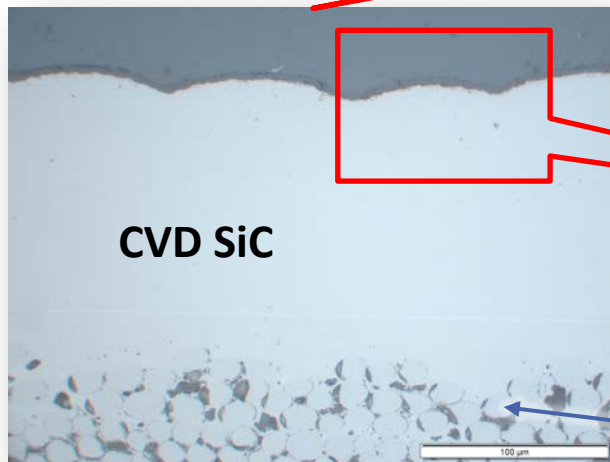
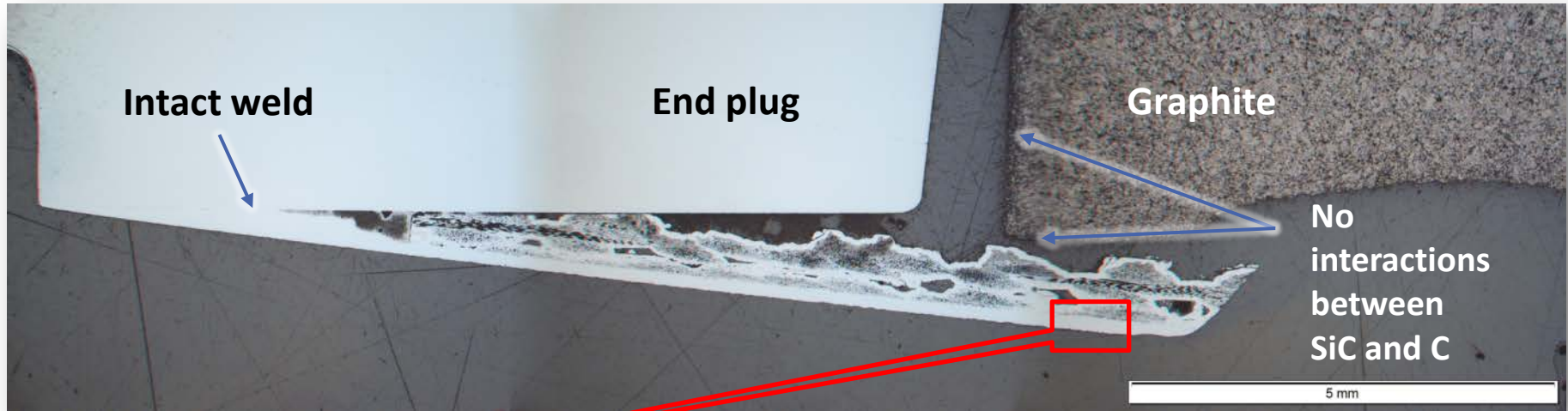
➡ Bubble formation, strong gas release, SiO_x volatilization above $\sim 1750^\circ\text{C}$

Transient test: Post-test appearance



Neutron tomography after transient test

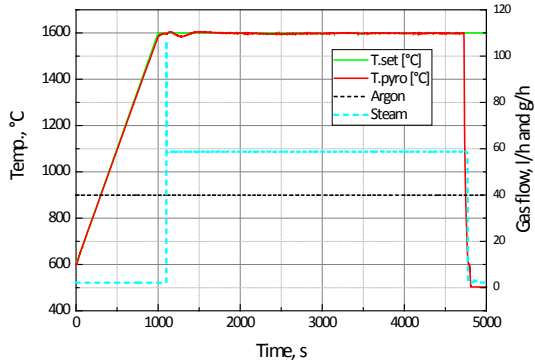
Transient test: Micrographs of longitudinal cross section



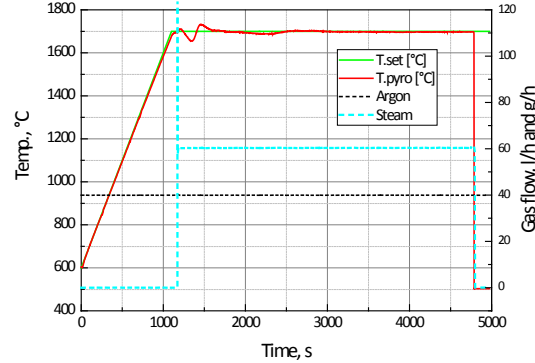
~4 μm
SiO₂

Isothermal tests: conduct and post-test appearance

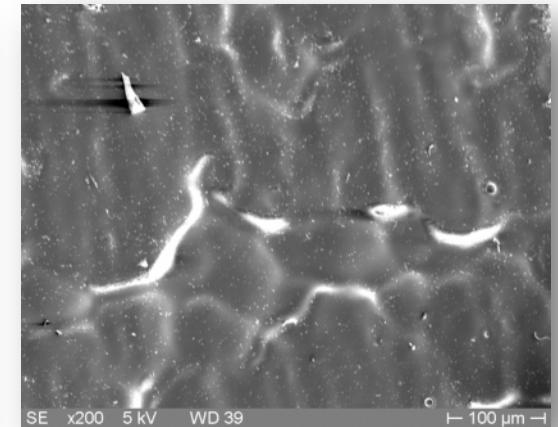
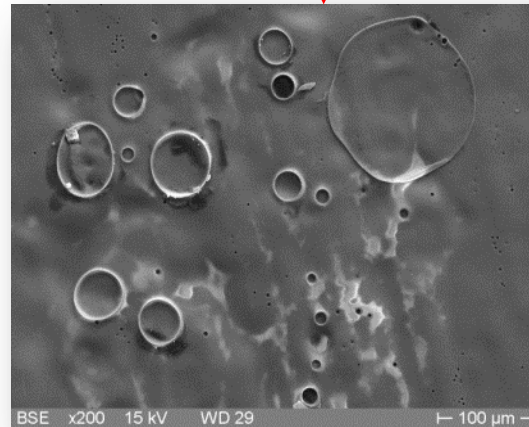
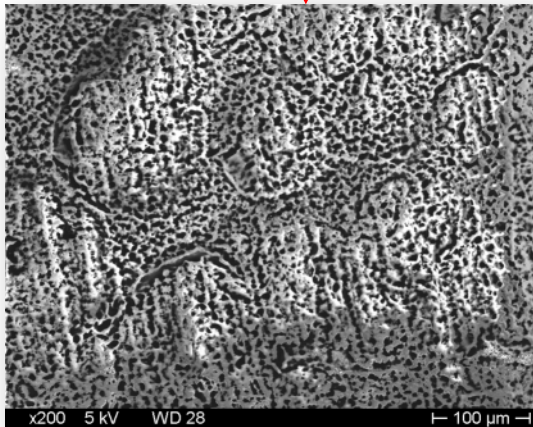
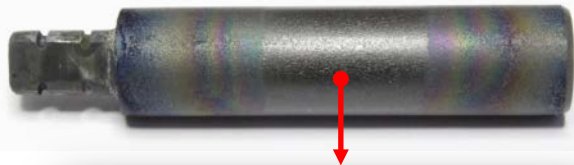
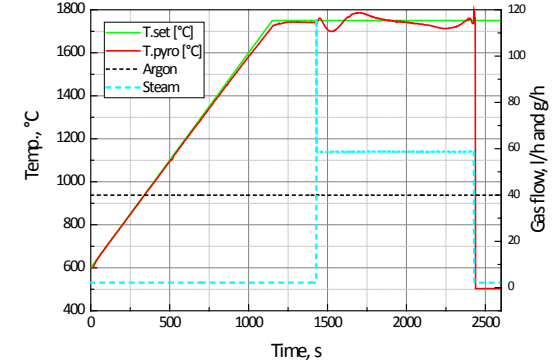
1 h @ 1600°C



1 h @ 1700°C

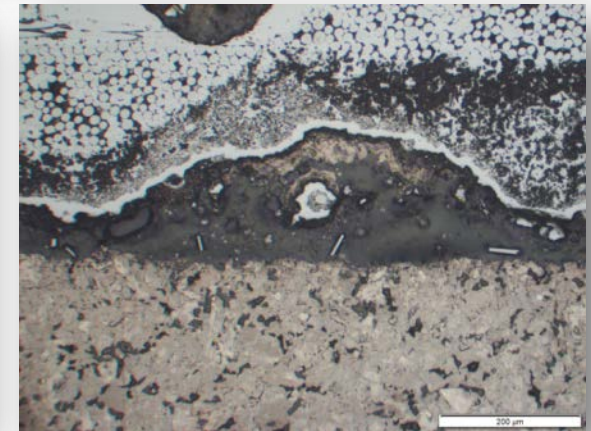
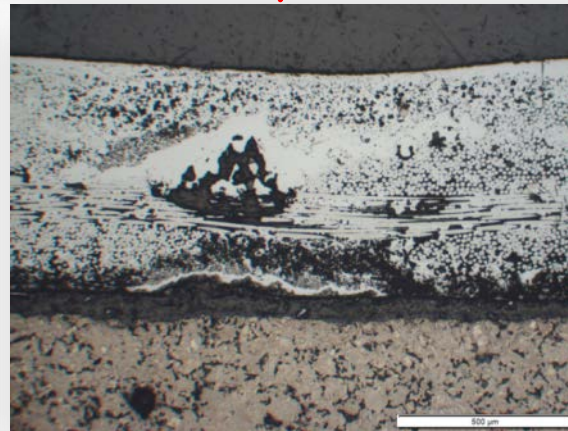
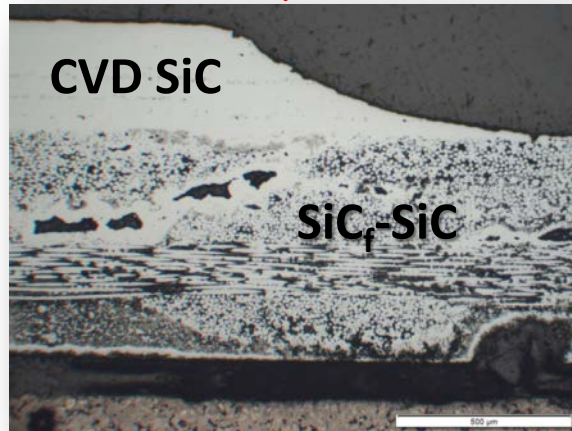


16 min @ 1750°C

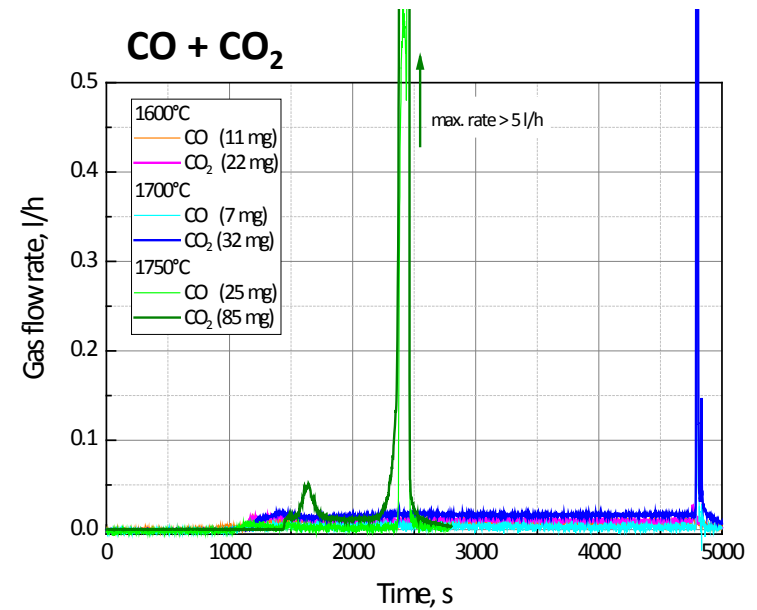
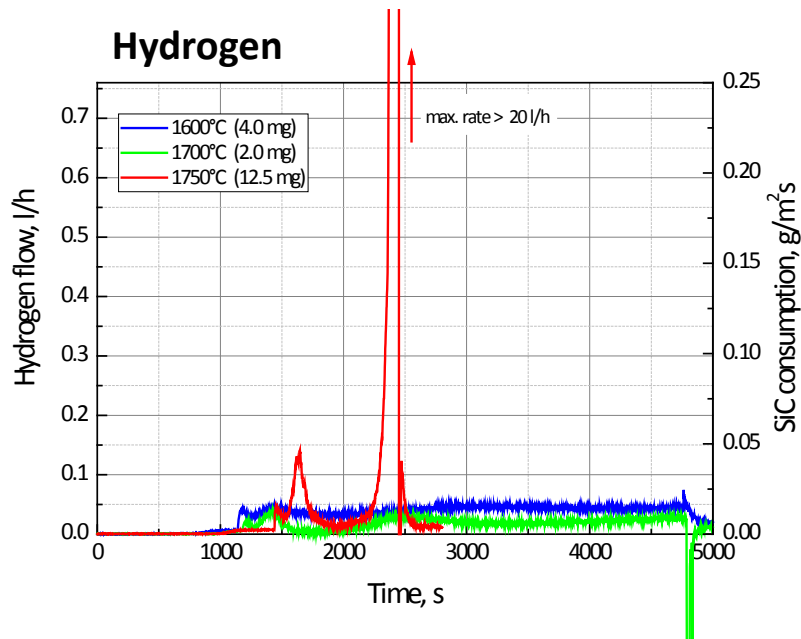


■ SEM/EDX: all surfaces are covered with SiO₂

Isothermal tests: Micrographs of longitudinal cross section



Isothermal tests: Gas release



- Very limited oxidation of the SiC_f-SiC cladding at 1600 and 1700°C
- Local failure of the sample after 16 min at 1750°C resulting in strong gas release
- Failure mechanism not yet identified

- PhD thesis
 - 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, follow-up program under discussion) as well as in the new TOPATF initiative
- Participation in the IAEA CRP on Accident Tolerant Fuel Concepts for Light Water Reactors (ACTOF)
- WP leader (coolant-cladding-fuel interaction) in 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/-19/-20 by various organisations
- Post-test calculations for QUENCH-18 in the framework of the NUGENIA QUESA project by GRS, PSI, IBRAE, LEI, EdF and of QUENCH-19 by GRS
- QUENCH data were used in the frame of IAEA FUMAC project
- RELAP5/SCDAPSIM analyses of various QUENCH tests
- 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: KIT Scientific Reports available online

■ LOCA summary paper planned for ASTM Symp. Zr in Nucl. Ind. 2019

■ Papers, book chapters and conference contributions (>15 Scopus references)

■ Plenary talk at NUMAT 2018

STP 1597, 2018 / available online at www.astm.org / doi: 10.1520/STP159720160041

Mirco Grosse,¹ Martin Steinbrück,¹ Burkhard Schilling² and Anders Kaestner¹

In Situ Investigations of the Hydrogen Uptake of Zirconium Alloys during Steam Oxidation

Citation

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ABSTRACT

The hydrogen uptake during steam oxidation of Zircaloy-4 and E110 was



Phenomenology of BWR fuel assembly degradation

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HIGHLIGHTS

• Phenomenology focusing on BWR fuel degradation is discussed, including several concerns arisen from the FDNPS accident.

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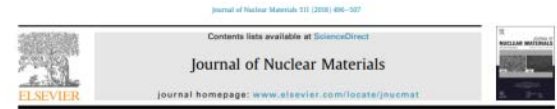
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Control blade
Channel box
Phase diagram
U-D-O system
UO₂-ZrO₂ system
K₂CO₃-NaNO₂ molten salt
K₂CO₃-ZrO₂ system
Radiation interaction model
Hydrogen uptake in Zr
B₂O₃ oxidation
Fukushima-daiichi nuclear power plant accident

Contents

1. Introduction	120
2. Integral tests with B ₂ O ₃ control blade	121
2.1. US and Canadian tests	121
2.2. CORA BWR tests	121
2.3. Control blade degradation test in JAEA	122
2.4. Potential effect of B ₂ O ₃ in the FDNPS accident	122
3. Integral tests with B ₂ O ₃ absorber rods	125

Abbreviation: FDNPS, Fukushima-Daiichi Nuclear Power Station; SA, Severe Accident; BWR, Boiling Water Reactor; PWR, Pressurized Water Reactor.
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Influence of composition and heating schedules on compatibility of FeCrAl alloys with high-temperature steam

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HIGHLIGHTS

• Catastrophic oxidation or protective effect was observed during high-temperature steam oxidation of FeCrAl-based alloys.
• The occurrence of catastrophic oxidation was not only determined by the chemical composition, but also considerably influenced by the applied heating schedules.
• Reactive element (Y) can significantly improve the high-temperature steam tolerance of FeCrAl alloys during the transient tests.
• The competition between mass transfer of Al from the substrate and transport of oxidizing gas through the scale determine the behavior of the alloys.

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ABSTRACT

FeCrAl alloys are proposed and being intensively investigated as alternative accident-tolerant fuel (ATF) cladding for nuclear fission applications. Herein, the influence of major alloy elements (Cr and Al), reactive element effect and heating schedules on the oxidation behavior of FeCrAl alloys in steam up to 1900 °C was examined in case of transient ramp tests, catastrophic oxidation, i.e. rapid and complete consumption of the alloys, occurred during temperature ramps up to above 1200 °C for specific alloys. The



Original Article High-temperature interaction of oxygen-preloaded Zr1Nb alloy with nitrogen

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ABSTRACT

Potential air ingress scenarios during accidents in nuclear reactors or spent fuel pools have raised the question of the influence of air, especially of nitrogen, on the oxidation of zirconium alloys, which are used as fuel cladding tubes and other structural materials. In this context, the reaction of zirconium with nitrogen-containing atmospheres and the formation of zirconium nitride play an important role in understanding the oxidation mechanism. This article presents the results of the interaction of the oxygen-preloaded niobium-bearing alloy M5[®] with nitrogen over a wide range of temperatures (800–1400 °C) and oxygen contents in the metal alloy (1–7 wt %). A strongly increasing oxidation rate with rising oxygen content in the metal was found. The highest reaction rates were measured for the saturated ZrO₂ as it exists at the metal-oxide interface, at 1300 °C. The temperature maximum of the reaction rate was approximately 100% higher than for Zircaloy-4, already investigated in a previous study [1]. The article presents results of thermogravimetric experiments as well as post-test examinations by optical microscopy, scanning electron microscopy (SEM), and microprobe elemental analyses. Furthermore, a comparison with results obtained with Zircaloy-4 will be made.
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1. Introduction

Zirconium alloys are used worldwide in the nuclear industry for fuel claddings and other structural materials because of their good mechanical properties and corrosion resistance at operational conditions as well as because of their low cross section for thermal neutrons. However, during loss-of-coolant accidents as well as severe nuclear accidents, the high-temperature oxidation of zirconium by water steam causes serious degradation of the mechanical properties of cladding tubes, resulting in the loss of barrier effect against the release of fission products, as well as in the production of hydrogen and chemical heat. The hydrogen source term due to the zirconium-steam reaction carries the risk of hydrogen detonation, as was seen during the Fukushima Daiichi accident. The chemical heat produced by the oxidation reaction may exceed the nuclear decay heat at high temperatures, thus becoming a driving force for thermal excursions of the reactor core [2].

Numerous studies on the oxidation of zirconium alloys in water steam have been conducted over a wide range of temperatures and conditions during the last decades, mainly triggered by the TMI-2

accident 1979 in the United States [1,4]. Air ingress scenarios in reactors have also been discussed for a long time; see for example the study by Powers et al. [5]. After the Fukushima accidents, air ingress accident scenarios for spent fuel pools came into focus for international research [6].

It is known from many experimental studies that the oxidation of zirconium alloys by steam or oxygen is strongly affected by the presence of nitrogen [7] [8–12]. Especially in the temperature range between 800 °C and 1200 °C, the reaction kinetics are strongly increased by nitrogen, resulting in more severe degradation of the cladding and higher hydrogen source terms (if steam is available) compared to oxidation in pure steam and oxygen atmosphere. The understanding of the mechanism of nitrogen attack is that zirconium nitride, ZrN, is temporarily formed and subsequently oxidized; this is connected with serious volume mismatches because of the different densities of the involved phases, and with this the formation of non-protective oxide scales. Furthermore, it is known that, on the one hand, the reaction of zirconium with nitrogen is very slow due to the formation of a thin protective nitride scale and low diffusion coefficients of nitrogen in Zr [7]; but, on the other hand, much ZrN is formed during the reaction of oxygen-stabilized zirconium with nitrogen [7]. Conditions favorable for the formation of ZrN are present locally at the metal-oxide interface after consumption of oxygen when nitrogen is in contact with the oxygen-

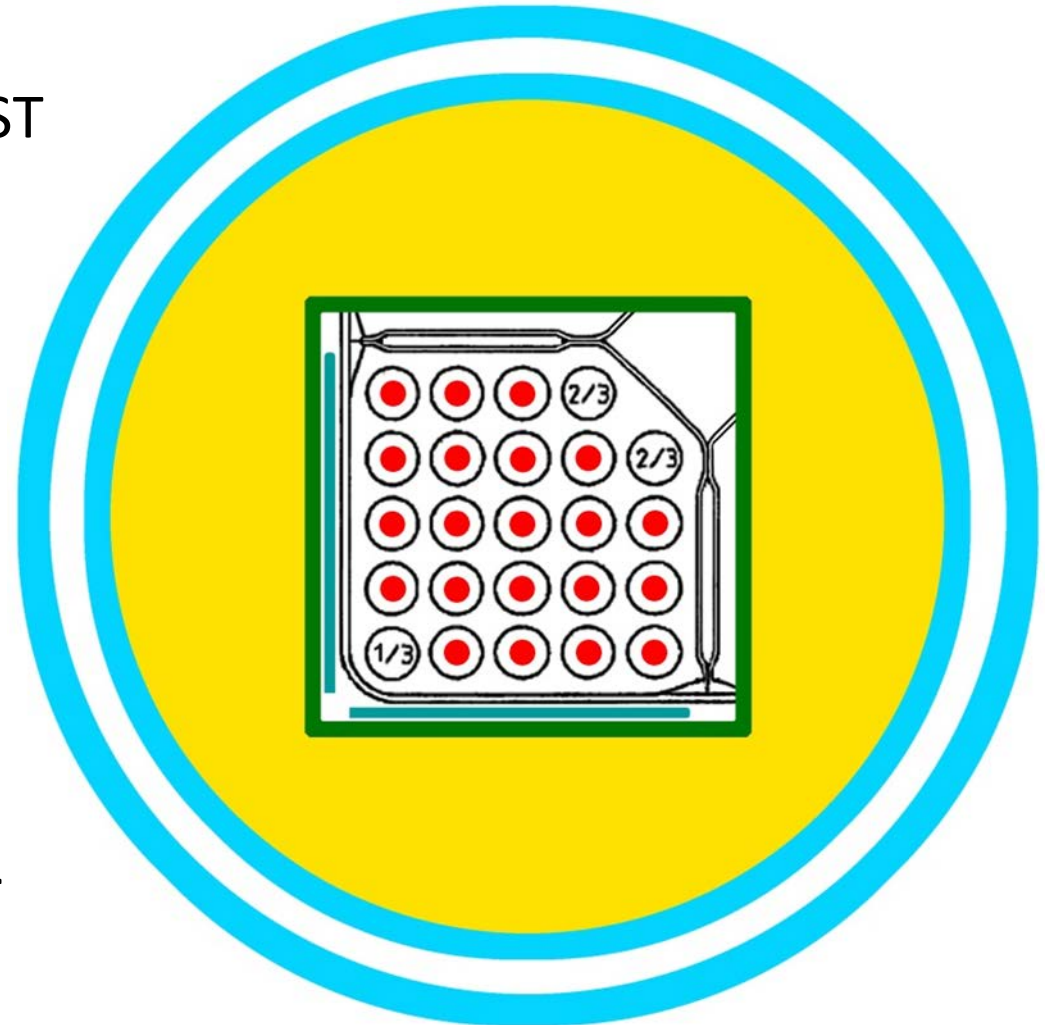
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- BWR bundle test within the framework of the EC SAFEST program beginning 2019
- Post-test examinations of QUENCH-18/-19/-20
- Discussion with various international partners on further bundle tests with ATF claddings
- SETs on various further topics with the focus on ATF cladding high temperature oxidation
- Activities on safety of long-term dry intermediate storage

BWR bundle test QUENCH-20

- Planned to be conducted beginning 2019 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



Co-operations

Programs

- NUGENIA
- HORIZON 2020
- IAEA
- OECD-NEA

Bilateral

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



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Update of the QUENCH Program

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

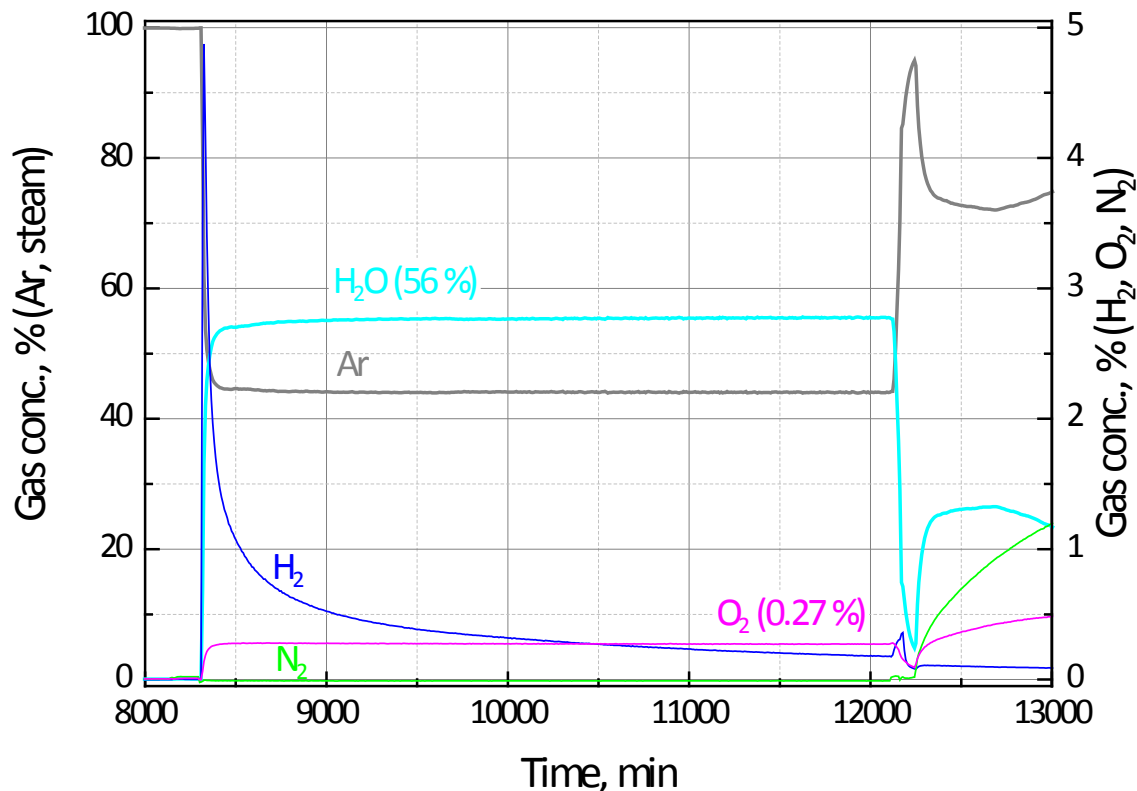
24th International QUENCH Workshop, Karlsruhe, 13-15 November 2018

Institute for Applied Materials, Programme NUSAFE



MS issue with O₂ signal in presence of steam

- Since July 2017 after change of filaments
- Small artificial O₂ signal (<1%) in presence of steam without any correlation to other signals (N₂, H₂)



Example:
Zry-4 oxidation in steam
at 1200°C