Lessons learned
from the QUENCH Programme at FZK

M. Steinbrück, **M. Große, L, Sepold, J. Stuckert**

15th Int. QUENCH Workshop, Karlsruhe Institute of Technology, 3-5 Nov. 2009

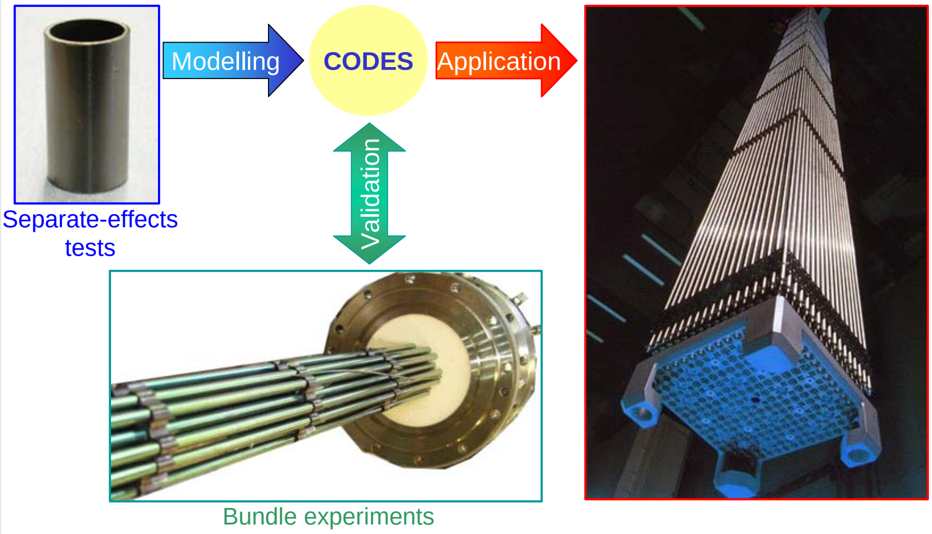
Institute for Materials Research I and III

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
  - TMI2, LOFT, PBF, CORA …
  - .. but only qualitative hydrogen measurements
- Coolability of a degraded core is a matter of high priority (SARNET-SARP, OECD-GAMA)

➨ QUENCH experiments (bundle+SET) provide data for development of models and validation of SFD code systems

QUENCH Programme



Basic information

Reflood
Water injection → steam production

Cooling effect — **Chemical heat production**

$Q_c \sim h\,(T_{Zr} - T_c)$

$Q_R \sim \Delta_R H\,(K/t)^{0.5}$

$Zr + 2\,H_2O \rightarrow ZrO_2 + 2\,H_2 + 580\,kJ$

Influenced by

- ΔT
- Flooding rate
- Cooling medium
- Pressure
- Hydraulic diameter
- …

- **Oxidation kinetics**
- Temperature (Arrhenius type)
- Degree of pre-oxidation
- Surface available for oxidation
- Material composition
- **Steam availability**

Simulation of bundle test with and without excursion

Quenching without temperature escalation

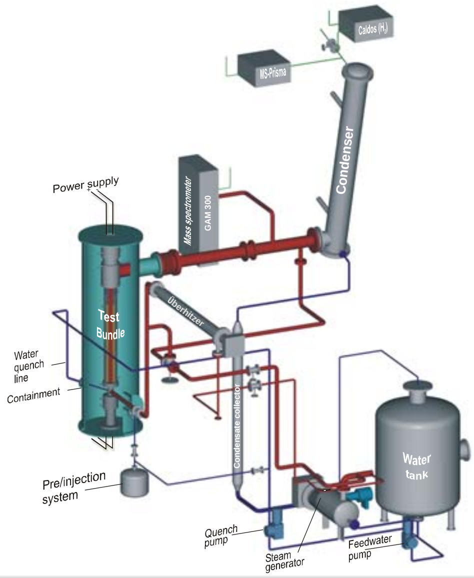
- Fast cool-down of the bundle
- Low to moderate hydrogen (FP) release
- Intact bundle

Quenching with temperature escalation

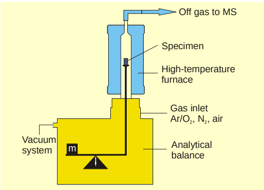
- Temperature escalation
- Strong hydrogen (FP) release
- Formation, relocation and oxidation of melts
- Strongly degraded bundle

QUENCH Facility

- Bundle with 21-31 fuel rod simulators of ~2,5 m length
- Electrically heated: ~1 m; max 70 kW
- Fuel simulator: ZrO_2 pellets
- Quenching (from bottom) with water or saturated steam
- Off-gas analysis by mass spectrometer (H_2, steam …)
- Extensive instrumentation for T, p, flow rates, level, etc.
- Removable corner rods during test



QUENCH Separate-effects tests: Main setups

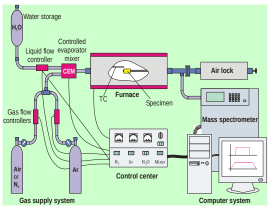


Thermo balance

1600 °C
1250 °C (steam)

Specimens: 0-2 cm

MS coupling



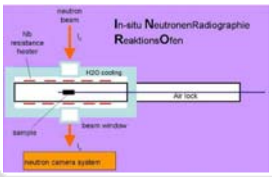
BOX Facility

1700 °C

Oxidising, reducing atmosphere (incl. steam)

Specimens: 1-2 cm

MS coupling

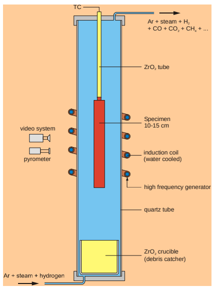


INRO Facility

1500°C

Specimens: 1-2 cm

Transparent for neutrons



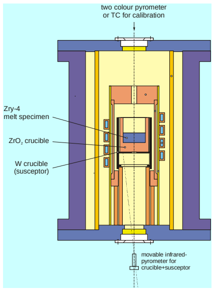
QUENCH-SR Rig

2000 °C

Oxidising, reducing atmosphere (incl. steam)

Specimens: 15 cm

MS coupling



LAVA Furnace

2300 °C

Inert, reducing atmosphere

Specimens: 1-2 cm

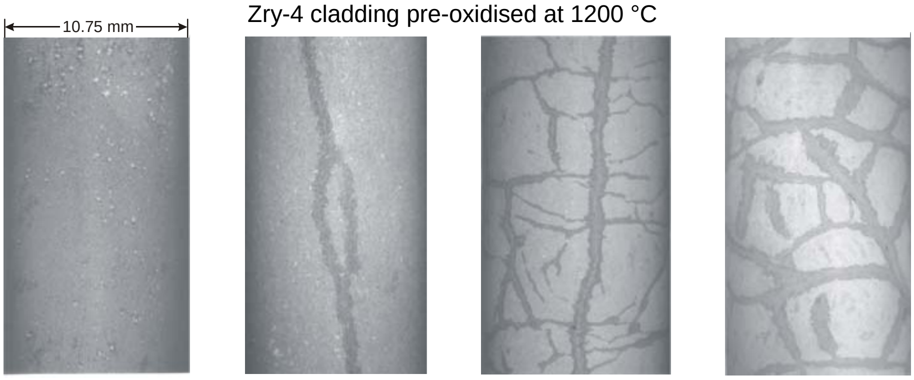
MS coupling

Phenomena of interest (for coolability)

- Crack oxidation
- Temperature at reflood initiation
- Influence of absorber materials (B_4C, AgInCd)
- Melt formation and oxidation
- Steam starvation
- Air ingress
- Reflood rate
- Materials, alloy composition
- Hydrogen absorption

Crack formation during reflood

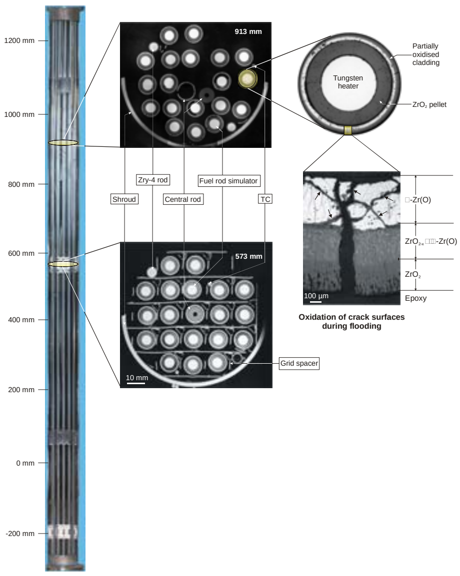
Zry-4 cladding pre-oxidised at 1200 °C



≈ 150 µm ≈ 200 µm ≈ 250 µm ≈ 350 µm

- Density of through-wall cracks: 0.5 mm/mm^2
- Oxidation of fresh-generated metallic surfaces
- Enhanced hydrogen absorption

Crack formation in QUENCH-01 bundle test



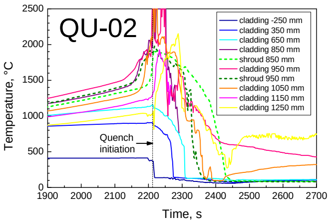
- Estimation of hydrogen production by crack oxidation gives only ca. 1-2 g for the whole bundle

But:

- Solution enthalpy of 1 g H_2 in 1 kg Zr increases temperature by 200 K

Temperature at reflood initiation

Test Date	Quench medium and injection rate	Temp. at onset of flooding	H_2 production before / during cooldown
QUENCH-01 Febr 26, 1998	**Water 52 g/s**	**≈ 1830 K**	**36 / 3**
QUENCH-02 July 7, 1998	**Water 47 g/s**	**≈ 2400 K**	**20 / 140**
QUENCH-03 Jan 20, 1999	**Water 40 g/s**	**≈ 2350 K**	**18 / 120**
QUENCH-04 June 30, 1999	**Steam 50 g/s**	**≈ 2160 K**	**10 / 2**
QUENCH-05 March 29, 2000	**Steam 48 g/s**	**≈ 2020 K**	**25 / 2**
QUENCH-06 Dec 13, 2000	**Water 42 g/s**	**≈ 2060 K**	**32 / 4**



- Escalation of temperatures and significant hydrogen release during tests with maximum temperatures exceeding Zry melting temperature

Bundle tests QUENCH-07/-08/-09 (B_4C effect)

Test Date	Quench medium and injection rate	Temp. at onset of flooding	H_2 production before / during cooldown	Remarks
QUENCH-07 July 25, 2001	**Steam 15 g/s**	**≈ 2100 K**	**66 / 120**	Impact of B_4C absorber rod failure on bundle degradation and H_2, CO, CO_2, and CH_4 generation.
QUENCH-08 July 24, 2003	**Steam 15 g/s**	**≈ 2090 K**	**46 / 38**	As QUENCH-07, no absorber rod
QUENCH-09 July 3, 2002	**Steam 49 g/s**	**≈ 2100 K**	**60 / 400**	As QUENCH-07, steam-starved conditions prior to cooldown.

- Strong influence of boron carbide on hydrogen source term and bundle degradation due to
  - Eutectic interactions between B_4C, SS, and Zry leading to rapid melt formation at about 1200 °C
  - High oxidation potential of boron carbide
    e.g. $B_4C + 8\,H_2O \rightarrow 2\,B_2O_3 + 8\,H_2 + CO_2 + 792$ kJ

Influence of B_4C absorber rod

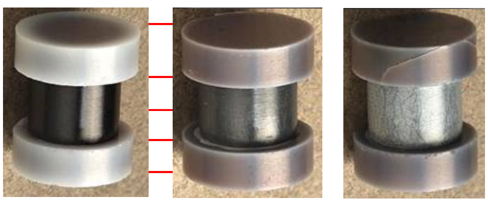
QUENCH-07 with B_4C

Bundle cross sections

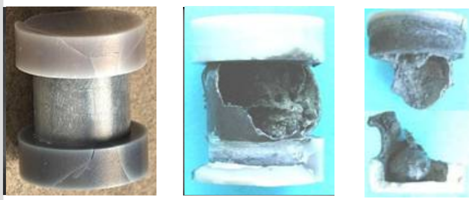
QUENCH-08 w/o B_4C

550 mm | 750 mm | 950 mm

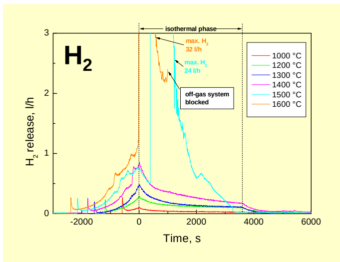
Degradation of boron carbide control rods

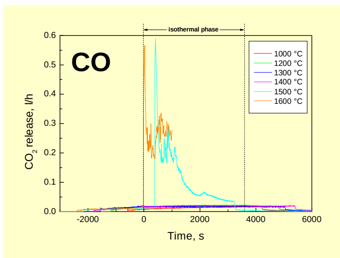


1000 °C **1200 °C** **1300 °C**



1400 °C **1500 °C** **1600 °C**

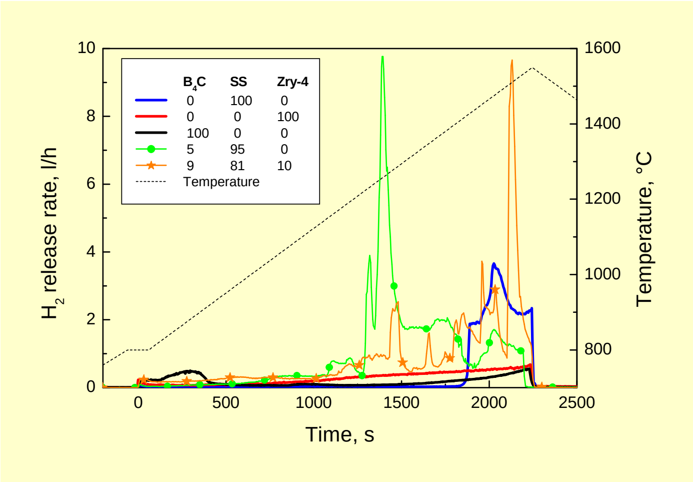




NO Methane!

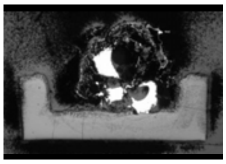
Forschungszentrum Karlsruhe in der Helmholtz-Gemeinschaft
Universität Karlsruhe (TH) Research University · founded 1825

Oxidation of B_4C-SS-Zry eutectic melts



5 B_4C + 95 SS

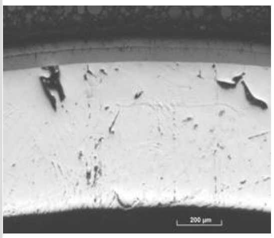
Eutectic melt

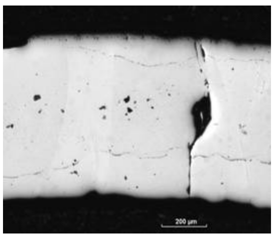


After oxidation

➧ Significant increase of oxidation rate above melting temperature

Steam starvation before reflood

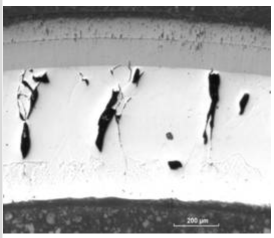


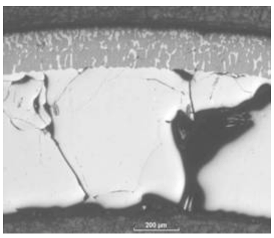


Dissolution of oxide scale

Oxidation

Steam starvation at 1700 K





Thinning of oxide scale and precipitation of α-Zr(O) in oxide

➧ Weakening of protective effect of ZrO_2 oxide layer

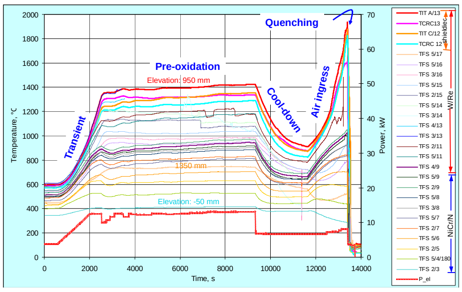
Influence of air ingress on coolability (QUENCH-10)

In comparison to steam:

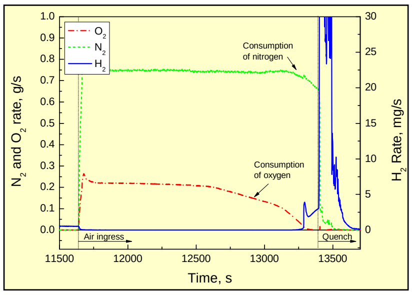
- Higher exothermal energy release
- Increased oxidation kinetics
- Less cooling effect

➡ Escalations from low temperatures

➡ Early loss of cladding barrier effect



Test phases, power input and temperatures in the QUENCH-10 bundle



Oxygen starvation and nitrogen consumption during QUENCH-10

Separate-effects tests on air oxidation

… under prototypical conditions, including

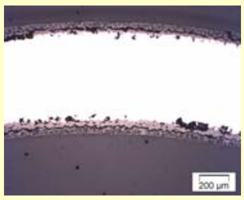
- Pre-oxidation in steam/O_2
- Tests in mixed air(N_2)-steam atmospheres

1 hour at 1200 °C in

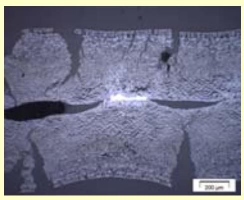
steam 50/50 steam/air air

➡ <u>Nitrogen is not an inert gas</u> under the conditions of a nuclear accident!

1 hour at 1000 °C in



steam



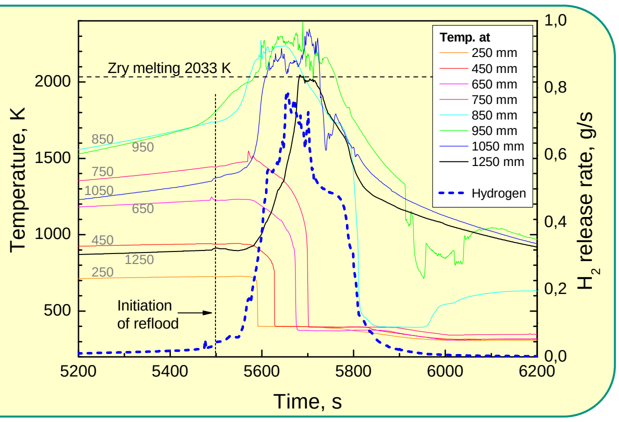
50/50 steam/N_2

Influence of reflood flow rate

➧ The three tests QUENCH-07/-08/-11 with reduced reflood flow rate <1 g/(s rod) resulted in temperature escalations and enhanced hydrogen production.

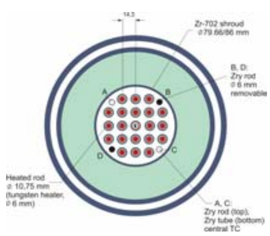
Example QUENCH-11

- SARNET benchmark
- Cooldown at bundle elevations < 800 mm
- Escalation at bundle elevations >800 mm
- Strongest hydrogen production when bundle temperatures exceeded Zry melting temperature

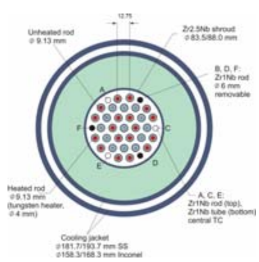


QUENCH-ACM: Influence of cladding alloy

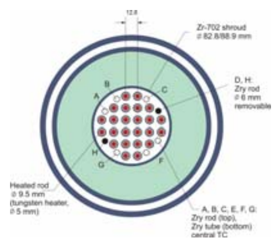
- ACM = Advanced cladding materials
- Optimised for operational conditions
- To be tested for DBA and BDBA conditions
- Bundle tests and separate-effects tests



QU-06 (Zry-4)
QU-14 (M5)

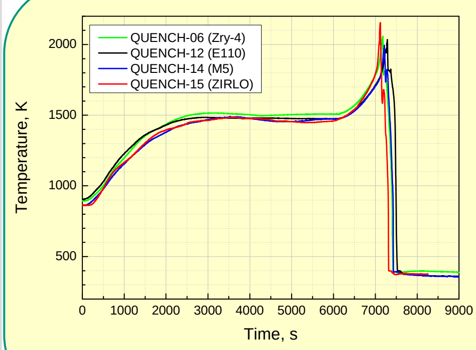


QU-12 (E110)

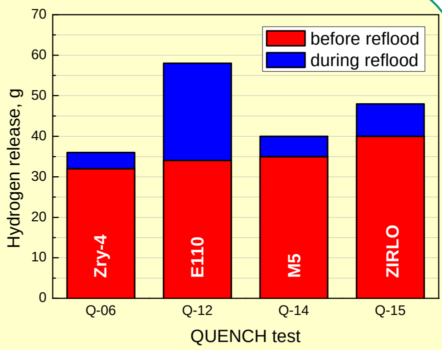


QU-15 (ZIRLO)

QUENCH-ACM: Influence of cladding alloy



Max. bundle temperatures

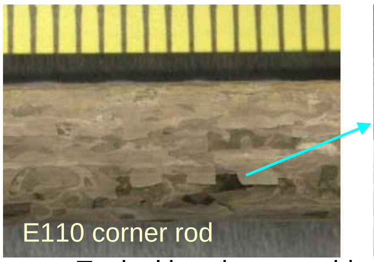


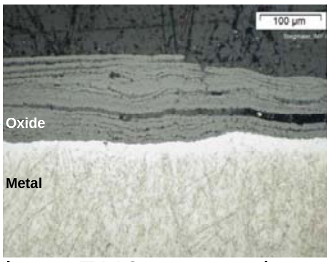
Hydrogen release

- Similar test conditions for all four experiments
- Only limited effect of cladding alloy on integral bundle behaviour
- Strongest oxidation during QUENCH-12

Post-test QUENCH-12 bundle with E110 cladding

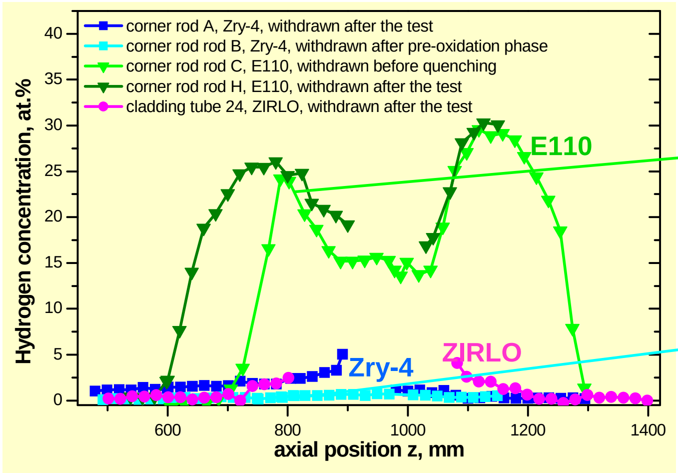
ZrO_2 debris formation and relocation to bundle foot

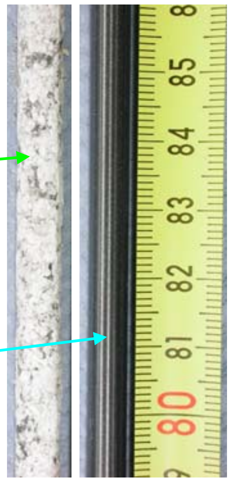




Typical breakaway oxidation at E110 corner rod

Hydrogen absorption during QUENCH-15





Strong correlation between oxide morphology and hydrogen uptake

ACM Separate-effects tests

Oxidation in steam and oxygen

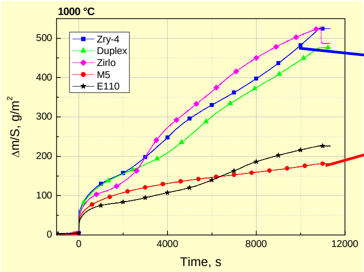
- Alloys: Zircaloy-4, Duplex, M5, ZIRLO, E110, Zr702, Hf
- Temperature: 600-1600 °C
- Isothermal and transient tests
- Test rigs: TG (online mass gain), BOX furnace
- Oxidation correlations
- Breakaway transition times and critical oxide thickness

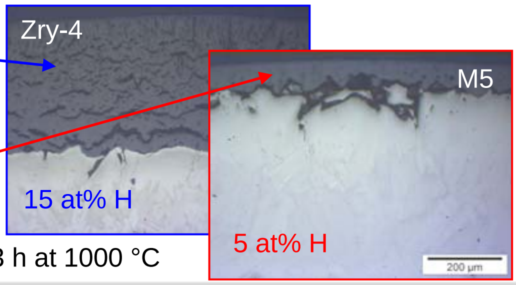
Hydrogen uptake

- In pure hydrogen (Sieverts' constants)
- During oxidation in steam
- Methods: Hot extraction and neutron radiography (post-test and in-situ)

ACM Separate-effects tests: Main results

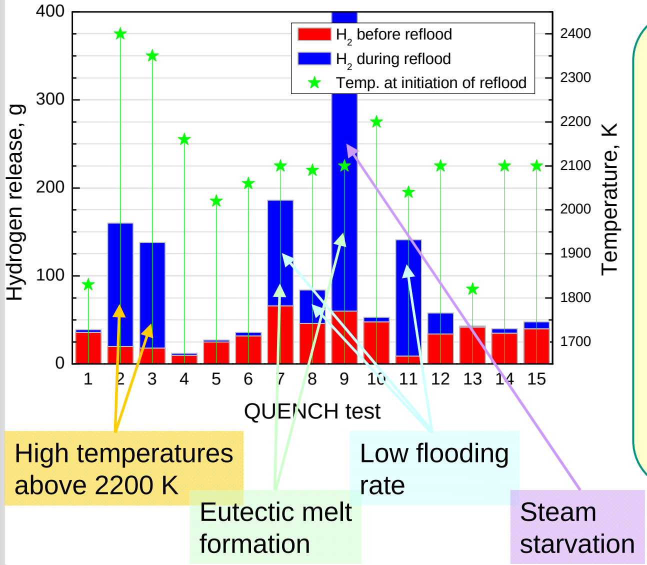
- Oxidation of Zr alloys with only slightly different composition is quite complex with strong differences up to 1000 °C, and less, but significant variation at higher temperatures.
- The oxidation kinetics are mainly determined by the oxide scale (breakaway, crystallographic phase, degree of sub-stoichiometry).
- Strong correlation between breakaway oxidation and hydrogen uptake was found.





3 h at 1000 °C

Summary



QUENCH bundle tests can be split into those:
- With successful flooding leading to immediate cooldown and low hydrogen release
- With temporary temperature escalation and strong hydrogen production

Conclusions

- Main factors affecting cooling and chemical heat production during reflood have been identified.
- The heat balance at initiation of reflood is crucial for further development of reflood process.
- Melt formation, relocation, and oxidation as well as steam starvation before reflood are most important processes for enhanced oxidation after reflood.
- Reflood rate must be > 1 g/(s rod) for successful cooldown.
- Experimental results must not directly applied to plant analyses, but they help to improve the SA code systems.