



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



KIT – The cooperation of Forschungszentrum Karlsruhe GmbH and Universität Karlsruhe (TH)

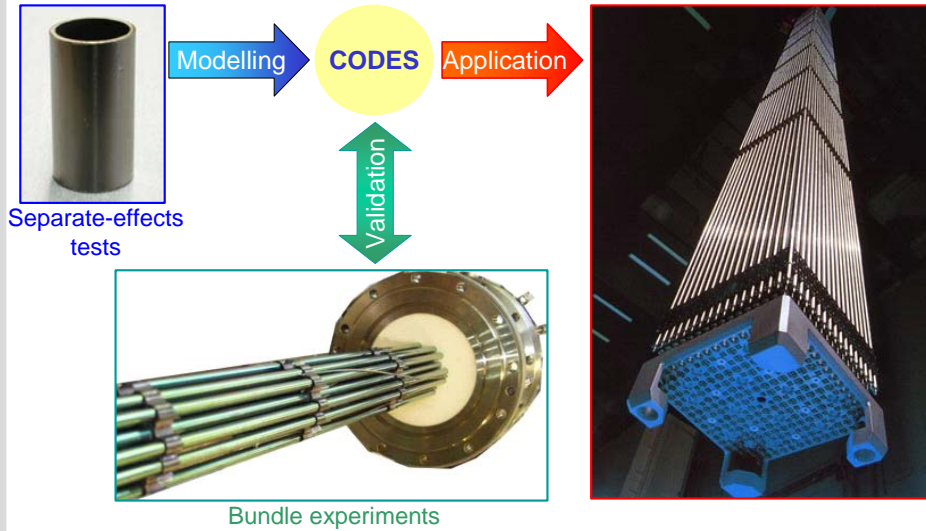


Motivation

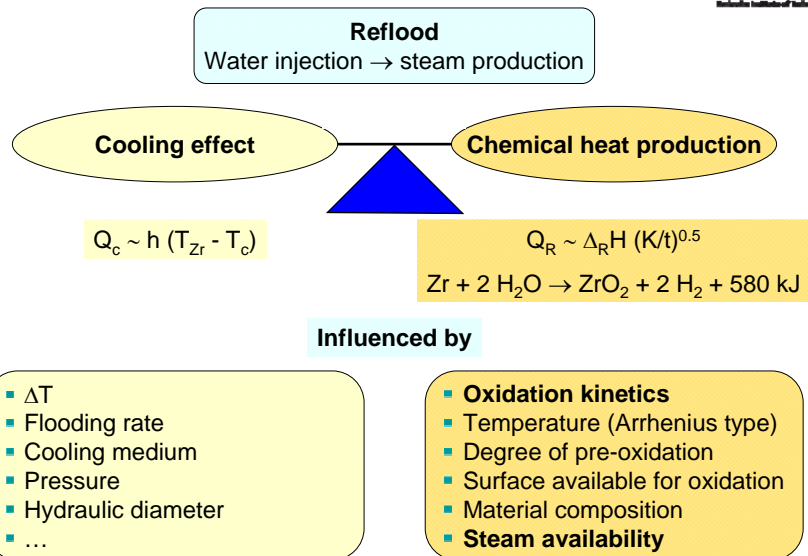


- Reflood is a prime accident management measure to terminate a nuclear accident
- Reflood may cause temperature excursion connected with increased hydrogen and FP release
 - TMI2, LOFT, PBF, CORA ...
 - .. but only qualitative hydrogen measurements
- Coolability of a degraded core is a matter of high 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



Simulation of bundle test with and without excursion



Quenching without temperature escalation

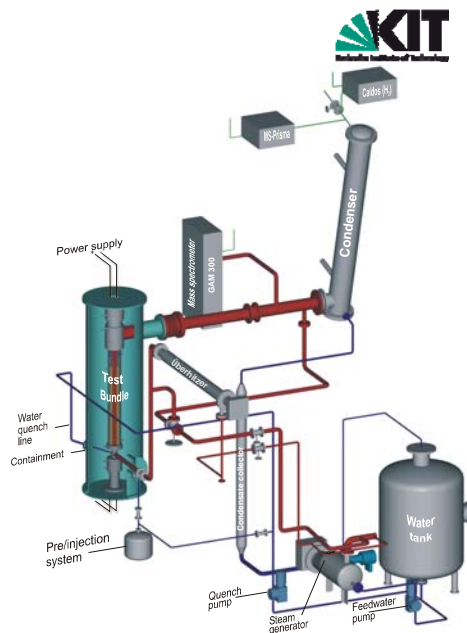
- Fast cool-down of the bundle
- Low to moderate hydrogen (FP) release
- Intact bundle

Quenching with temperature escalation

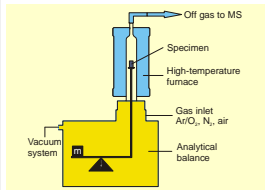
- Temperature escalation
- Strong hydrogen (FP) release
- Formation, relocation and oxidation of melts
- Strongly degraded bundle

QUENCH Facility

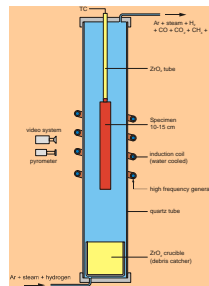
- Bundle with 21-31 fuel rod simulators of ~2,5 m length
- Electrically heated: ~1 m; max 70 kW
- Fuel simulator: ZrO_2 pellets
- Quenching (from bottom) with water or saturated steam
- Off-gas analysis by mass spectrometer (H_2 , steam ...)
- Extensive instrumentation for T, p, flow rates, level, etc.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



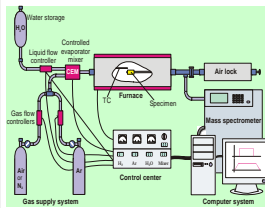
QUENCH Separate-effects tests: Main setups



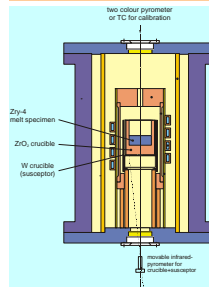
Thermo balance
 1600 °C
 1250 °C (steam)
 Specimens: 0-2 cm
 MS coupling



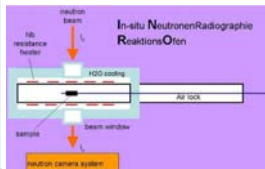
QUENCH-SR Rig
 2000 °C
 Oxidising, reducing atmosphere (incl. steam)
 Specimens: 15 cm
 MS coupling



BOX Facility
 1700 °C
 Oxidising, reducing atmosphere (incl. steam)
 Specimens: 1-2 cm
 MS coupling



LAVA Furnace
 2300 °C
 Inert, reducing atmosphere
 Specimens: 1-2 cm
 MS coupling



INRO Facility
 1500 °C
 Specimens: 1-2 cm
 Transparent for neutrons

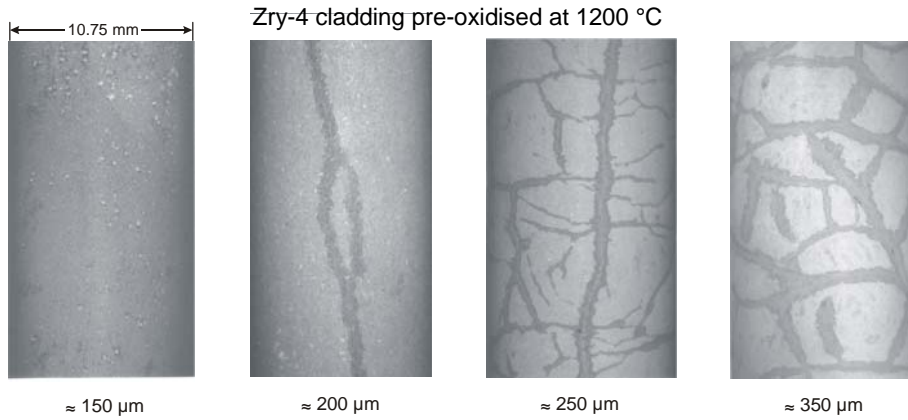
Phenomena of interest (for coolability)



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

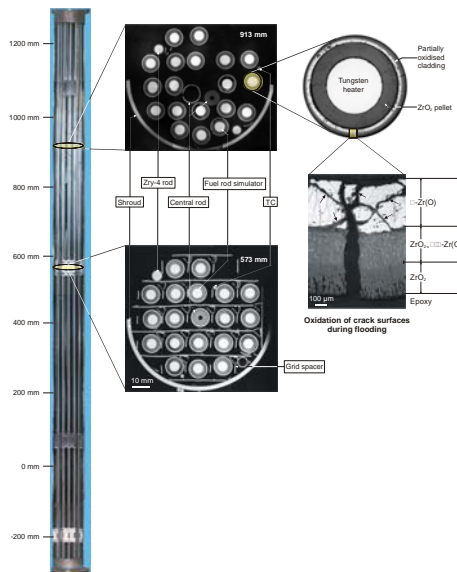


Crack formation during reflow



- Density of through-wall cracks: 0.5 mm/mm²
- 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

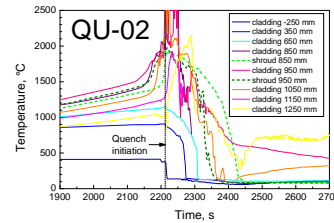
But:

- Solution enthalpy of 1 g H₂ 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 (B₄C 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 ₄ C 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 B₄C, SS, and Zry leading to rapid melt formation at about 1200 °C
- High oxidation potential of boron carbide
e.g. $B_4C + 8 H_2O \rightarrow 2 B_2O_3 + 8 H_2 + CO_2 + 792 \text{ kJ}$

Influence of B₄C absorber rod



QUENCH-07
with B₄C



**Bundle
cross
sections**

QUENCH-08
w/o B₄C

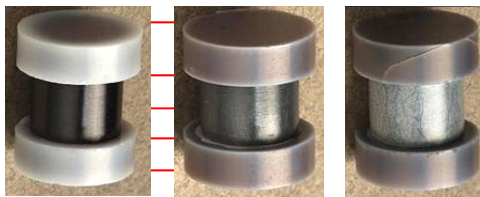


550 mm

750 mm

950 mm

Degradation of boron carbide control rods



1000 °C

1200 °C

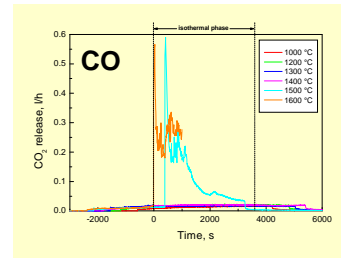
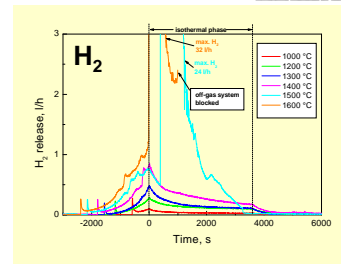
1300 °C



1400 °C

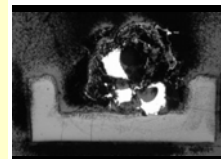
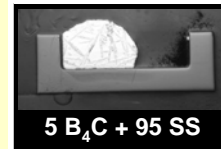
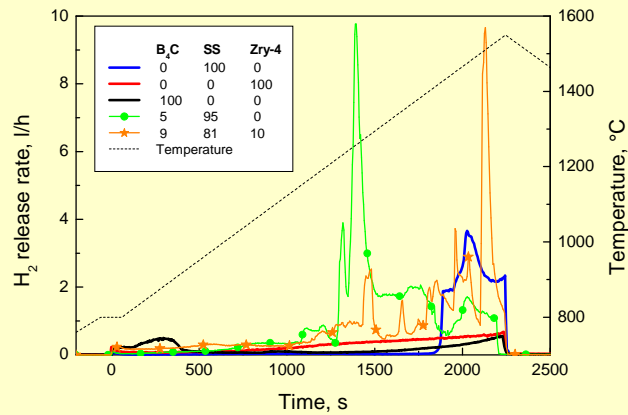
1500 °C

1600 °C



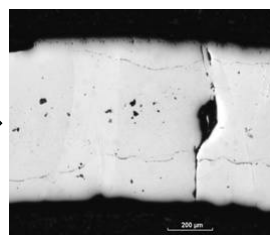
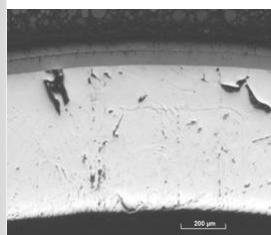
NO Methane!

Oxidation of B₄C-SS-Zry eutectic melts

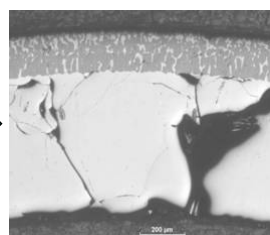
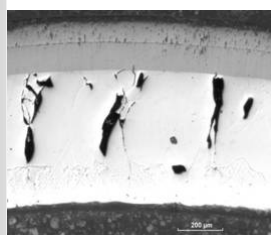


➡ Significant increase of oxidation rate above melting temperature

Steam starvation before reflow



Dissolution of
oxide scale



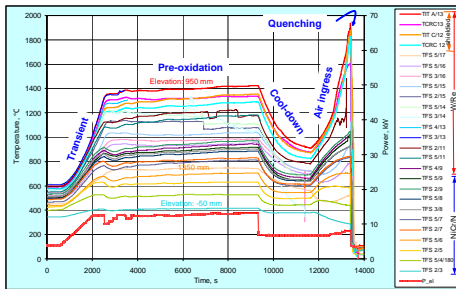
Thinning of oxide scale and
precipitation of α-Zr(O) in
oxide

➡ Weakening of
protective effect of
ZrO₂ oxide layer

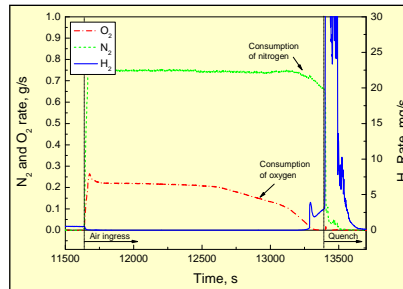
Influence of air ingress on coolability (QUENCH-10)

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/O₂
- Tests in mixed air(N₂)-steam atmospheres

1 hour at 1200 °C in



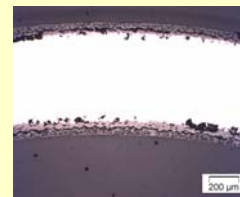
steam

50/50 steam/air

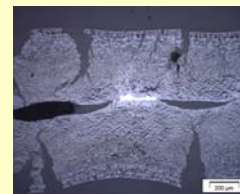
air

➤ Nitrogen is not an inert gas under the conditions of a nuclear accident!

1 hour at 1000 °C in



steam



50/50 steam/N₂

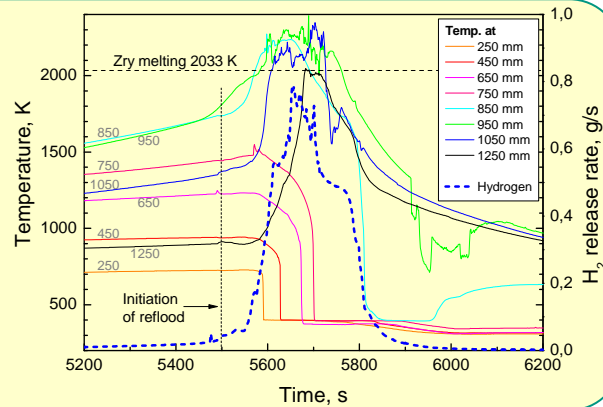
Influence of reflow flow rate



- The three tests QUENCH-07/-08/-11 with reduced reflow flow rate <math>< 1 \text{ g/(s rod)}</math> resulted in temperature escalations and enhanced hydrogen production.

Example QUENCH-11

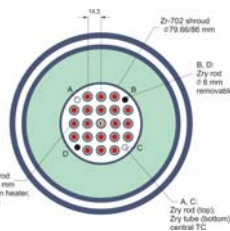
- SARNET benchmark
- Cooldown at bundle elevations <math>< 800 \text{ mm}</math>
- Escalation at bundle elevations >math>800 \text{ mm}</math>
- Strongest hydrogen production when bundle temperatures exceeded Zry melting temperature



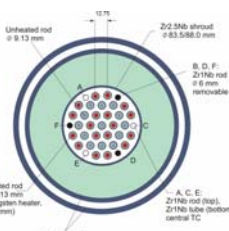
QUENCH-ACM: Influence of cladding alloy



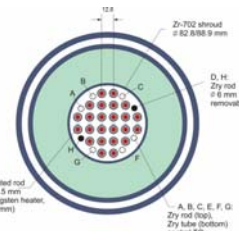
- ACM = Advanced cladding materials
- Optimised for operational conditions
- To be tested for DBA and BDBA conditions
- Bundle tests and separate-effects tests



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

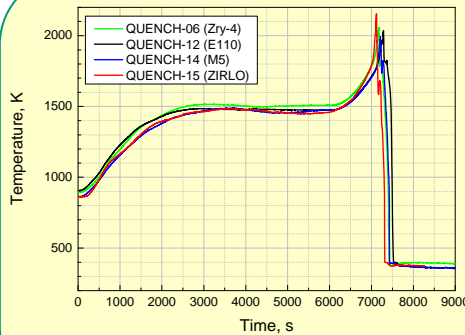


QU-12 (E110)

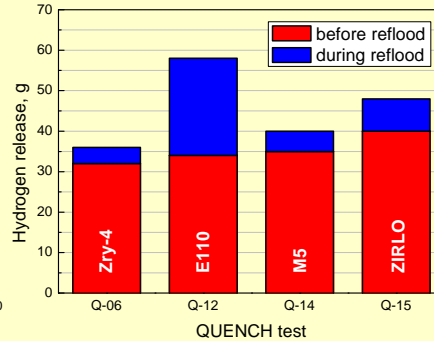


QU-15 (ZIRLO)

QUENCH-ACM: Influence of cladding alloy



Max. bundle temperatures



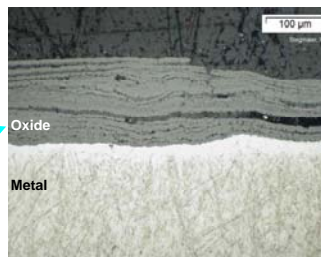
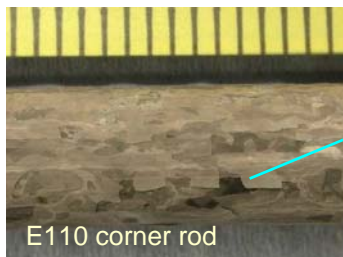
Hydrogen release

- ➔ 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

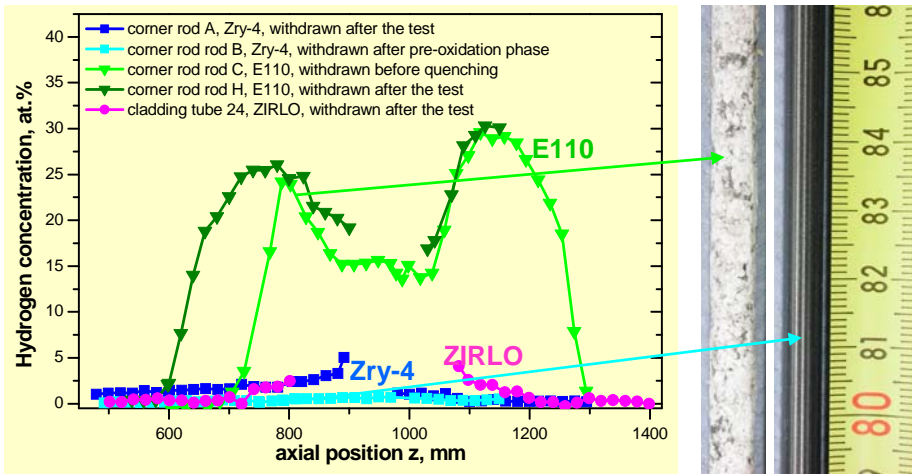


ZrO₂ debris formation and relocation to bundle foot



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

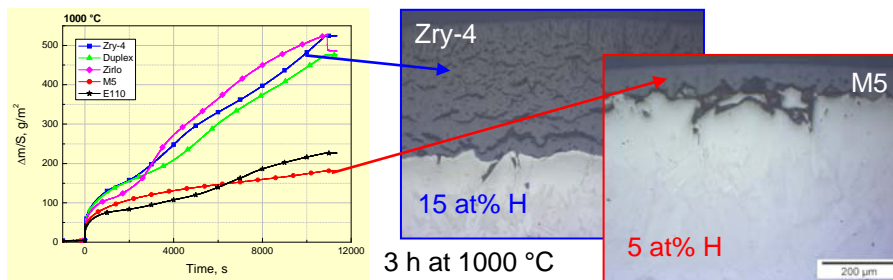
- In pure hydrogen (Sieverts' constants)
- During oxidation in steam
- Methods: Hot extraction and neutron radiography (post-test and in-situ)



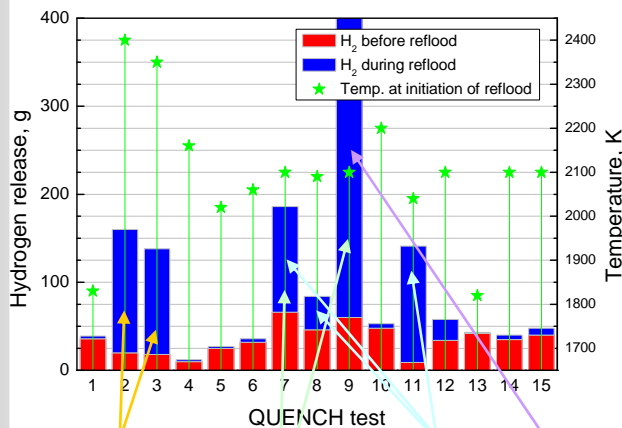
ACM Separate-effects tests: Main results



- 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.



Summary



High temperatures above 2200 K

Eutectic melt formation

Low flooding rate

Steam starvation

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 $> 1 \text{ g/(s rod)}$ for successful cooldown.
- Experimental results must not directly applied to plant analyses, but they help to improve the SA code systems.