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Results of cladding hydrogenation in tube furnace at temperatures between 500 and 1100 °C

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The past and current regulatory basis for LOCA embrittlement criteria (maximum cladding oxidation of 17 % and maximum temperature of 1200 °C) is to preserve the ductility of the rod cladding. While still preserving these criteria, recent LOCA experiments have raised questions, particularly on the behaviour of the ballooned rod region. It seems that upon clad rupture, the so-called secondary hydriding of the cladding has a major effect on the residual strength of the cladding material. Presently, it is believed that the existing embrittlement limit of 17 % is still valid for unirradeated cladding *with no hydrogen*. In a new criterion (which is not yet defined) the rod cladding's content of hydrogen, which is a measure of burn-up, has to be taken into account. Recent investigations performed at ANL showed that the cladding hydrogen content can reach *4000 wppm* due to secondary hydriding.

In the context of searching for updated LOCA embrittlement criteria, a new test series in the QUENCH facility is being launched within the Nuclear Safety Program of the Karlsruhe Institute of technology (KIT). The tests will be performed with test rod bundles to study LWR fuel rod behaviour under LOCA conditions, particularly to understand hydriding effects in the ballooned region as well as further aspects of post-quench mechanical behaviour and post-quench mechanical properties. The post-test determination of the residual strength and ductility of tested claddings will be performed with tensile and ring-compression tests. For development and enhancement of mechanical test method the single rod tests on hydrogenation of different cladding materials were performed at KIT.

New LORA facility with 3-zones tube furnace with gas channel length of 600 mm was used for hydrogenation of cladding tubes. The length of cladding probes was 150 – 200 mm. The hydrogenation was performed at temperatures 500 – 900 °C in hydrogen – argon gas mixture at hydrogen partial pressures 37, 90 and 150 mbar. 55 hydrogenated samples of the Zry-4 alloy, 18 samples of the M5® alloy and 9 samples of the E110 alloy were prepared for tension tests. Hydrogen content was measured by weighing of probes and was between *500* and *6000 wppm*. Some samples hydrogenated at temperatures lower 800 °C were bent due to not homogeneous axial hydrogen uptake and phase transition in Zr-alloy during hydrogenation. These samples were therefore prepared only for ring compression tests.

The hydrogen content for mostly samples reached not the solubility limit at a given conditions and have a radial hydrogen gradient across the cladding, what is preferable to preparation of prototypical LOCA samples with short-term secondary hydrogen uptake. Estimated hydrogen diffusion coefficients in metal for cases of hydrogen partial pressures of 37 and 90 mbar were less of known for partial pressure of 1000 mbar (by a factor of 10 in comparison with data from Steinbrück, 2004) due to significant role of transport mechanisms in gas phase.

Prepared samples will be used for further enhancement of tension and ring compression test methods in framework of the QUENCH-LOCA program.

Results of cladding hydrogenation in tube furnace at temperatures between 500 and 1100 °C

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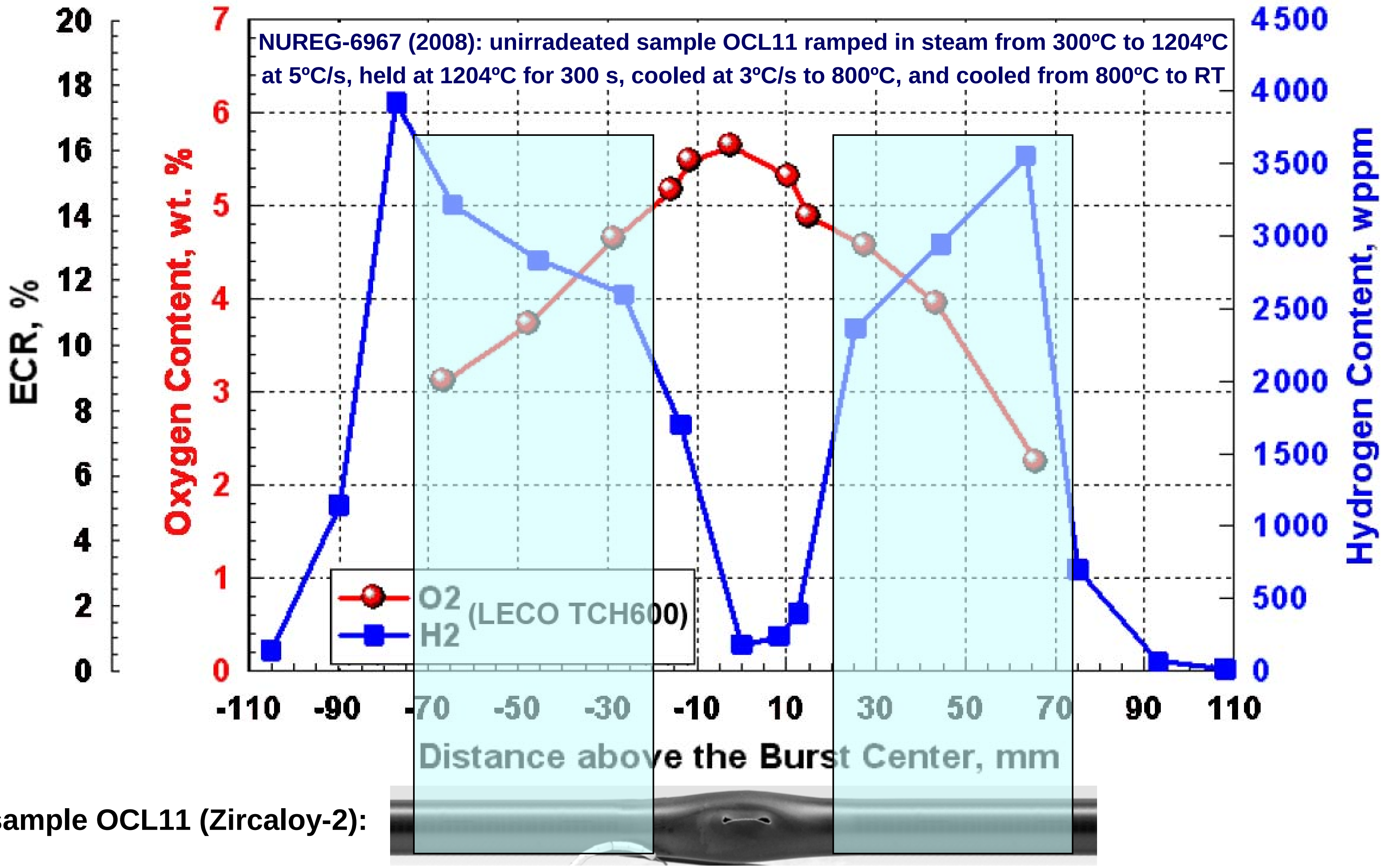
Objectives

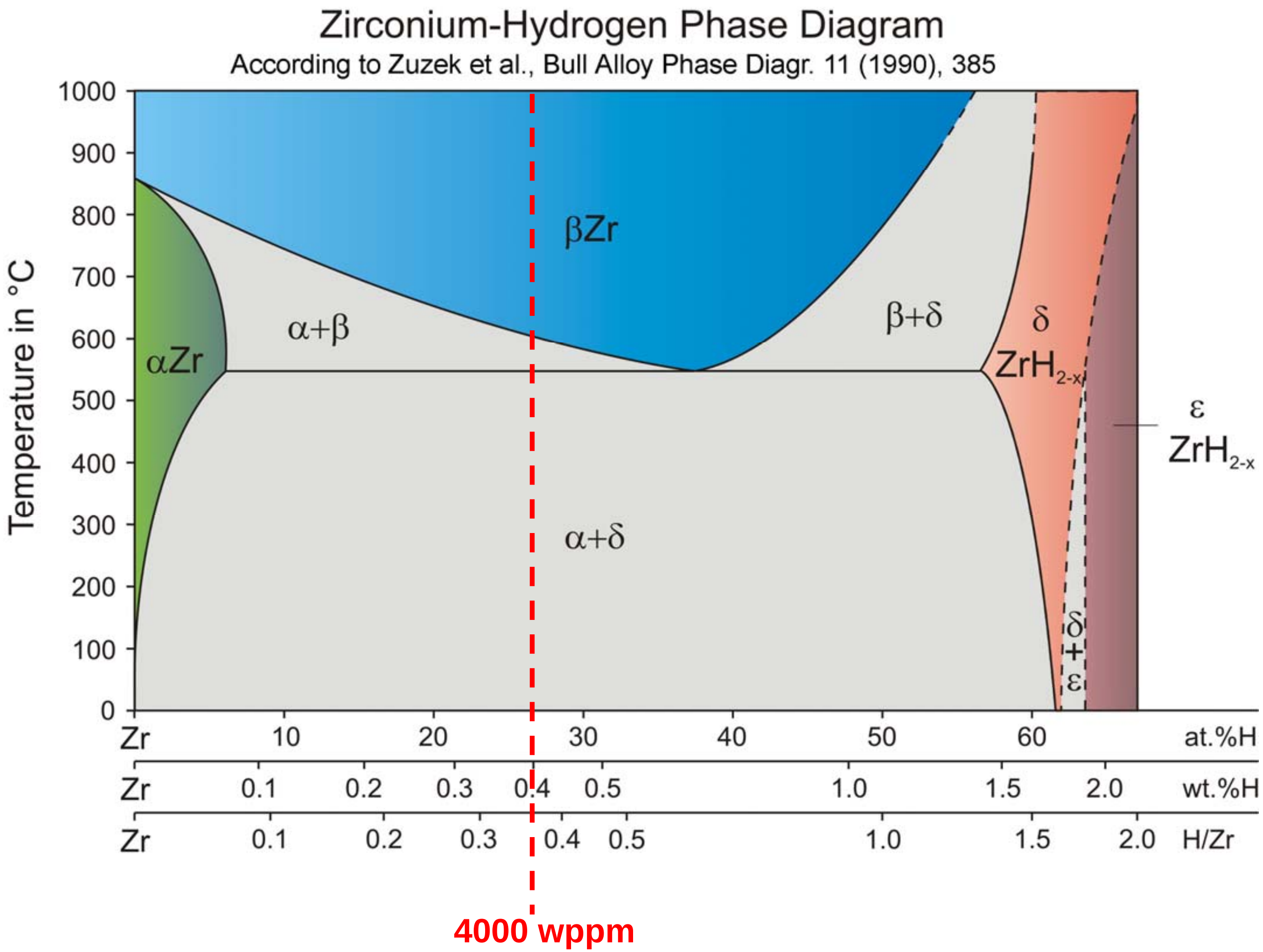
- Preparation of hydrogenated probes for mechanical tests
- Short term hydrogen uptake to investigation of samples with radial gradient of hydrogen content
- Evaluation of oxygen and hydrogen absorption by claddings produced from different zirconium alloys

Short term secondary hydrogenation after ballooning and burst: hydrogen uptake increased rapidly up to 4000 ppm (significant higher than ductility limit of 500 ppm)



sample OCL11 (Zircaloy-2):

Phase composition of system Zr-H at different temperatures



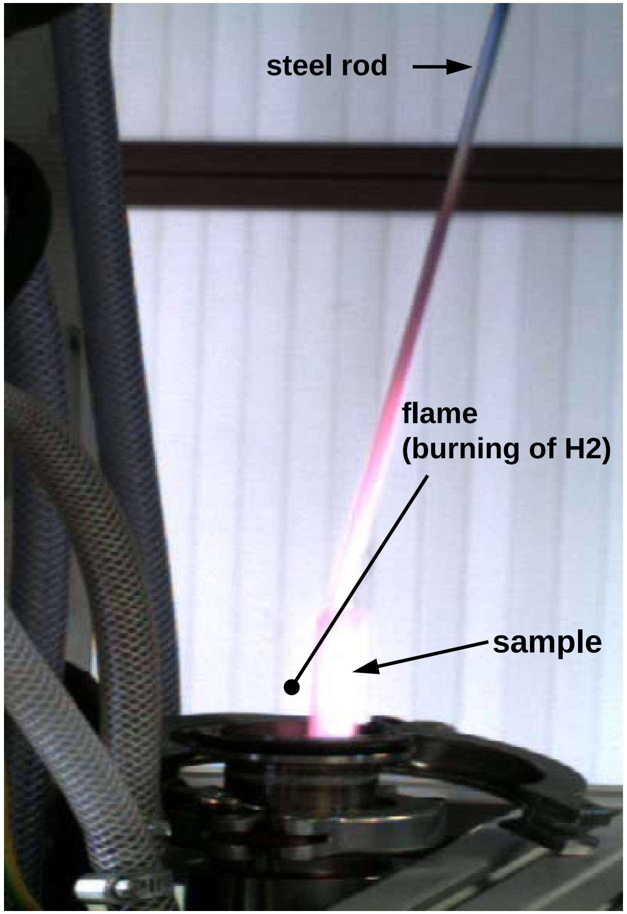
Hydrogenated standard samples for mechanical tests

Tensile tests to investigation of mechanical properties

Comparative ring compression tests

Hydrogenation facility

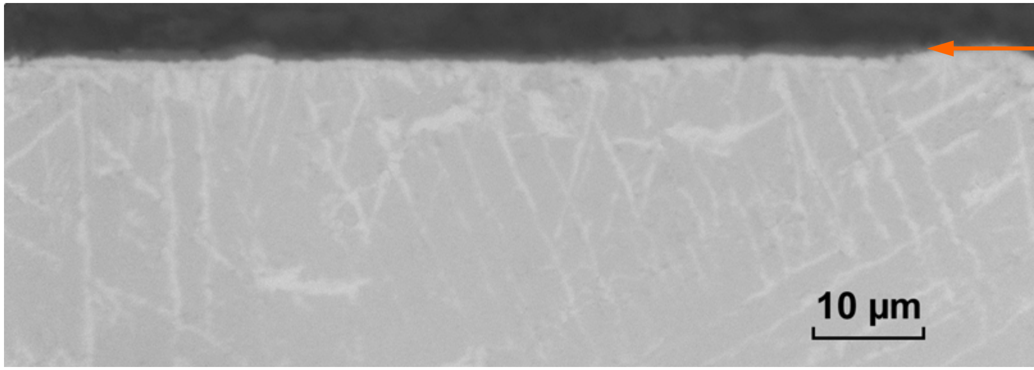
vertical 3-zones tube furnace LORA
(height 60 cm)



sample extraction

sample
15 – 20 cm

Influence of sample extraction from furnace into air atmosphere

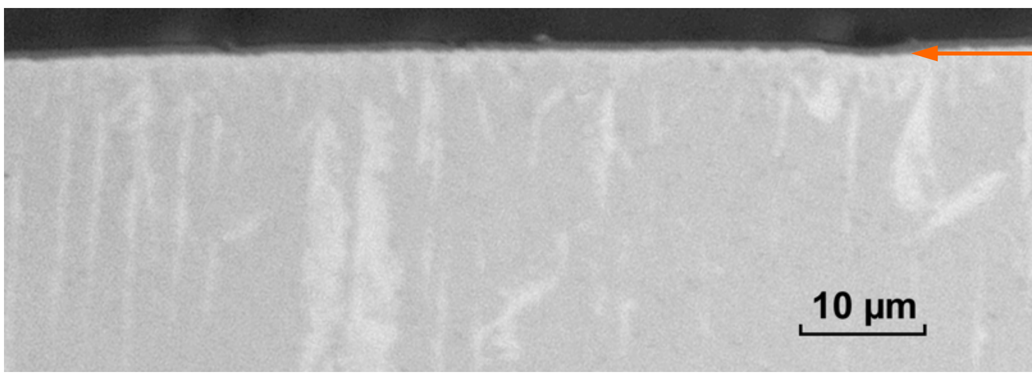


sample H3Z4 **annealed at 900 °C during 480 s** with H_2 of 90 mbar

oxide layer: 1.3 µm
/protection against H2 release during cool down/

total sample mass increase: 4600 wppm,

mass increase due to oxidation on air 900: wppm



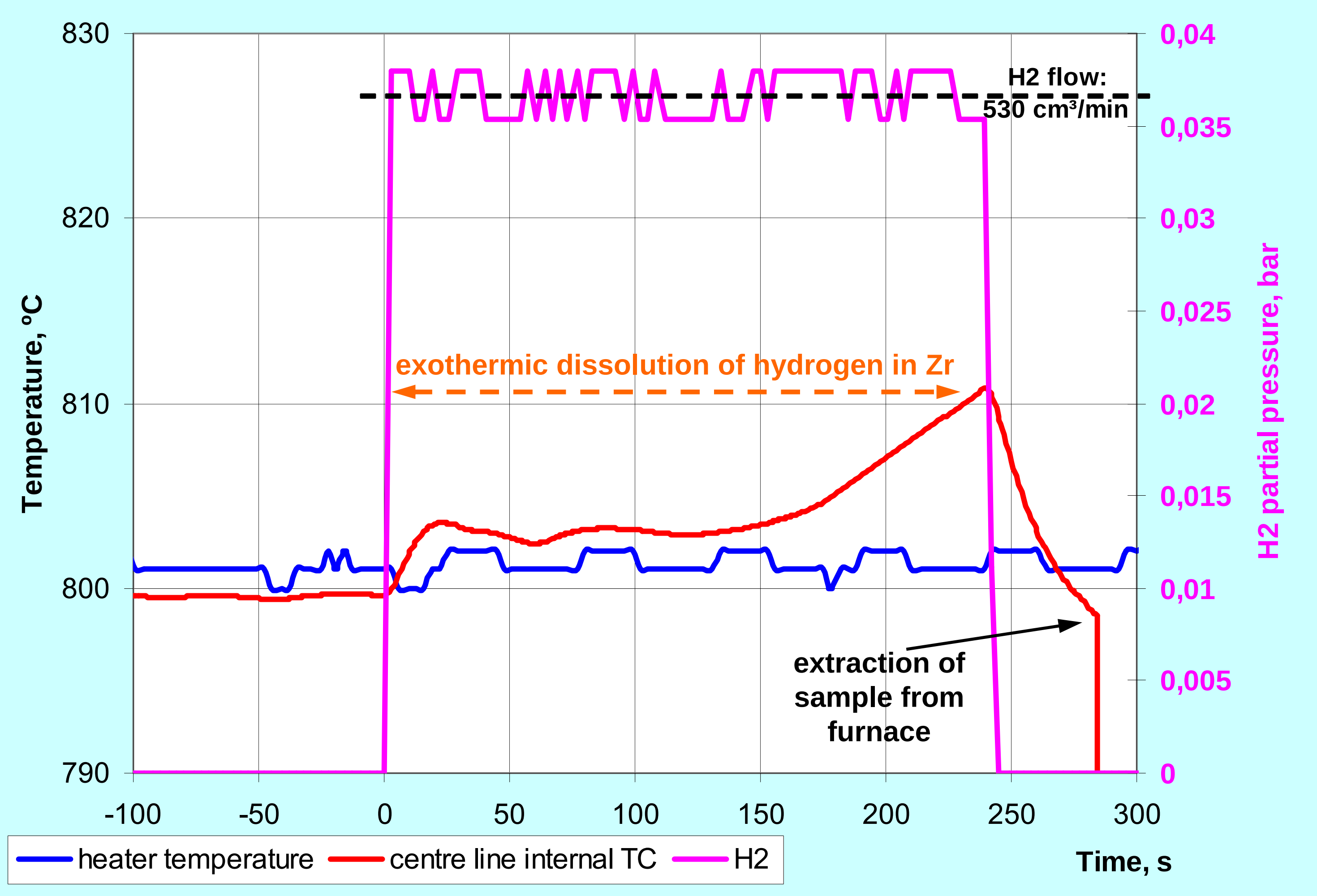
sample H9M5 **annealed at 900 °C during 240 s** with H_2 of 90 mbar

oxide layer: 0.8 µm
/protection against H2 release during cool down/

total sample mass increase: 3700 wppm,

mass increase due to oxidation on air 700: wppm

General test conduction



Test with closed probe (hydrogenation only from outside) and centre line thermocouple

Samples:

open (double sided hydrogenation) and closed

Cladding materials:

Zircaloy-4 (55 samples), **M5** (18 samples), **E110** (9 samples)

H2 partial pressure (mbar):

37 (530 cm³/min), **90** (1370 cm³/min), **150** (2500 cm³/min)

Temperatures (°C):

500, 600, 700, 800, 900, 1100

Durations:
mostly: **50 s – 1000 s;**
few: **90 min – 180 min**

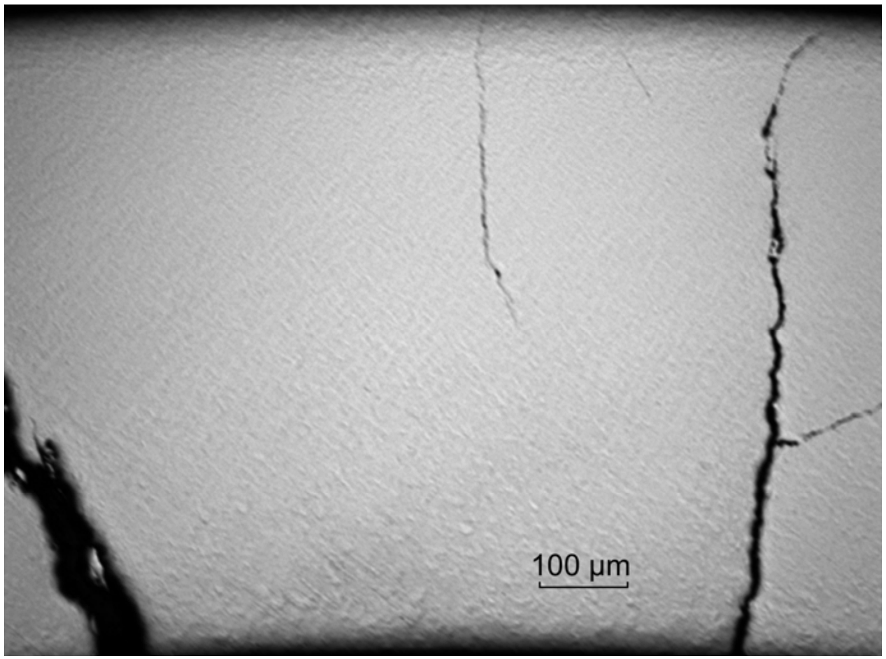
Long term hydrogenation at 500 °C: self-destroying of samples

sample H18M5
annealed
with H2 of 65 mbar
during 90 min

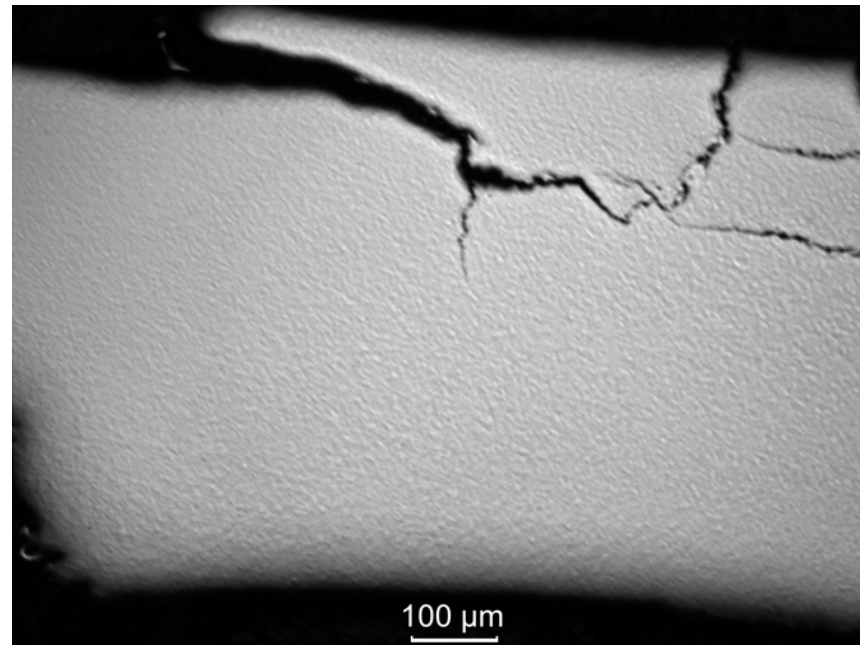
sample H6Z4
annealed
with H2 of 90 mbar
during 120 min

sample H5Z4
annealed
with H2 of 150 mbar
during 180 min

Long term hydrogenation at 500 °C : homogeneous coarse microstructure



sample H18M5
annealed
with H2 of 65 mbar
during 90 min;
as polished



sample H5Z4
annealed
with H2 of 150 mbar
during 180 min;
as polished

Bending of Zry-4 samples due to axially distributed phase transition from α-Zr to β-Zr at T<900°C

sample H22Z4
annealed at 700 °C
without H2

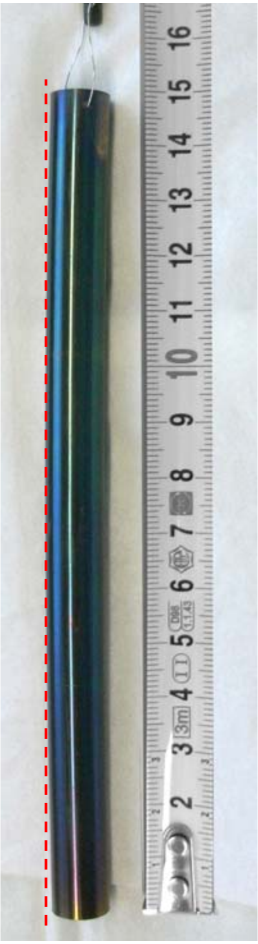
sample H36Z4
annealed at 600 °C
with H2 (37 mbar)
Δt=600 s
Δm_H=1800 wppm

sample H15Z4
annealed at 700 °C
with H2 (37 mbar)
Δt=360 s
Δm_H=2100 wppm

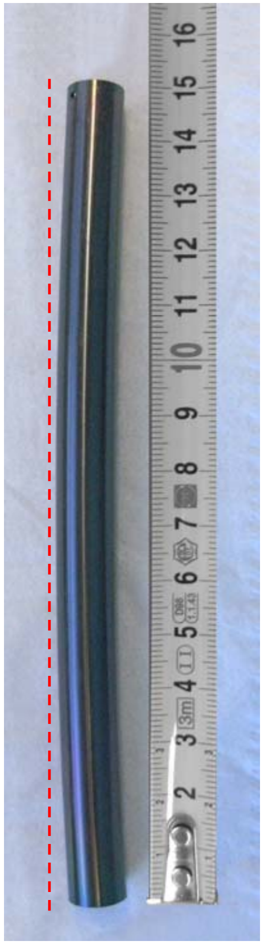
sample H18Z4
annealed at 800 °C
with H2 (37 mbar)
Δt=120 s
Δm_H=600 wppm

sample H 40Z4
annealed at 900 °C
with H2 (37 mbar)
Δt=960 s
Δm_H= 2100 wppm

Bending of the Nb-bearing samples due to phase transition from α-Zr to β-Zr at T<800°C



sample H23**M5**
annealed at 600 °C
with H2 (37 mbar)
Δt=360 s
Δm_H=1300 wppm



sample H20**M5**
annealed at 700 °C
with H2 (37 mbar)
Δt=240 s
Δm_H=1200 wppm

sample H15**M5**
annealed at 800 °C
with H2 (37 mbar)
Δt=960 s
Δm_H=4000 wppm

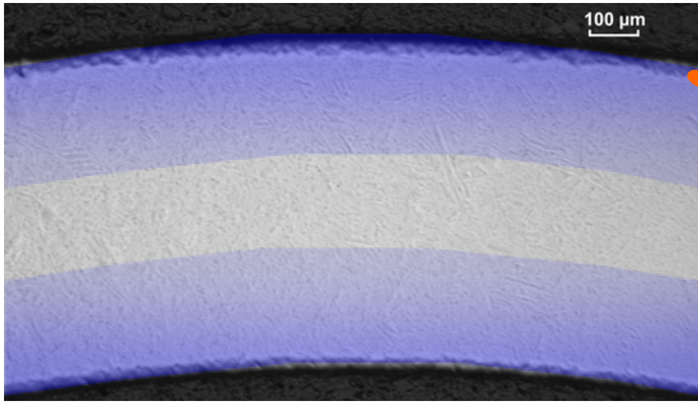
sample H9**E110**
annealed at 800 °C
with H2 (37 mbar)
Δt=480 s
Δm_H=4000 wppm

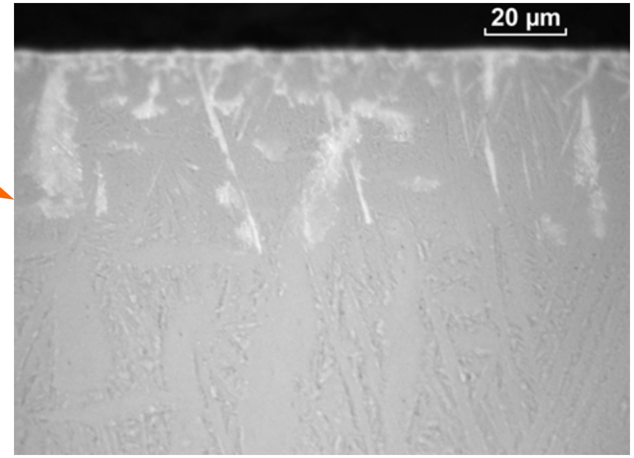
Is the phase transition boundary for M5 and E110 lower than for Zry-4?

Hydrogenation development

1) Hydrogenation of sample under different conditions

2) Influence of different H content on metallographic preparation



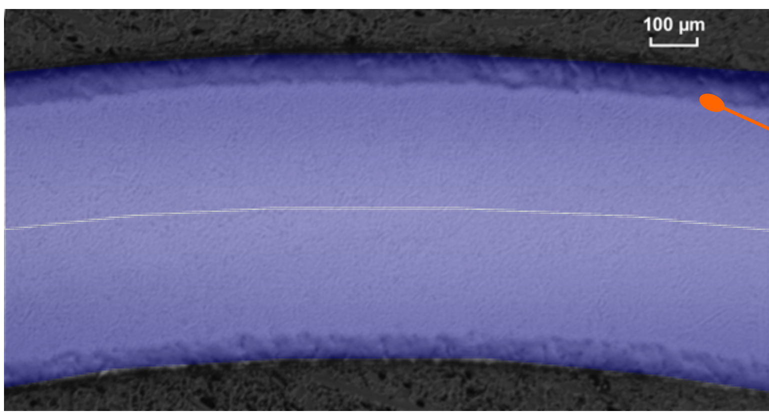


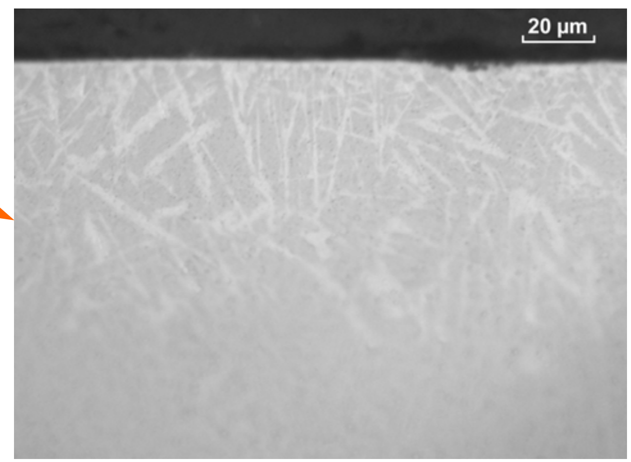
45 µm

fewer hydrogen: coarse edge of cross-section after polishing is narrow

M5 sample H7M5**:** annealed in H_2 (p. p. ***90 mbar***) at *900°C* during 240 s; *solubility: 4300 wppm;*

Δm_H = 3200 wppm (partial sample hydrogenation), effective hydrogenated layer ~250 µm





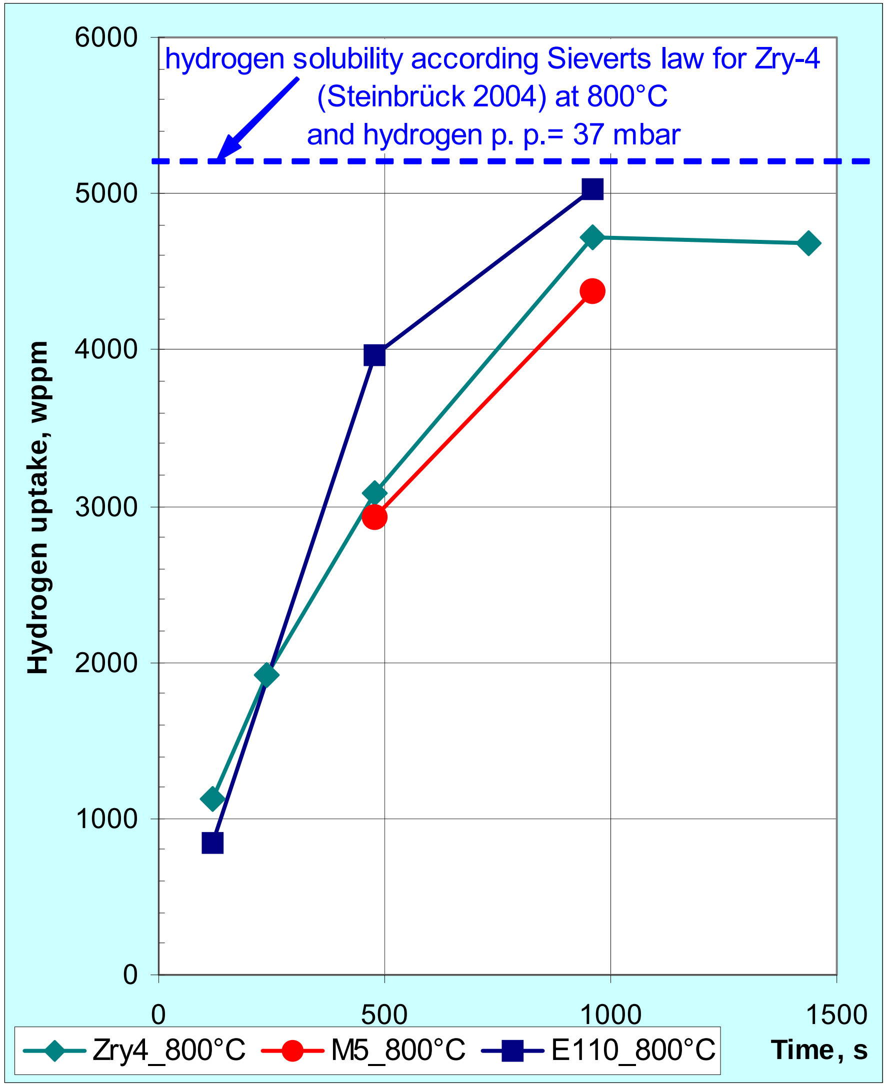
70 µm

more hydrogen: coarse edge of cross-section after polishing is wide

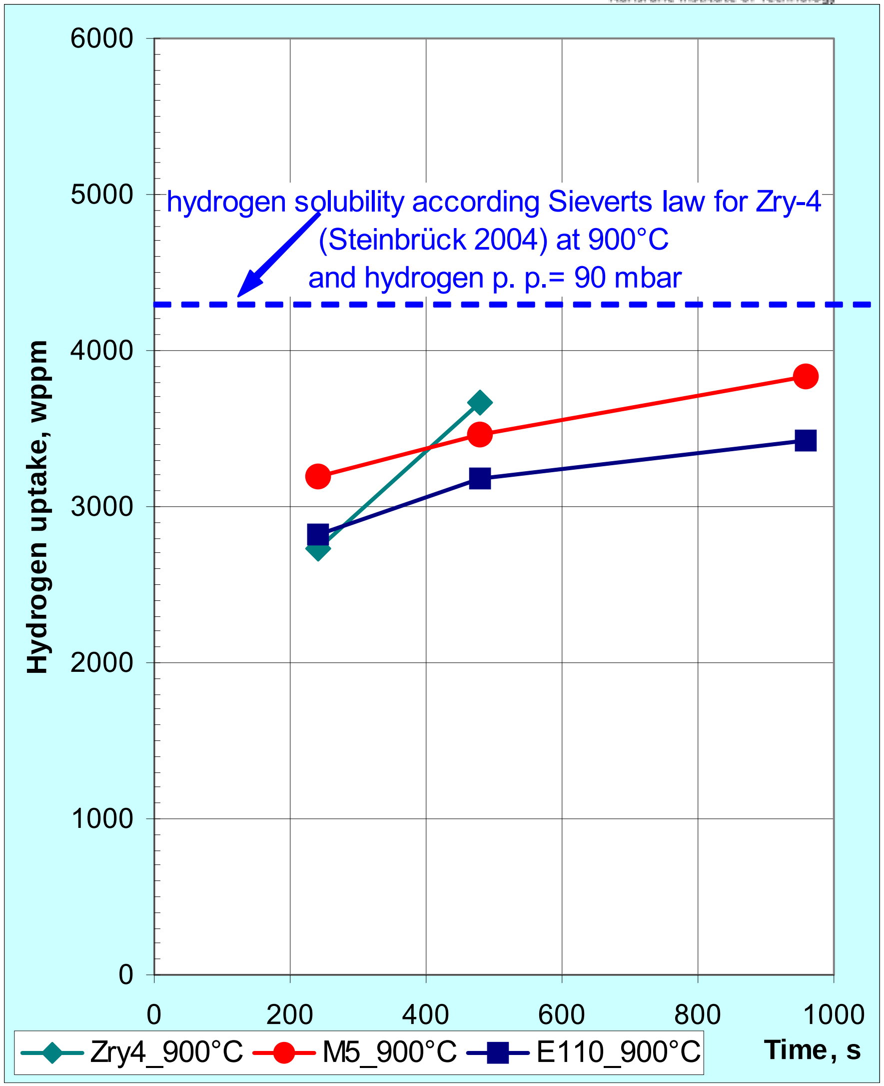
Zry-4 sample H4Z4: annealed in H_2 (p. p. ***150 mbar***) at *900°C* during 480 s; *solubility: 5600 wppm;*

Δm_H = 5500 wppm (full sample hydrogenation)

Similar hydrogenation development for different alloys

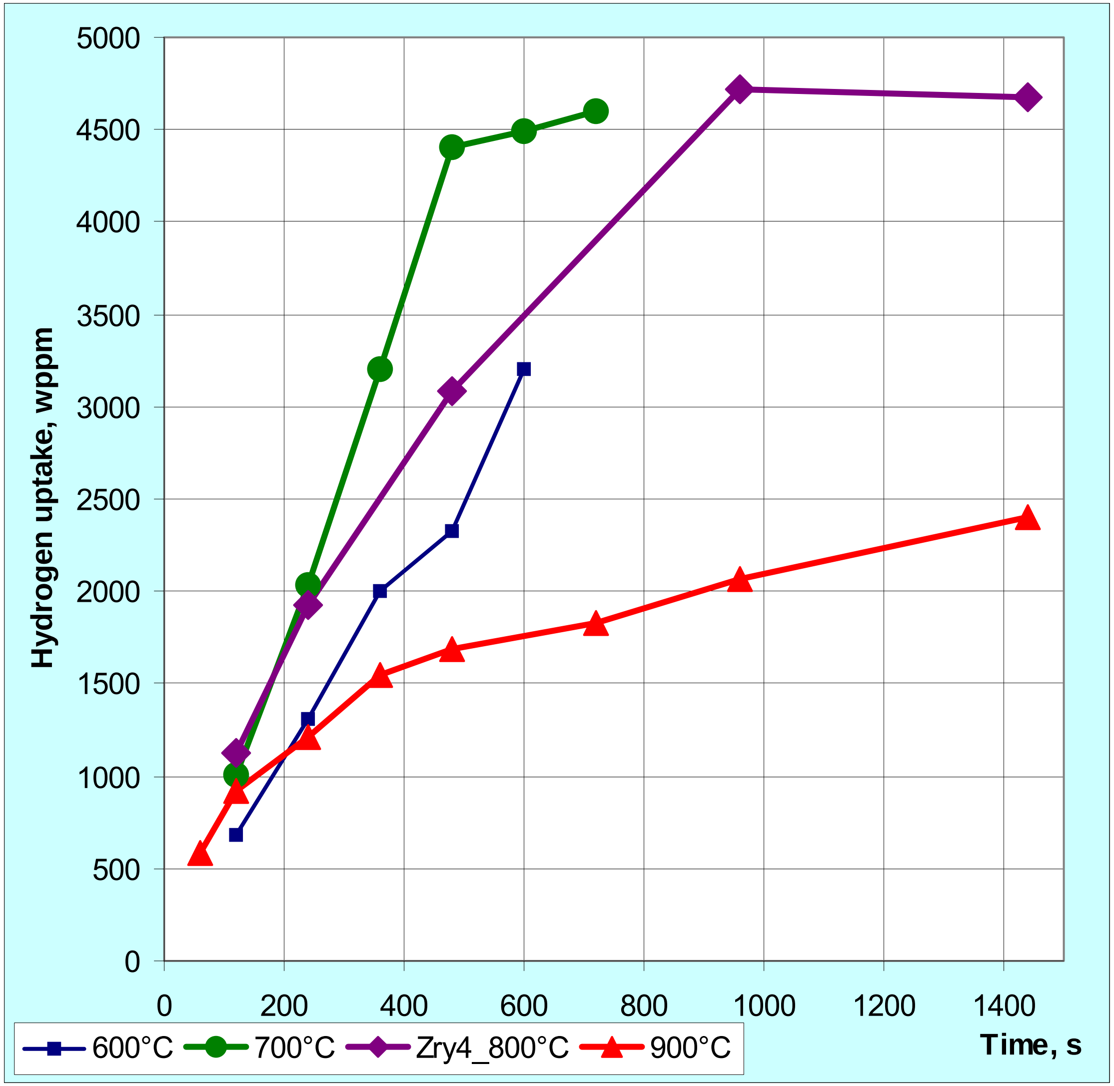


Temperature 800 °C, H_2 partial pressure 37 mbar



Temperature 900 °C, H_2 partial pressure 90 mbar

Mass gain for Zry-4 at partial pressure of 37 mbar

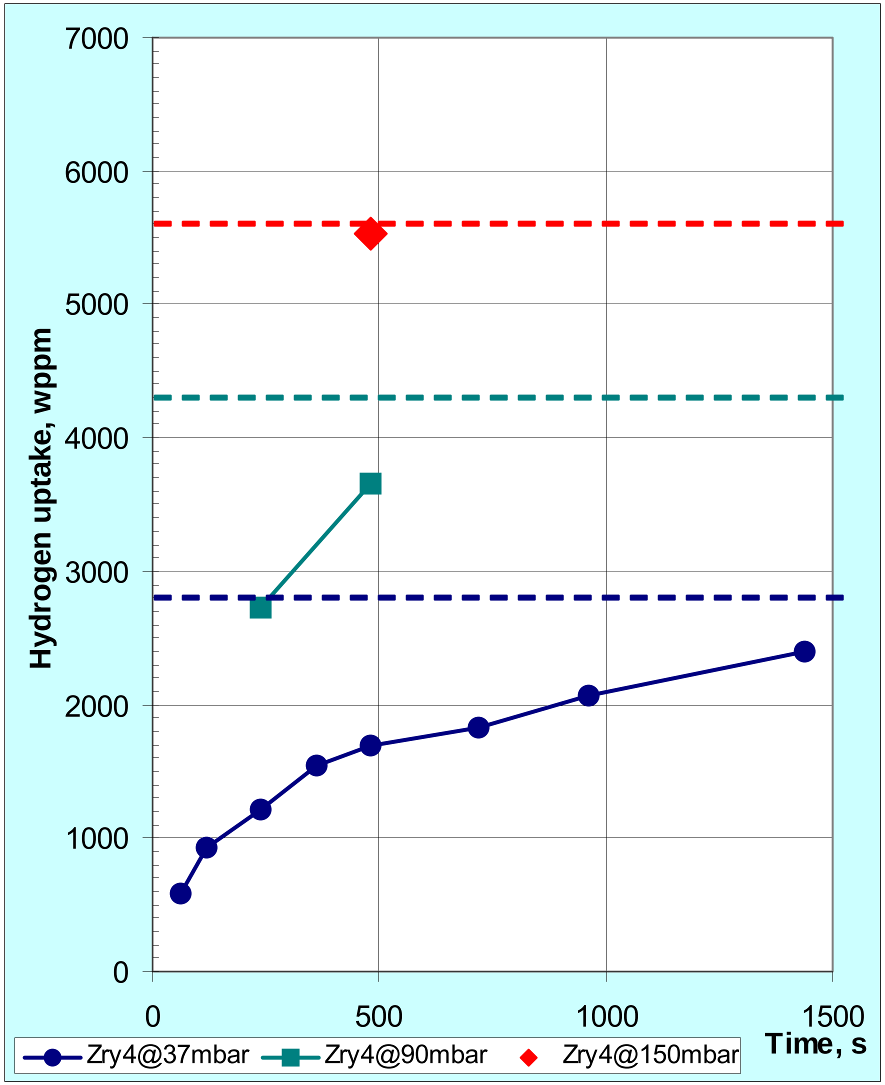


1) Saturation of processes will be not reached before 1500 s for all temperatures at this hydrogen partial pressure.

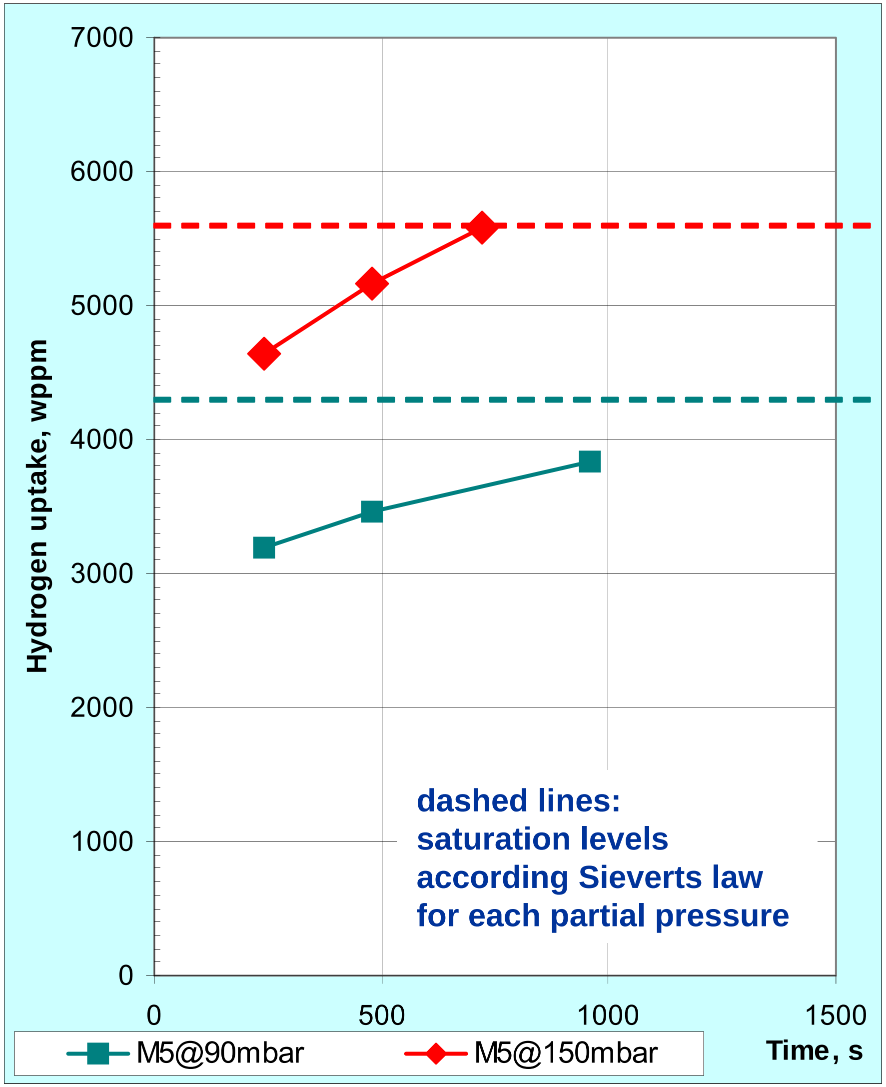
2) Competition of two processes with counterpart dependency on temperature: hydrogen diffusion and hydrogen solubility.

As result – no Arrhenius dependency (drop out of 600°C case) for mass increase before saturated final state.

Saturation achievement at 900 °C for different hydrogen partial pressures

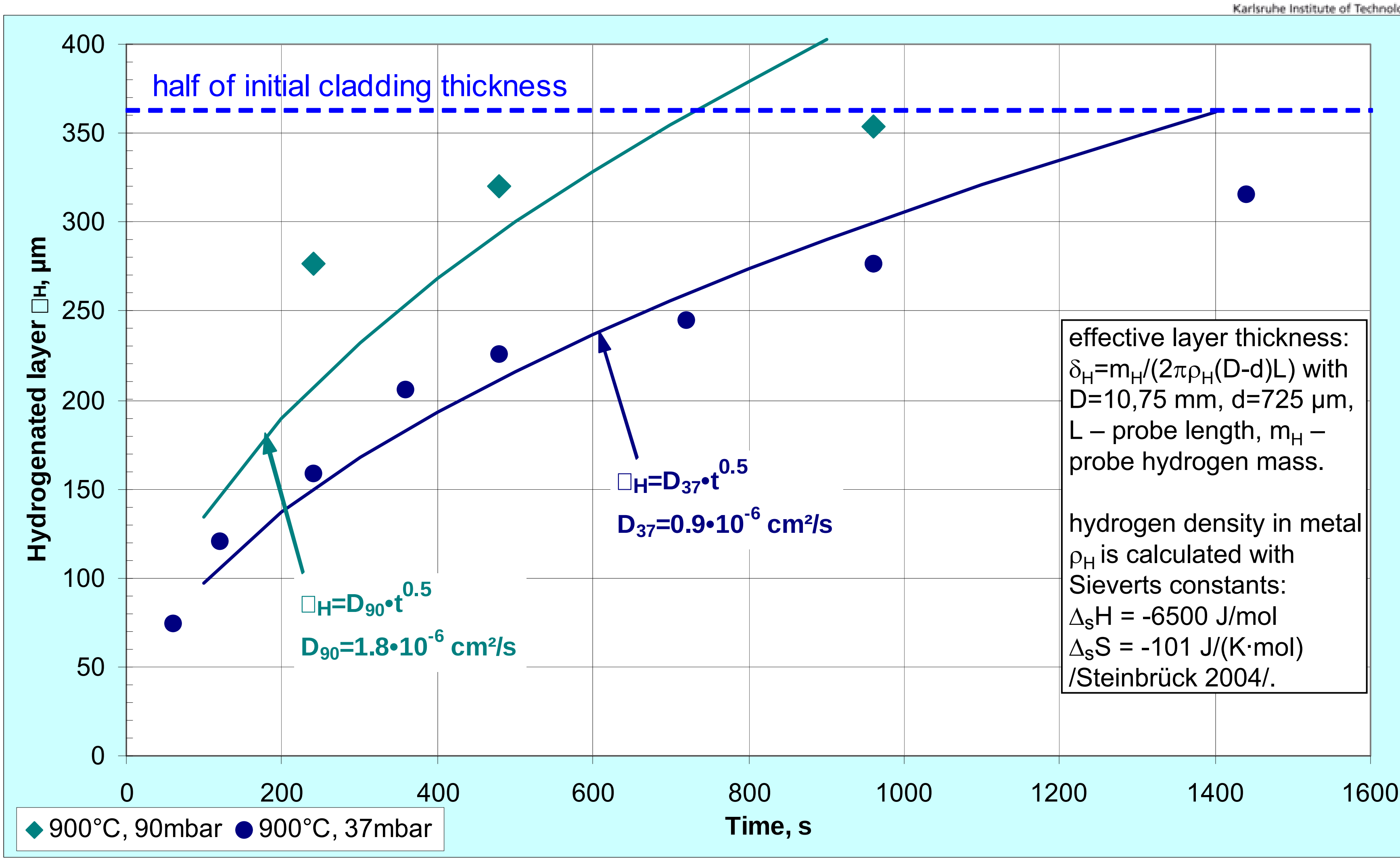


Zry-4: H_2 partial pressures 37, 90 and 150 mbar



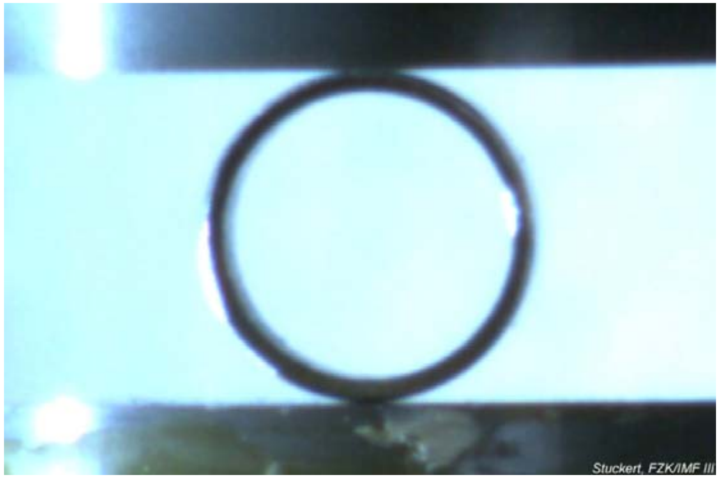
M5: H2 partial pressures 90 and 150 mbar

Estimation of development of effective hydrogenated layer (95% hydrogen inside of saturated region)



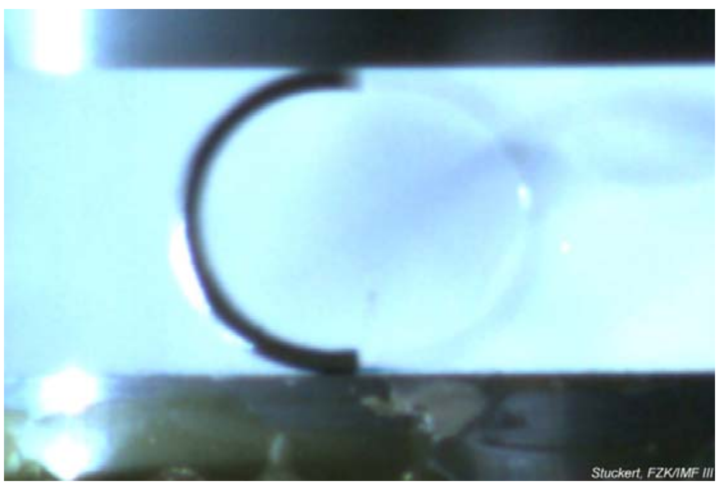
Effective "diffusion coefficients" less of known for "normal" H_2 partial pressure of 1000 mbar due to significant role of gas phase diffusion

Ring compression tests (*displacement rate 17 µm/s*): M5 alloy hydrogenated to 5000 wppm hydrogen

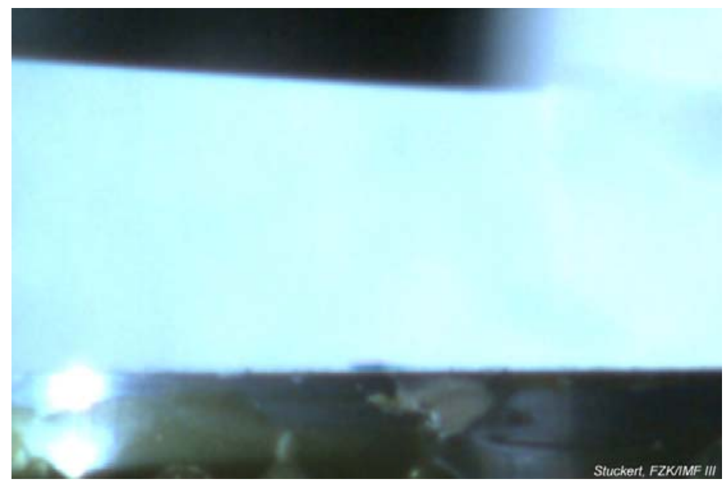


0 s: intact

43.909 s: two cracks



73.527 s: beginning of probe disintegration



73.607 s: complete disintegrated

Conclusions

➢ New LORA facility with 3-zones tube furnace was used for hydrogenation of cladding tubes to preparation of samples for mechanical tests. Hydrogenation was performed at temperatures 500 – 900 °C in hydrogen – argon gas mixture at hydrogen partial pressures 37, 90 and 150 mbar.

➢ 55 hydrogenated samples of the Zry-4 alloy, 18 samples of the M5® alloy and 9 samples of the E110 alloy were prepared for tension tests. Hydrogen content was measured by weighing and reached values between 500 and 6000 wppm (excluding reference probes with 19000 wppm). Some samples were bent during hydrogenation, therefore these samples could be used only for ring compression tests.

➢The hydrogen content for mostly samples reached not the solubility limit at a given conditions and have a radial hydrogen gradient across the cladding, what is preferable to preparation of prototypical LOCA samples with secondary hydrogen uptake.

➢ Prepared samples will be used for further enhancement of tension and ring compression test methods in framework of the QUENCH-LOCA program.

Acknowledgements:

● Mss. F. Baudin and U. Peters for probe preparation, test conduct and post-test probe processing

● Ms. U. Stegmaier for post-test probe investigation

Following activity:

• Metallographic investigations of samples microstructure

Thank you for your attention!