Results of the SPIZWURZ bundle test

J. Stuckert, M. Große, J. Moch, C. Rössger, U. Peters, T. Y. Lin, S. Weick

Abstract

The 21-rod SPIZWURZ test bundle with three types of unirradiated claddings (opt. ZIRLO, Zry-4, DX-D4) was used to conduct a long-term integral experiment, approximately simulating dry storage conditions. A variety of parameters typical of an integrated test allowed for the acquisition of a large amount of experimental data necessary for verifying the corresponding computer codes. The claddings were preliminarily hydrogenated to concentrations of 100 and 300 wppm in a specially designed HOKI tubular furnace, distributed as uniformly as possible over a 1.3 m length. Hydrogenation was carried out at 450 °C by sequentially feeding fixed masses of hydrogen through a specially treated inner surface of the claddings. It was noted that the rate of hydrogenation of the opt. ZIRLO claddings is 1.5 times lower than for the Zry-4 claddings. After the hydrogen loading of the cladding tubes, the axial distribution of hydrogen was determined by laser scanning profilometry calibrated by hot gas extraction.

During the experiment, two values of internal rod pressure were used: 106 and 146 bar, which were maintained constant throughout the experiment (250 days). The peak cladding temperature decreased in steps of ≈15 K from 400 to 165 °C (average cooling rate ≈0.9 K/day). The maximal cooling rate during each temperature step was 6 K/h, step duration was about 10 h.

The post-test laser scanner measurements of the outer cladding diameter showed significant creep: radial deformation values are between 0.2 and 3.3% (diameter increase and the corresponding wall thinning). The largest creep of 3.3% was measured for opt. ZIRLO claddings hydrogenated to 300 wppm. The corresponding maximum creep value was 0.93% for Zry-4 and 1% for DX-D4. A clearly visible dependence of the degree of creep on the hydrogen concentration is observed for the opt. ZIRLO claddings: the creep of claddings hydrogenated to 300 wppm is 1.2-1.5 times higher than that of claddings hydrogenated to 100 wppm. A number of claddings show radially asymmetric wall thinning, which can be associated with the radial shift of the pellets from the central axis of the rod and the corresponding asymmetric heat supply along the circumference of the cladding.

The metallographic investigations revealed a uniform distribution of hydrides throughout the entire cladding circumference for all three cladding types used. In the DX-D4 claddings, hydrogen primarily diffused toward the outer liner. The degree of hydride reorientation was significantly higher in the Zry-4 claddings compared to the opt. ZIRLO claddings.

The difference in the behavior of the Zry-4 and opt. ZIRLO may be due, in part, to their different grain microstructures. Opt. ZIRLO claddings have a finer grain size than Zry-4. Moreover, although the temperatures during hydrogenation (450 °C) and the experiment itself (max. 400 °C) were relatively low, EBSD measurements showed grain growth from approx. 6 µm for the initial state to post-test 16 µm for Zry-4 (12 µm after hydrogenation), and from initial 3 µm to post-test 4.3 µm for opt. ZIRLO.

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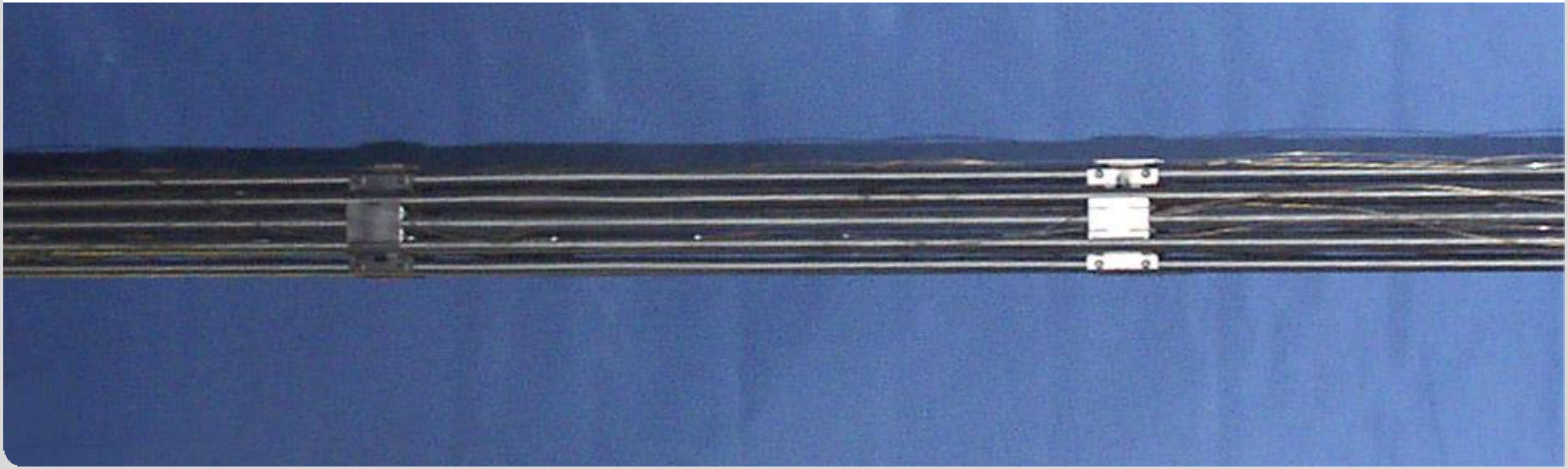
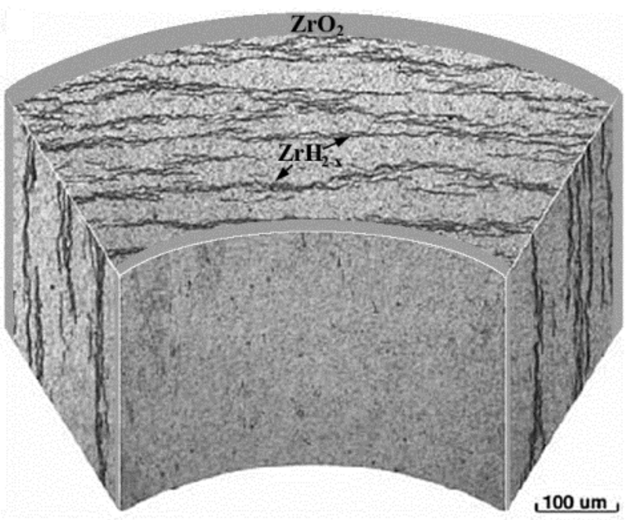


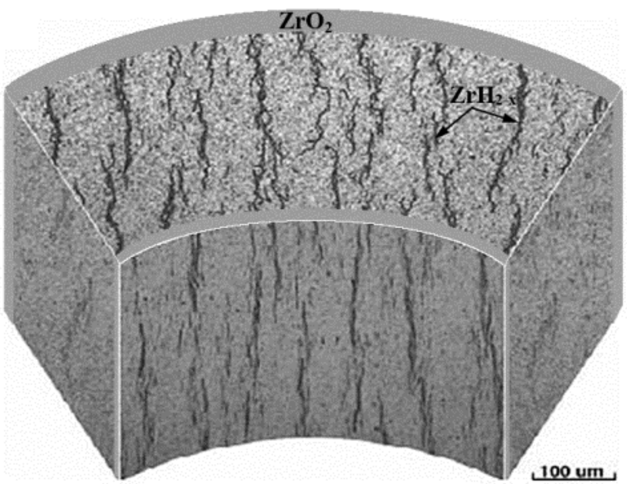
Goal of <u>bundle</u> test:

investigation of behavior of hydrides during the long-time dry storage of spent fuel with

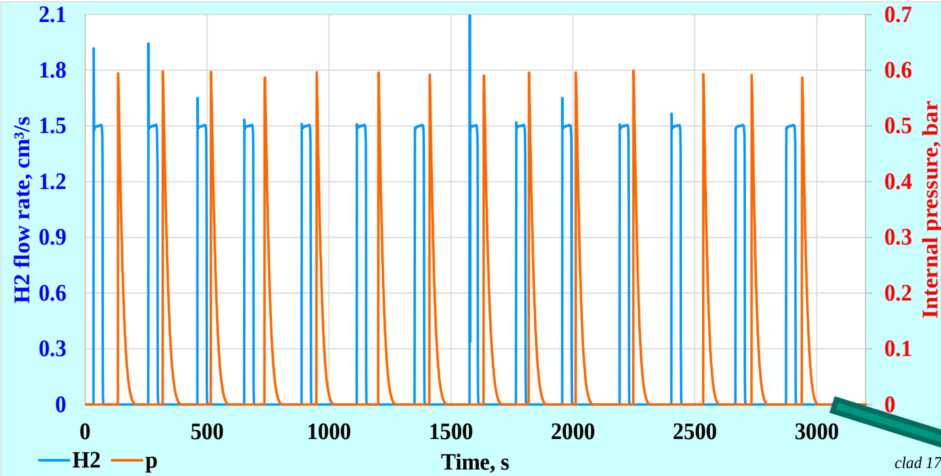
- ***different cladding materials (Zry-4, opt. ZIRLO, DUPLEX)***
- ***different cladding inner pressures (140, 100 bar)***
- ***different hydrogen contents (100, 300 wppm)***
- ***different temperature histories (due to axial T profile in the bundle)***

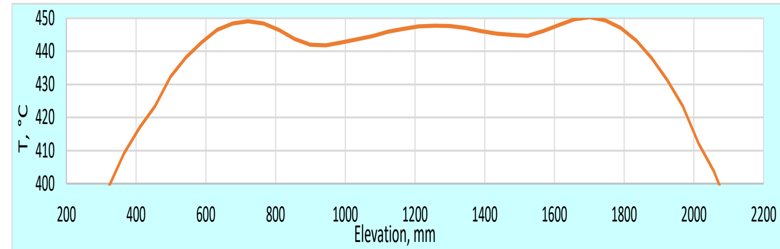


long time cool-down: reorientation of hydrides



Hydrogenation of each cladding in extended pulse mode to prevent rapid formation of hydrides near to the inner cladding surface





pulse filling of the internal cladding volume after special treatment of the inner cladding surface and evacuation of the cladding tube: repeated depressurization of inner cladding volume due to H_2 absorption

1) stepwise hydriding of cladding to 300 wppm H

***14 injections** with **p_{max}=0.6 bar** →*

$$\text{total } m_{H2} = 14 \times \left(\frac{\mu p_{max} V_{hot}}{R T_{hot}} + \frac{\mu p_{max} V_{cold}}{R T_{cold}}\right) = 0.05 \text{ g}$$

$M_{clad\ 125\ cm}$ = 170 g C_H = 0.05/170 ≈ **300 wppm**

2) stepwise hydriding of cladding to 100 wppm H

***5 injections** with **p_{max}=0.6 bar** →*

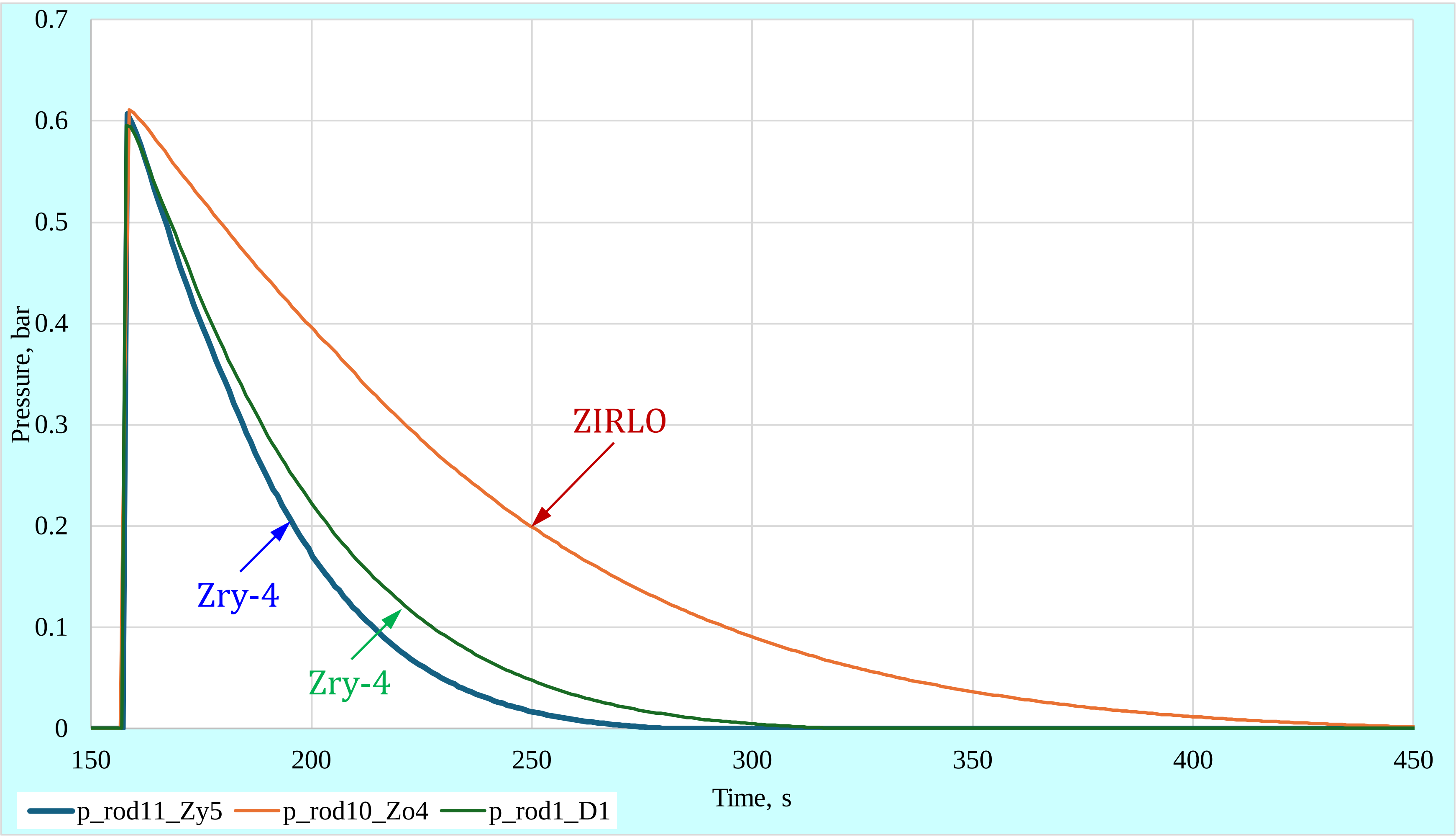
$$\text{total } m_{H2} = 5 \times \left(\frac{\mu p_{max} V_{hot}}{R T_{hot}} + \frac{\mu p_{max} V_{cold}}{R T_{cold}}\right) = 0.0179 \text{ g}$$

$M_{clad\ 125\ cm}$ = 170 g C_H = 0.0179/170 ≈ **100 wppm**

HOKI oven with axial temperature profile: T ≈ 450 °C along the length of ≈ 1250 mm

*after pulse filling followed **homogenization** stage at 450 °C during 4 h and then almost linear cooldown to 50 °C during 12 h*

Hydrogenation duration (1 cycle) for different claddings materials



Slower charging of ZIRLO in comparison to Zry-4 (difference in diffusion coefficients?)

Determination of axial distribution of hydrogen concentration by increase of circumferential strain at each elevation

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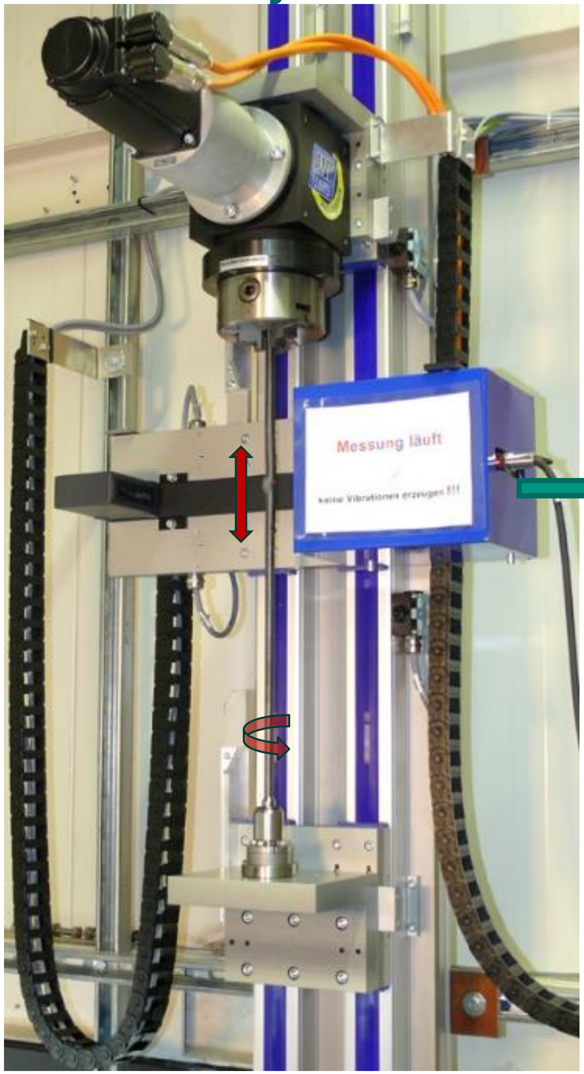
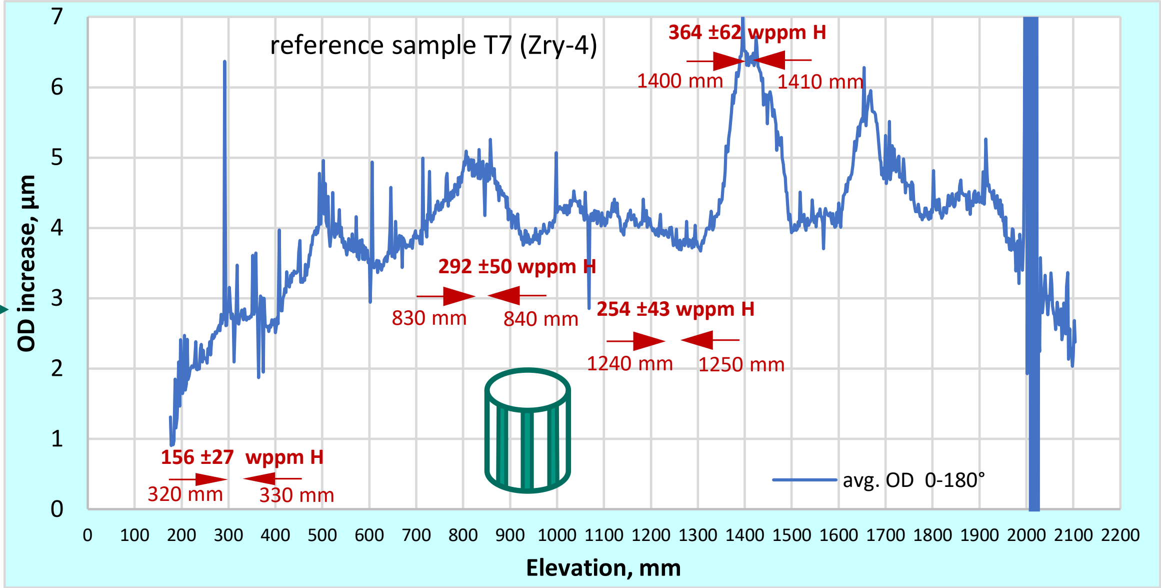


profilometry of clads with laser scanner

steps: 3° tangential, 2 mm axial

(thermal expansion ±0.3 µm for ±5 °C)

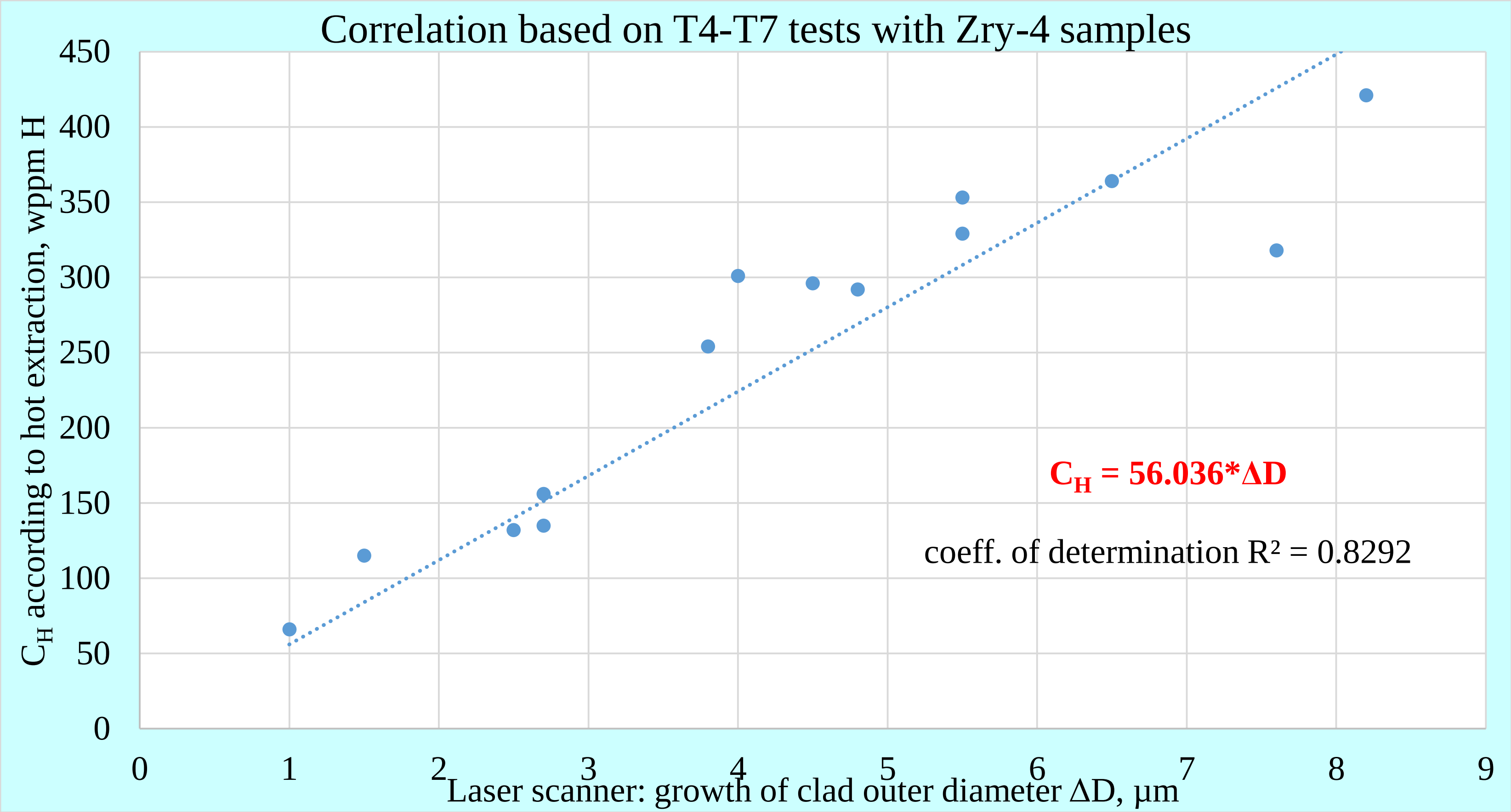


axial profile of cladding diameter increase (laser scanner)

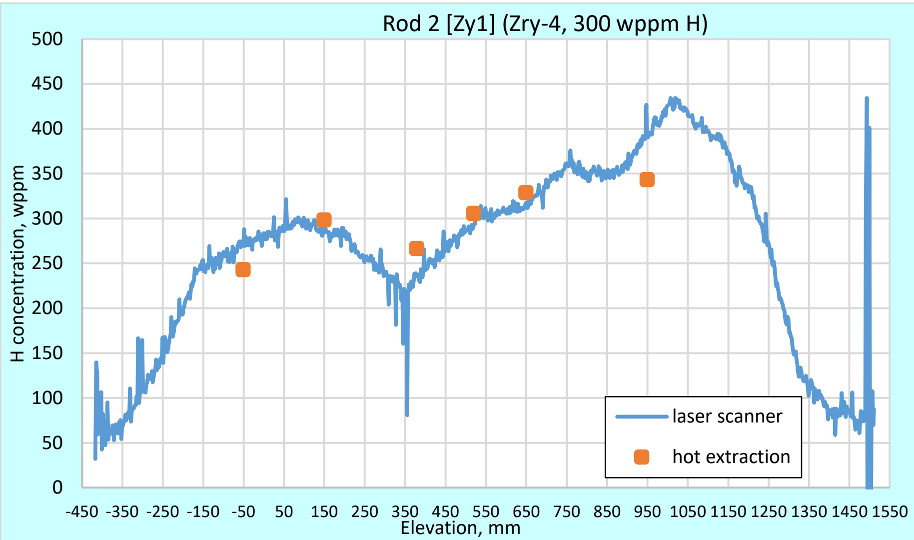
and corresponded hydrogen concentrations (hot extraction from 3 lamellas 10x1 mm)

Correlation between hydrogen content and increase of averaged cladding diameter

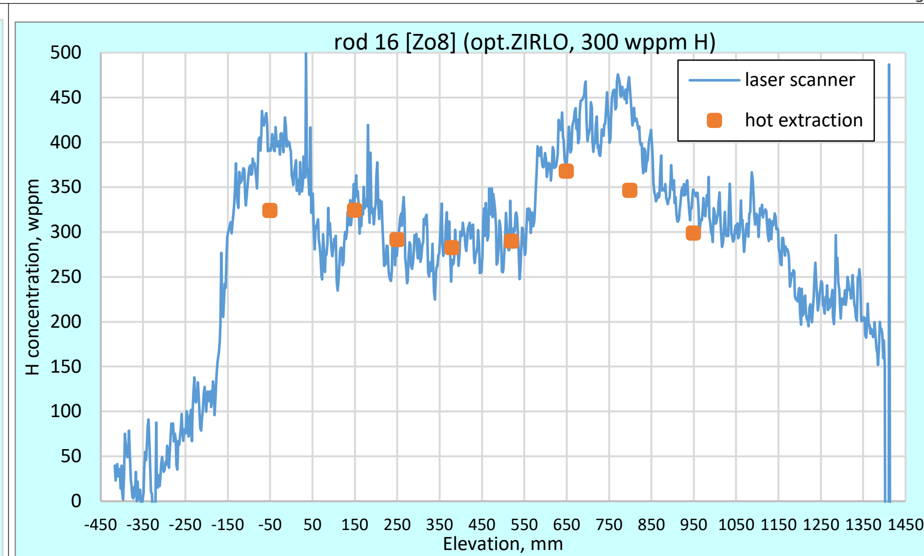
sample	ref. T4			ref. T5			ref. T6				ref. T7			
axial position, mm	750	1250	1650	750	1250	1650	405	755	1155	1455	325	835	1245	1405
OD increase, µm (laser scanner)	**7.6**	**4**	**5.5**	**8.2**	**5.5**	**4.5**	**1**	**2.5**	**2.7**	**1.5**	**2.7**	**4.8**	**3.8**	**6.5**
hot extraction, wppm	**318**	**301**	**329**	**421**	**353**	**296**	**66**	**132**	**135**	**115**	**156**	**292**	**254**	**364**
± wppm	48	45	49	105	88	74	17	33	34	29	27	50	43	62
SD				25	3	2	27	2	1	5	4	13	6	3



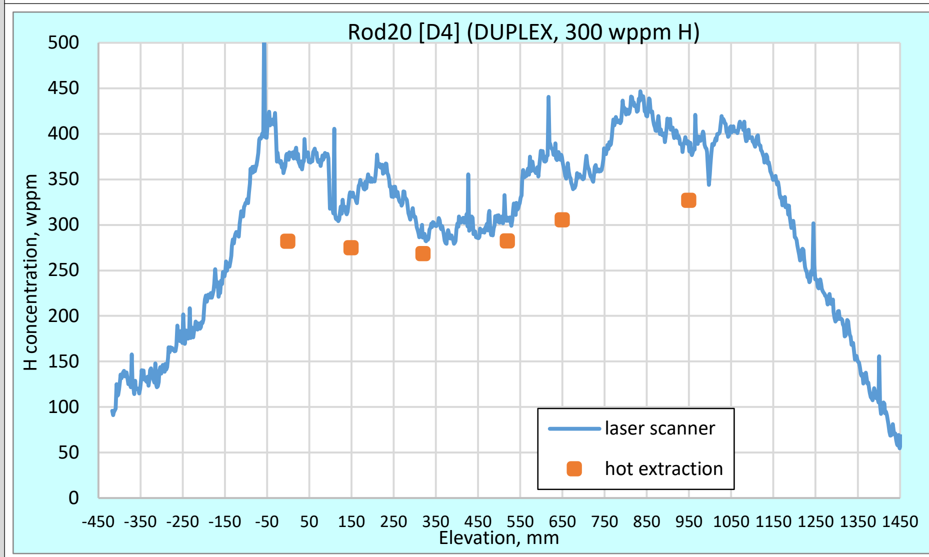
Axial hydrogen distribution for claddings: comparison of laser scanner data and post-test hot extraction



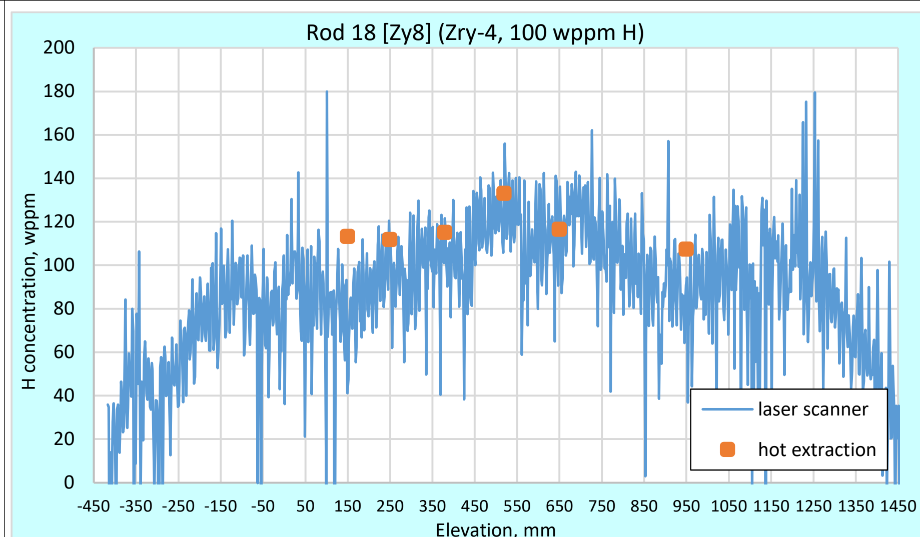
Zry-4, 300 wppm



opt. ZIRLO, 300 wppm

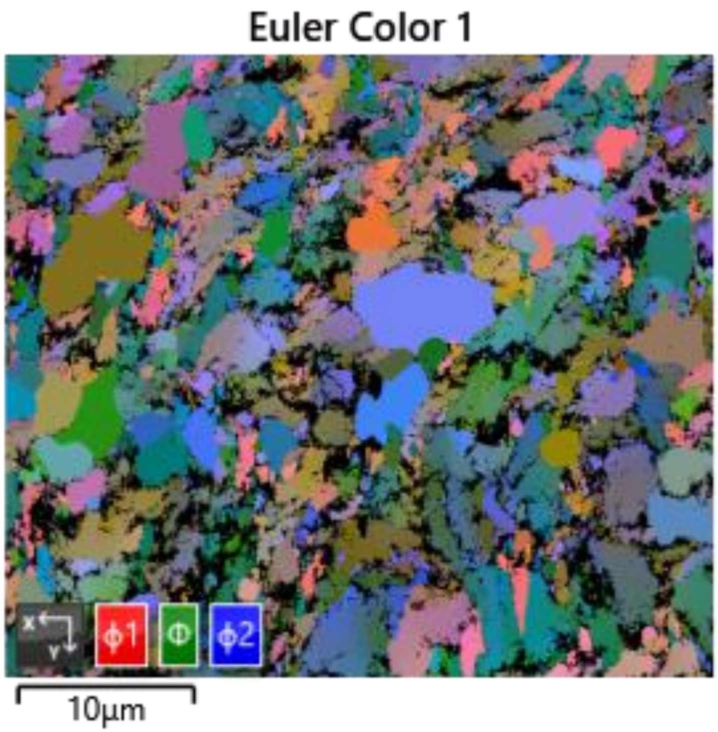


DX-D4, 300 wppm

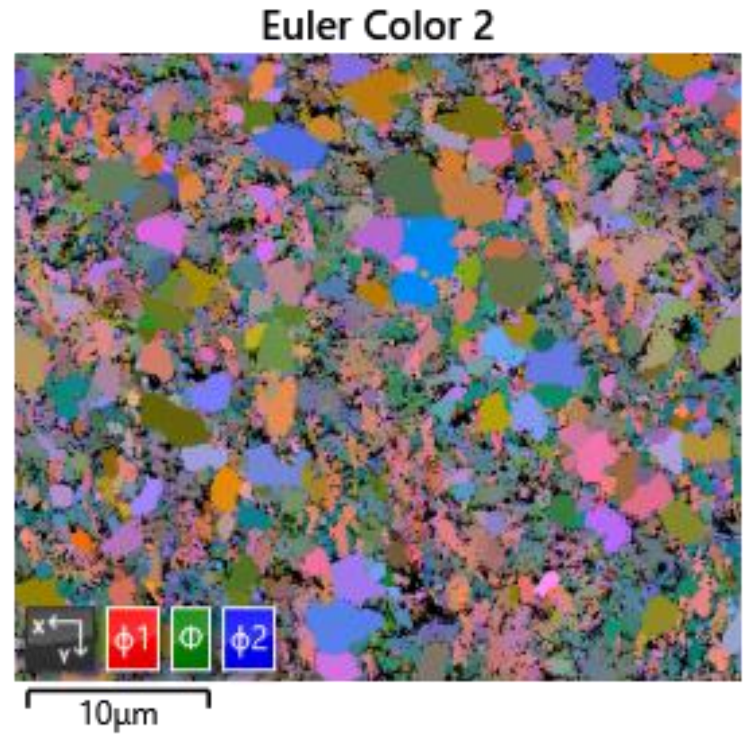


Zry-4, 100 wppm

Microstructure of as-received claddings: EBSD analysis

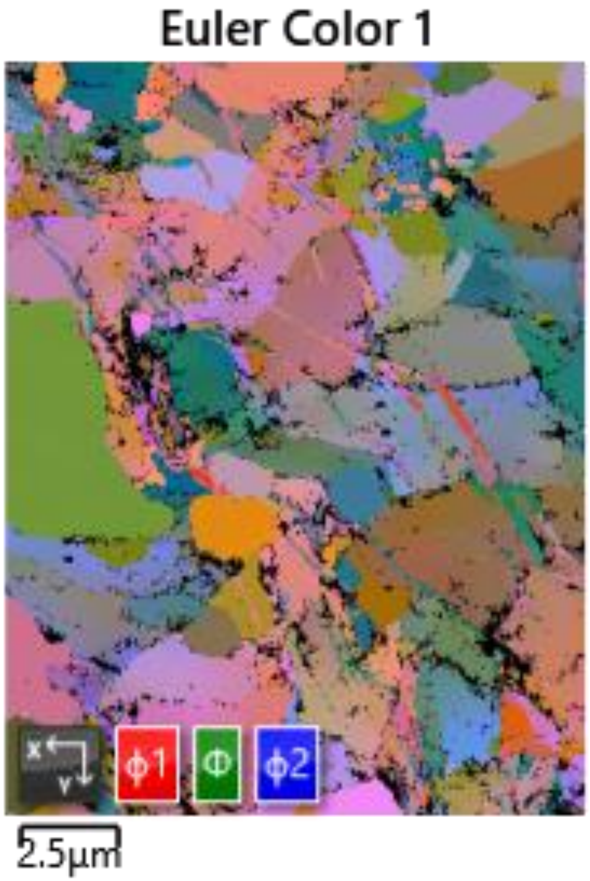


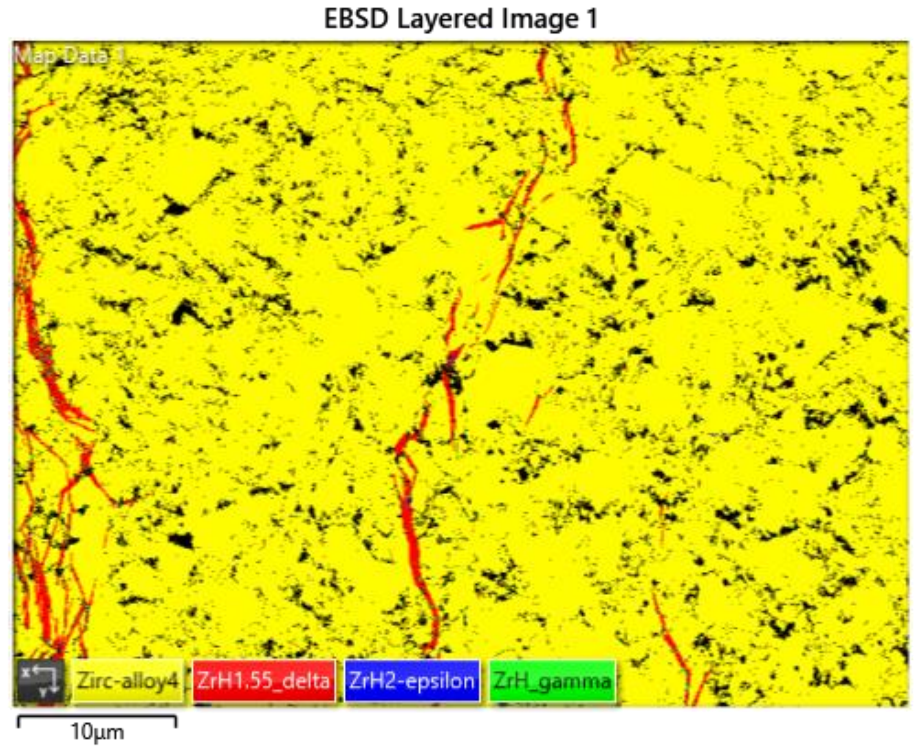
cold-worked and stress-relieved (CWSR) **Zry-4**:
avg. grain area: weight mean **5.74 μm²**
(biaxial grains $\overline{D} \approx 3.6$ μm, $\bar{d} \approx 2.0$ μm)



partially recrystallized (PRXA) **opt. ZIRLO**:
avg. grain area: weight mean **2.92 μm²**
($\overline{D} \approx 2$ μm)

Microstructure of <u>hydrogenated</u> claddings: EBSD analysis





CWSR **Zry-4** hydrogenated at 450 °C (total heat treatment ≈ 1 day): avg. grain area weight mean **12.79 µm²** (biaxial grains $\overline{D}$ ≈ 4.9 µm, $\bar{d}$ ≈ 3.3 µm)	Zry-4: inter- and intra-granular δ-hydrides with avg. length of ≈10 µm and width ≈0.3…0.6 µm; radial distance between hydrides ≈20 µm
<u>**doubling of the grain area**</u> in comparison to as-received cladding	

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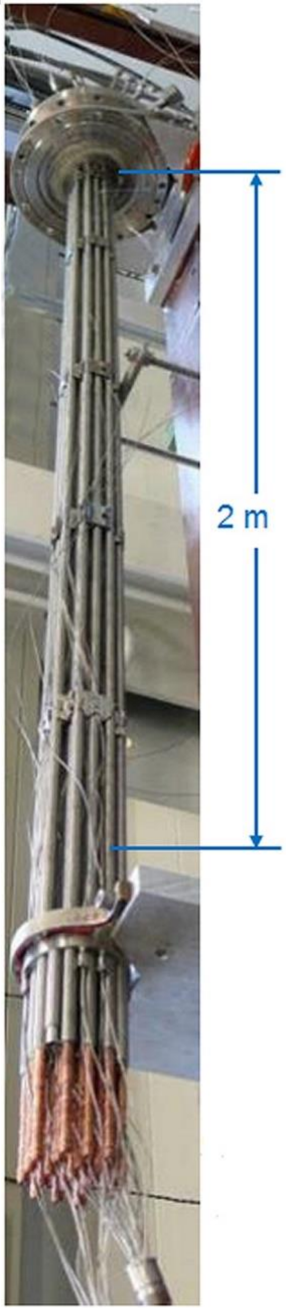
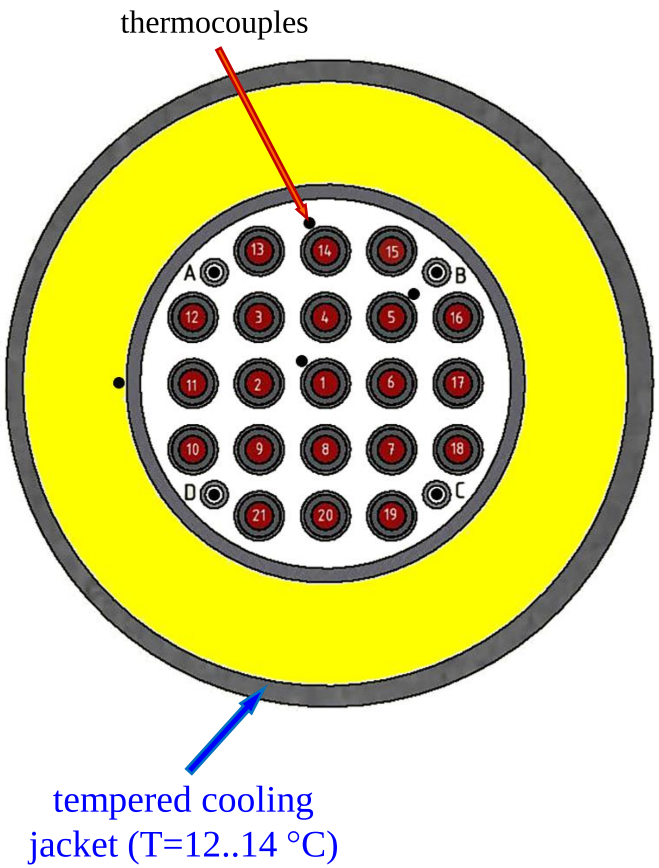
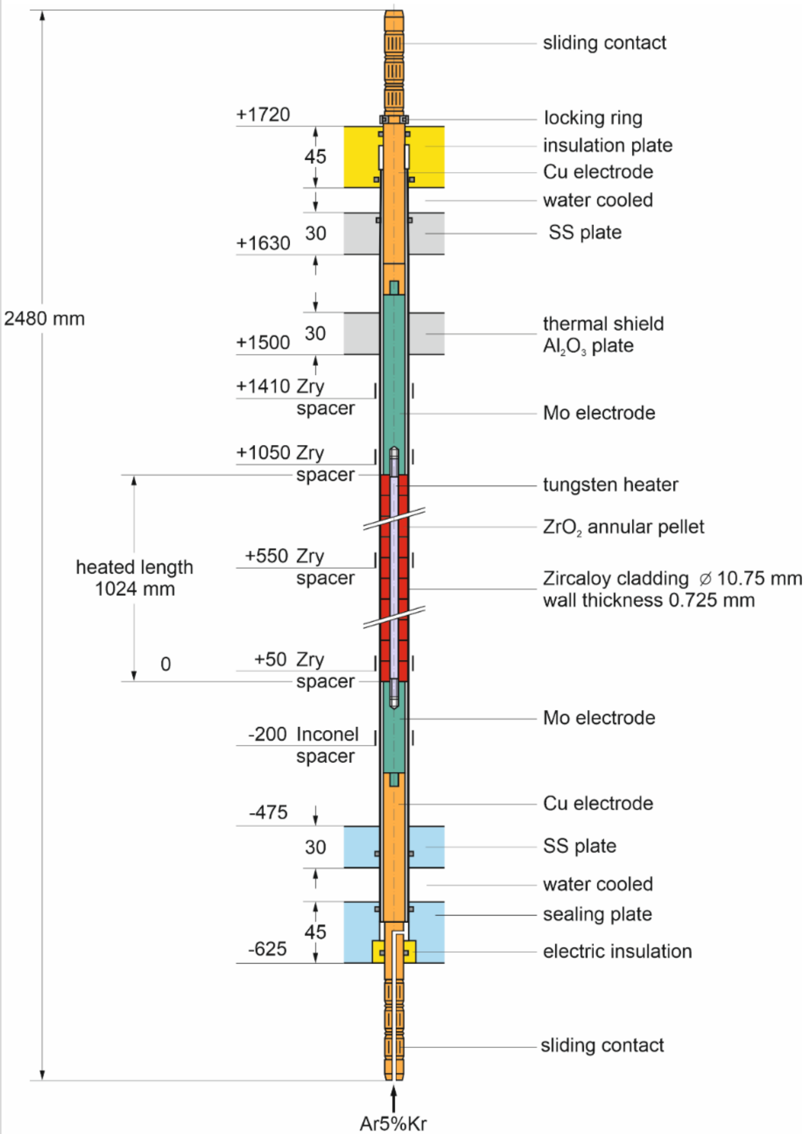
SPIZWURZ bundle





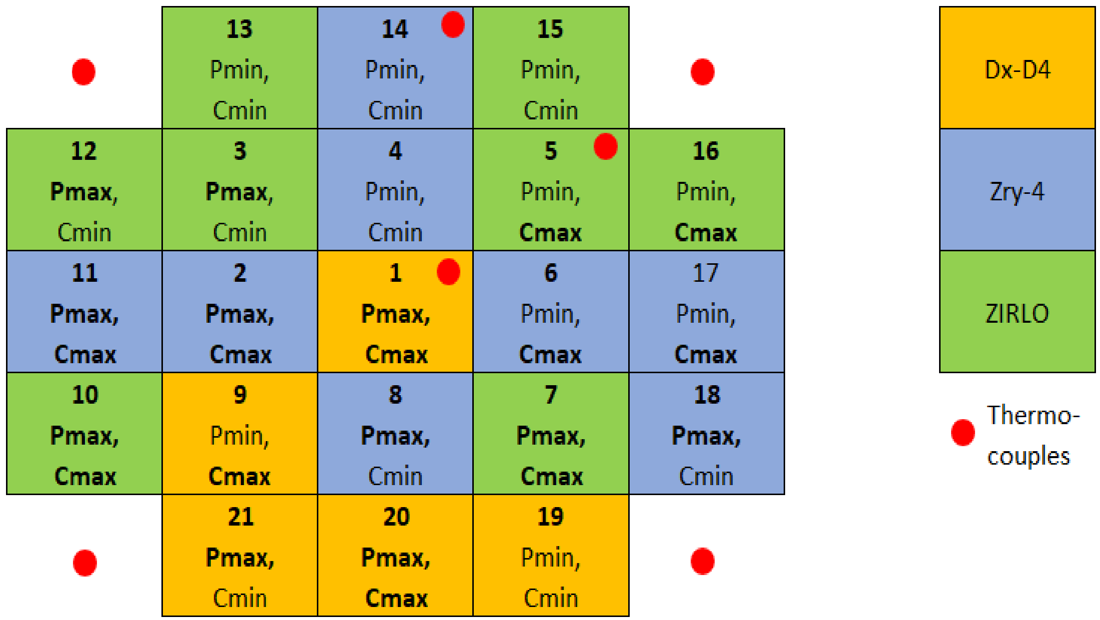
Bundle size		21 heated rods
Pitch		14.3 mm
Corner rod (4)	material	Zircaloy-4
	instrumented (A, B, C, D)	tube ∅ 6x0.5 (bottom: -1140 mm)
Grid spacer	material	Zircaloy-4
	length	42 mm
	sheet thickness	0.5 mm
	elevation of lower edge	Zry: -100, 150, 550, 1050, 1410 mm
Shroud	material	Zirconium 702 (flange: Zry-4)
	wall thickness	3.17 mm
	outside diameter	86.0 mm
	length (extension)	1600 mm (-300 mm to 1300 mm)
Shroud insulation	material	ZrO_2 fiber
	insulation thickness	~ 36 mm
	elevation	-300 to ~1000 mm
Cooling jacket	Material: inner/outer	Inconel 600 (2.4816) / SS (1.4571)
	inner tube	∅ 158.3 / 168.3 mm
	outer tube	∅ 181.7 / 193.7 mm
Thermocouples	at cladding surfaces	rods 1, 5, 9; totally 3x15=45
	inside corner rods	one at each elevation 2-16, totally 15
	at shroud outer surface	one at each elevation 3-15, totally 13

Heated rod



Cladding OD / ID		**10.75 / 9.3 mm**
Cladding thickness		**0.725 mm**
Cladding length	**(position in the bundle)**	**2278 mm (between -593 and 1685 mm)**
Rod length	(elevations)	2480 mm (-690 to 1790 mm)
Internal rod pressure; gas		5.5 MPa abs.; Kr
Material of middle heater	 surface roughness	Tungsten (W) Ra=1.6 µm
Tungsten heater length		1024 mm (between 0 and 1024 mm)
Tungsten heater diameter		4.6 mm
Annular pellet	material dimensions surface roughness	ZrO_2;Y_2O_3-stabilized ∅ 9.15/4.75 mm; L=11 mm Ra=0.3 µm
Pellet stack		0 mm to ~1020 mm
Molybdenum heaters and copper electrodes	length of upper part length of lower part outer diameter: prior to coating after coating with ZrO_2 coat. surface roughness borehole of Cu-electrodes	766 mm (576 Mo, 190 mm Cu) 690 mm (300 Mo, 390 mm Cu) 8.6 mm 9.0 mm Ra=6-12 µm diameter 2 mm, length 96 mm
Gas volume inside the rod	**heated**	**15 cm³**
Gas volume outside the rod	**not heated (room T)**	**20 cm³**

Bundle composition

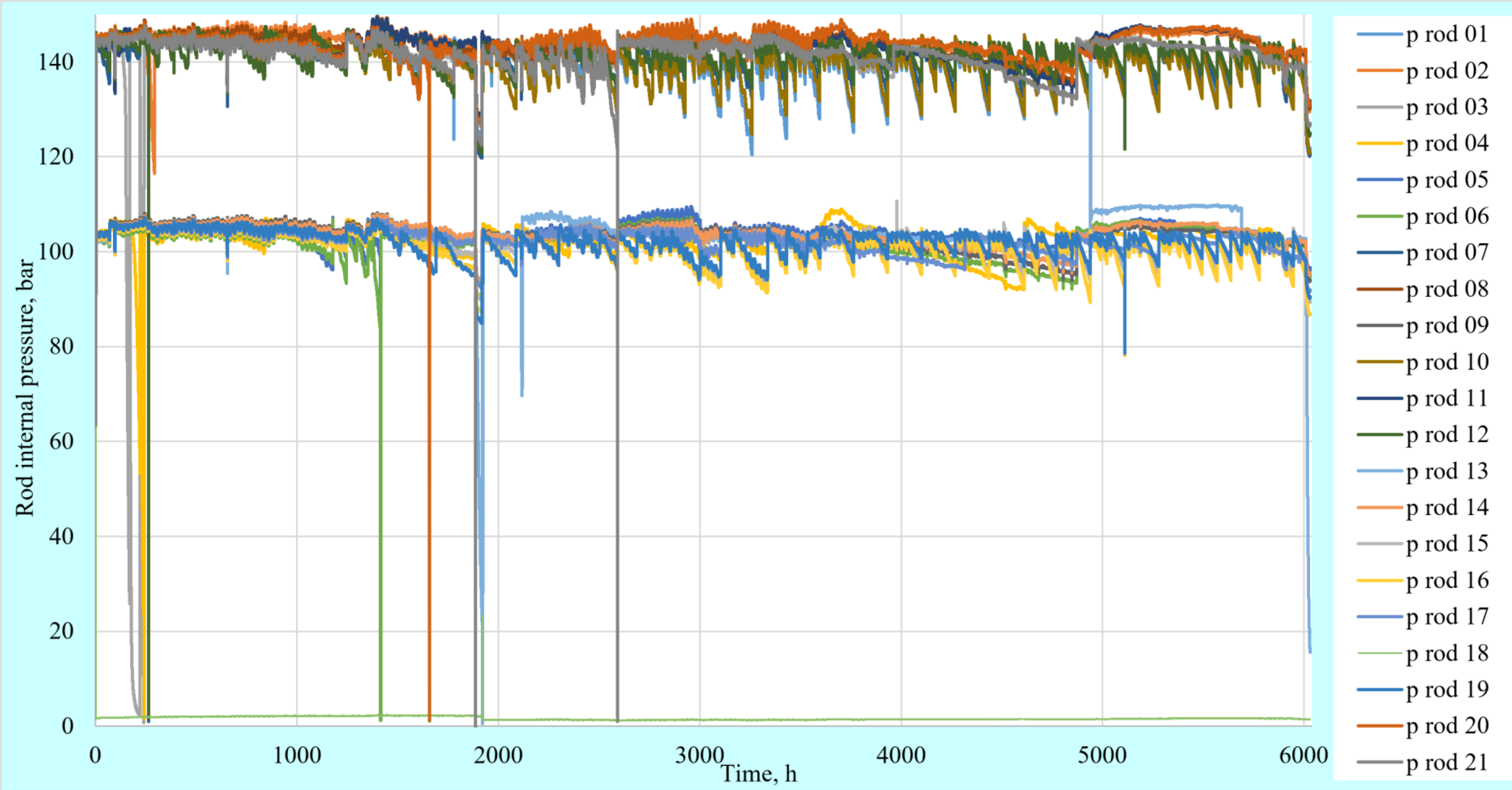


number	rod	alloy	H conc., wppm	mark
D1	1	*DUPLEX*	300	D1
Zry0212	2	*Zry-4*	300	Zy1
Zo049	3	*ZIRLO*	100	Zo1
Zy31	4	*Zry-4*	100	Zy2
Zo087	5	*ZIRLO*	300	Zo2
Zy85	6	*Zry-4*	300	Zy3
Zo156	7	*ZIRLO*	300	Zo3
Zry197	8	*Zry-4*	100	Zy4
D2	9	*DUPLEX*	300	D2
Zo165	10	*ZIRLO*	300	Zo4
Zry199	11	*Zry-4*	300	Zy5
Zo220	12	*ZIRLO*	100	Zo5
Zo221	13	*ZIRLO*	100	Zo6
Zy914	14	*Zry-4*	100	Zy6
Zo332	15	*ZIRLO*	100	Zo7
Zo351	16	*ZIRLO*	300	Zo8
Zry1021	17	*Zry-4*	300	Zy7
	18	*Zry-4*	100	Zy8
D3	19	*DUPLEX*	100	D3
D4	20	*DUPLEX*	300	D4
D5	21	*DUPLEX*	100	D5

hydrogen content in cladding
Cmax = 300 wppm H
Cmin = 100 wppm H

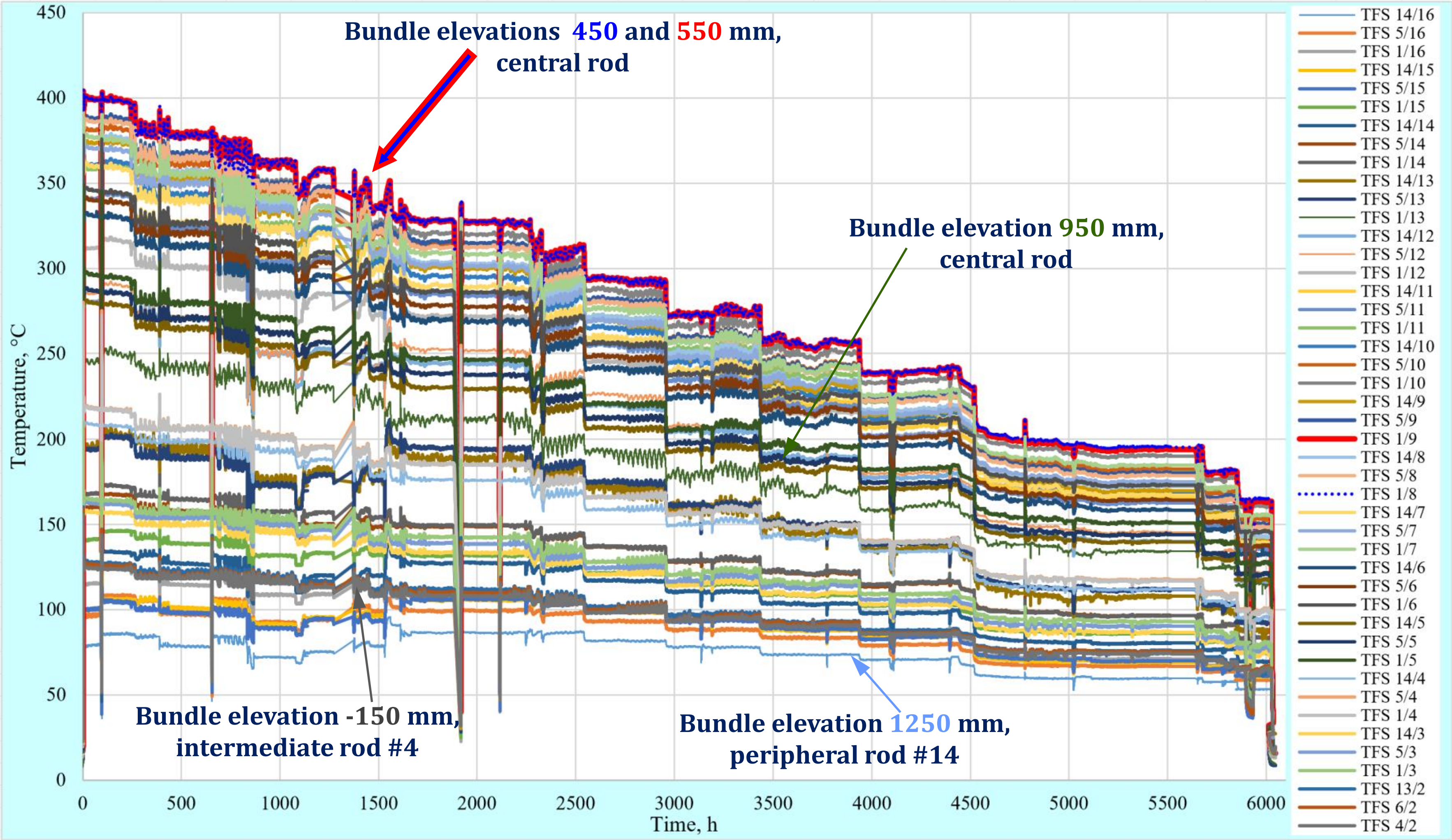
pressure inside the rod
Pmax = 146 bar
Pmin = 106 bar

Pressures inside the rods between 12.05.2023 and 18.01.2024



- two pressurization levels of **106 and 146 bar**
- daily refill of 7 rods with $Ar+O_2$ due to small leakages
- short depressurizations of 6 rods due to change of sealing rings

Reading of cladding surface thermocouples (TFS) between 12.05.23 and 18.01.24



- peak cladding temperature decreased in steps with 15 K decrement and average duration of 400 h, average cooling rate 0.9 K/day
- periodic daily temperature fluctuations ≈±1.5 K for each thermocouple
- 4 el. power breakdowns

Smooth temperature decrease on the time of the power step reduction

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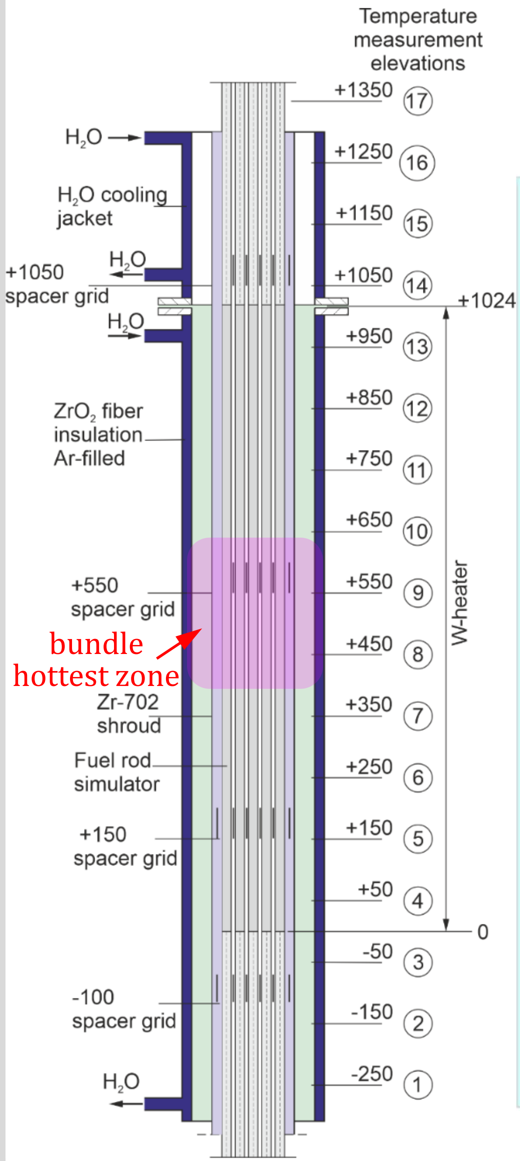
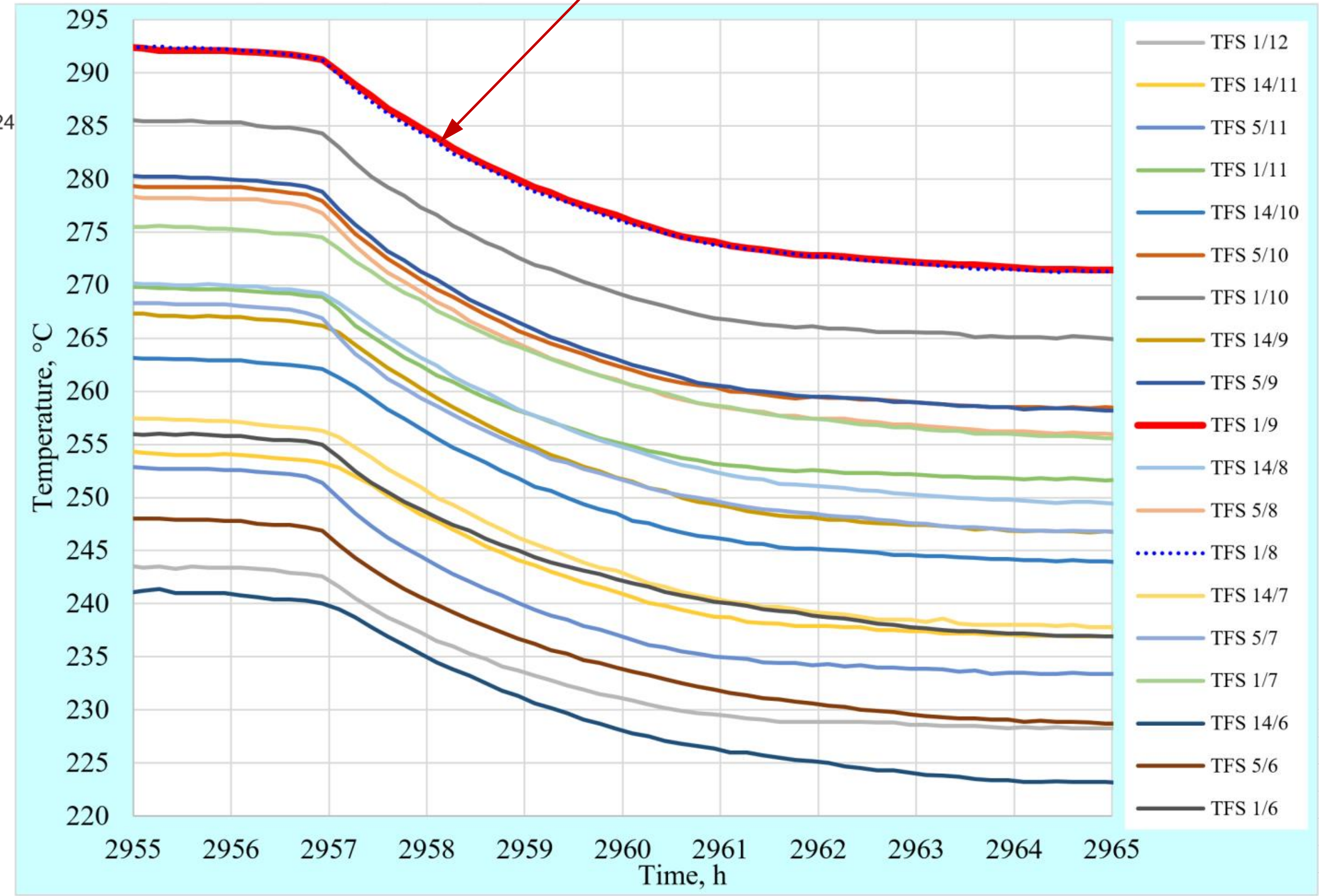
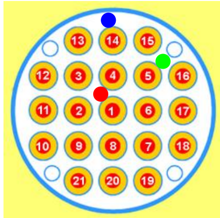
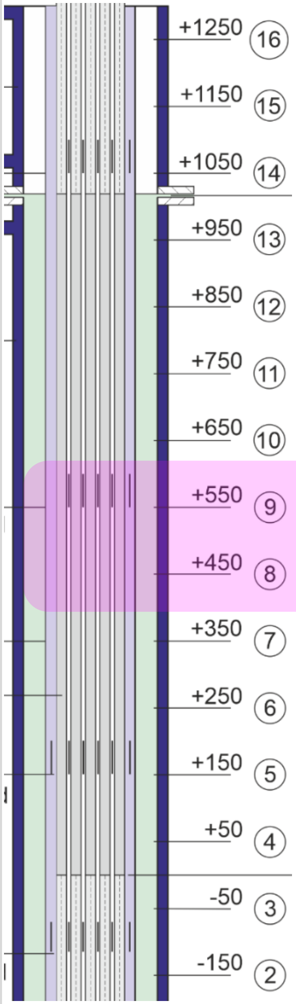


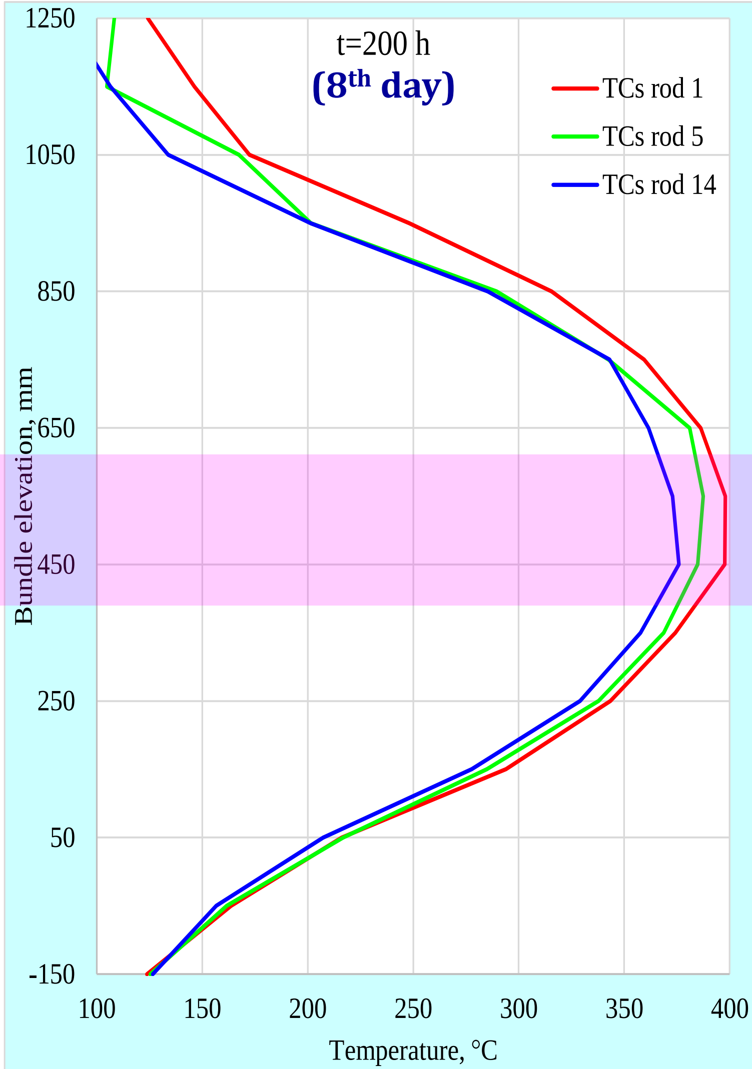
cooling rate decreased from 6 K/h ($1.7*10^{-3}$ K/s) to 0 K/s during 10 h

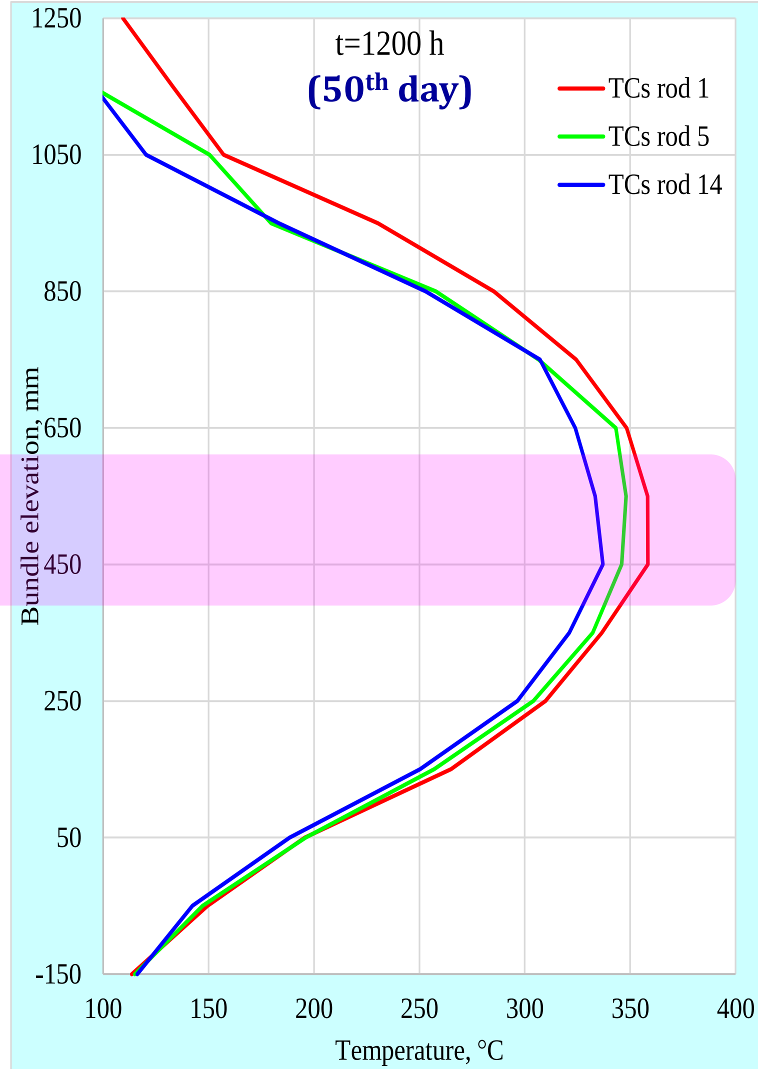


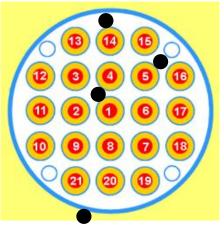


Axial temperature distribution for three rod groups

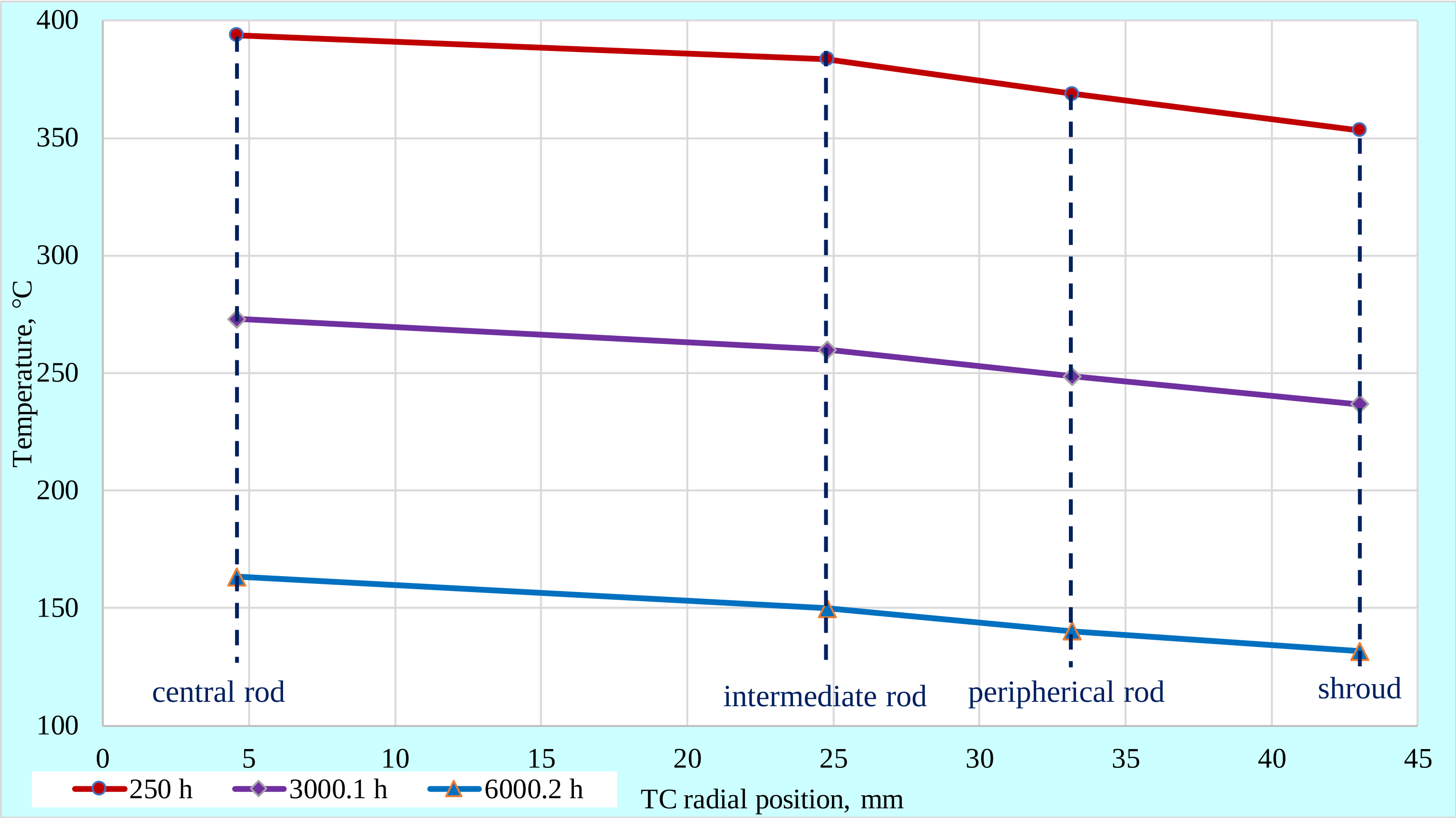








Radial temperature distribution



The radial temperature gradient: 40 K (test start), 30 K (test end)

Indirect estimation of radial symmetry in bundle: comparison of corner rods and shroud temperatures

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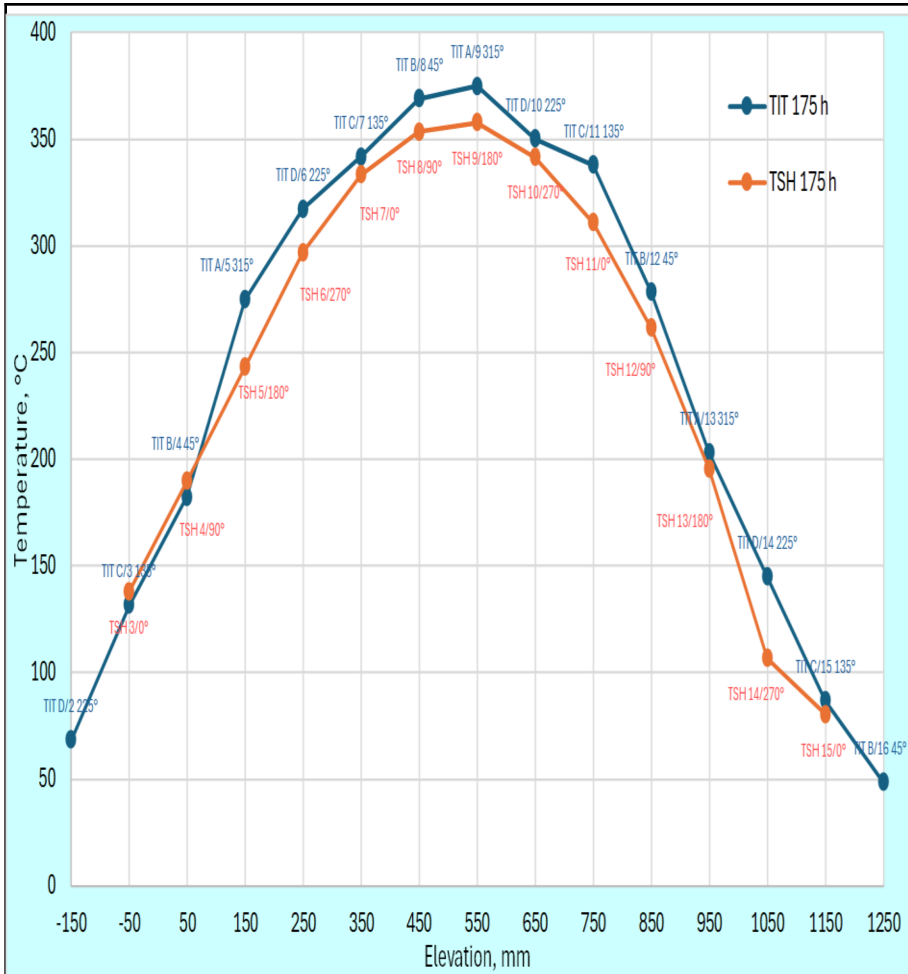


t = 175 h

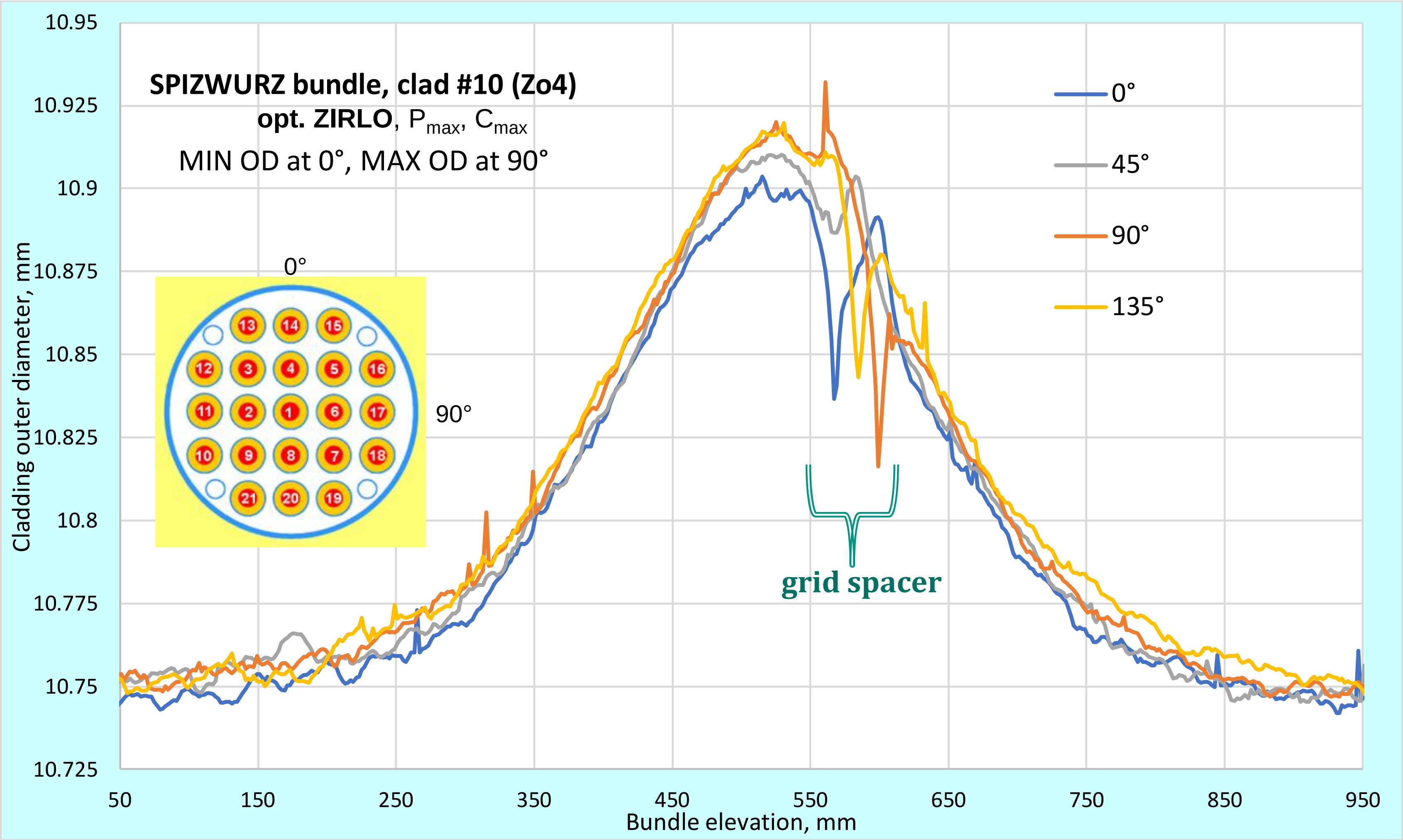
t = 4000 h

The circumferential temperature scatter is ±8 °C at the beginning of the test and ±5 °C before the end of the test

SPIZWURZ bundle: post-test view between two middle grid spacers

			
0°	90°	180°	270°

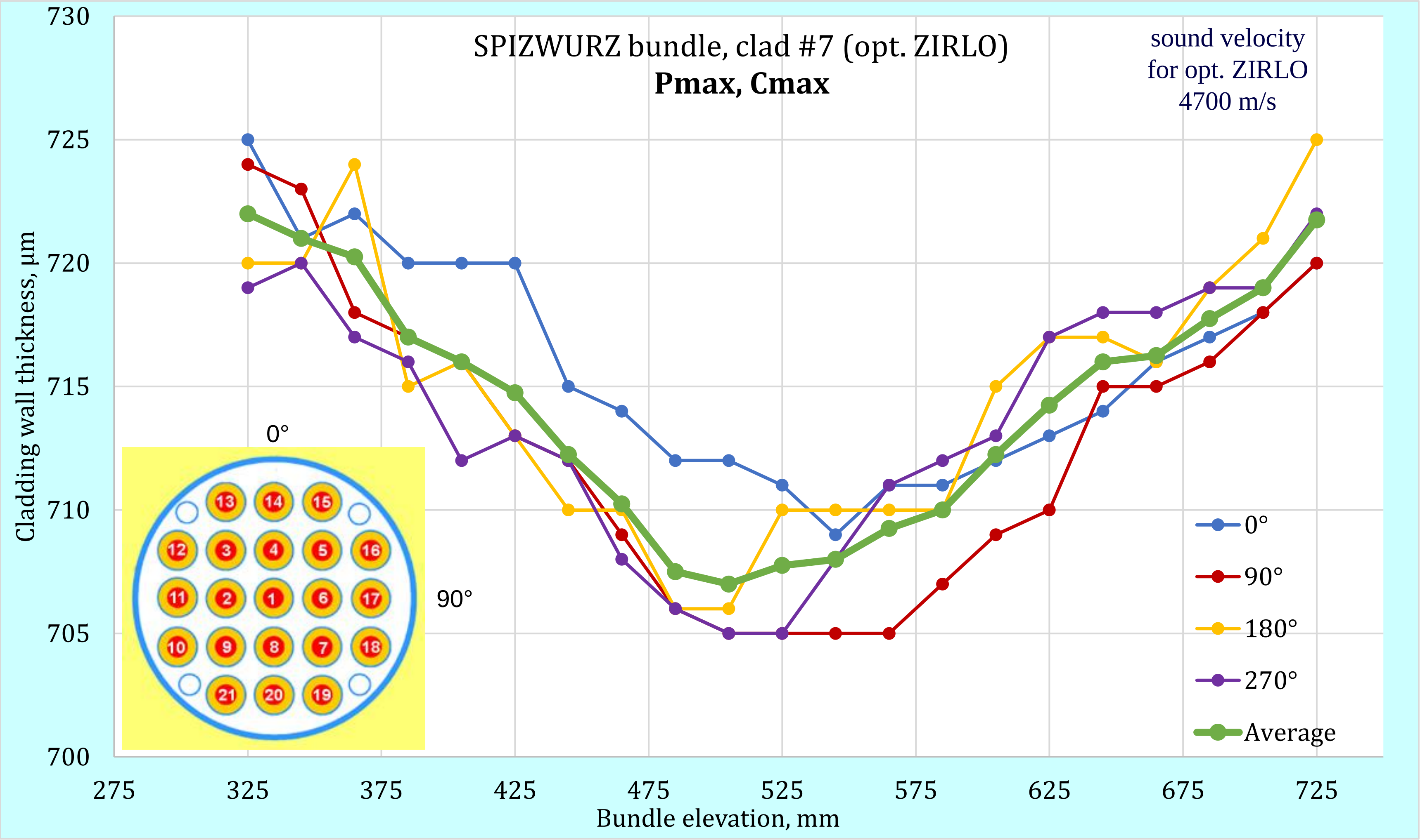
Results of creep measurements with the laser scanner: 1) small ovality of cladding (comparison of scanning at 4 angles); 2) influence of grid spacer springs (creep limitation)



Max cladding ovality OD_{max}- OD_{min} = 20 µm at 520 mm (hottest bundle elevation);

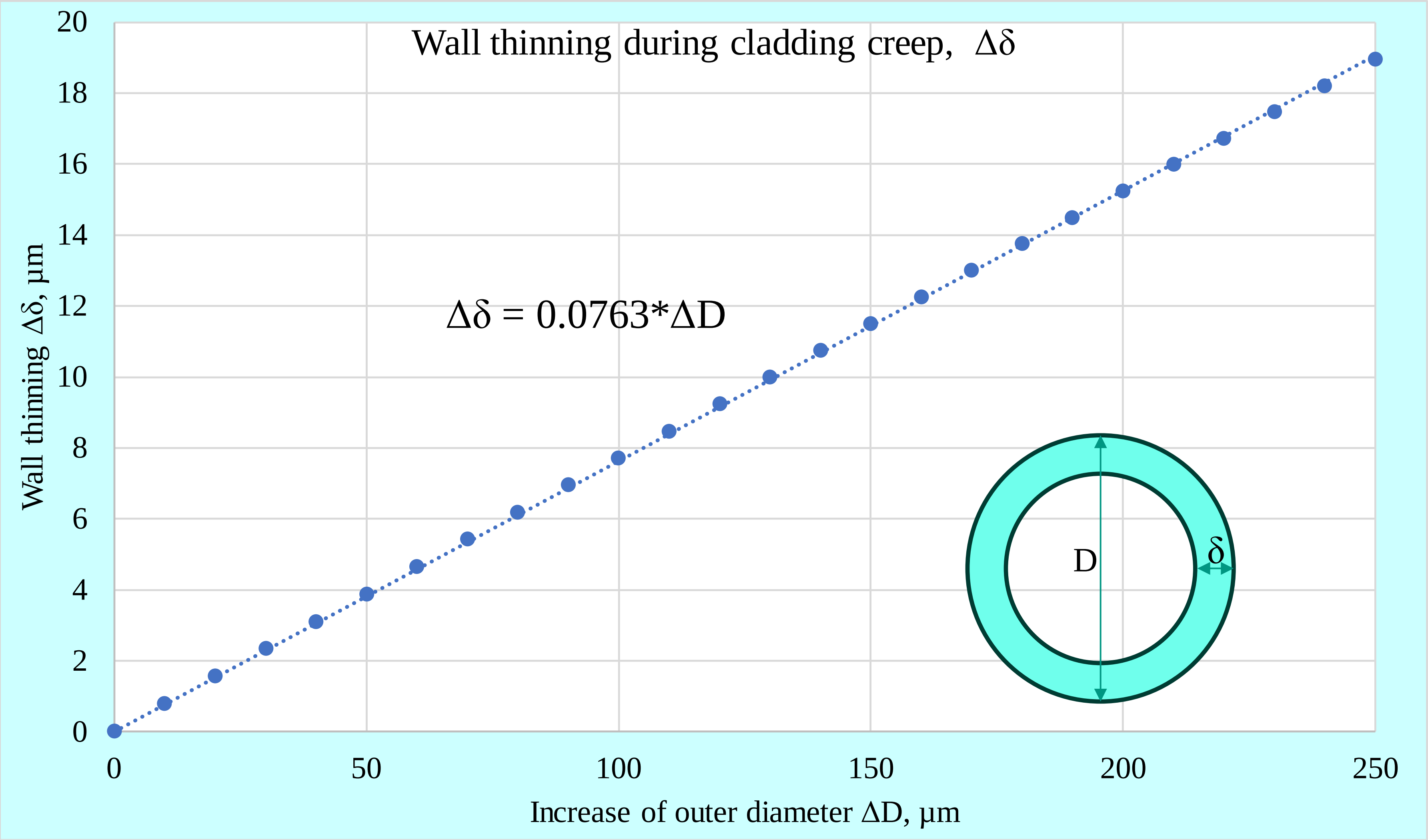
average ovality along the cladding is 8 µm, what is comparable with delivered ovality of 5 µm

Ultrasound measurements of wall thickness along four cladding sides: not symmetrical wall thinning at each bundle elevation

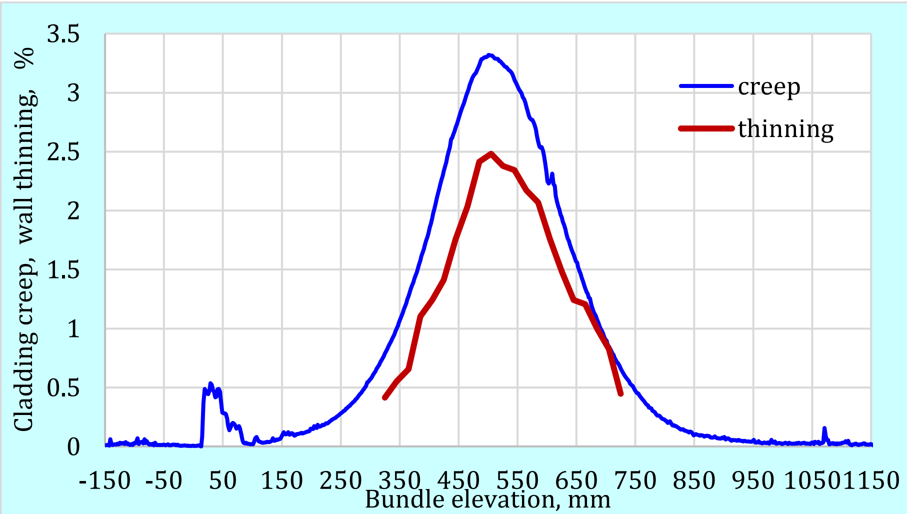


The probable reason of the asymmetrical thinning could be a shift of the pellets from the midline

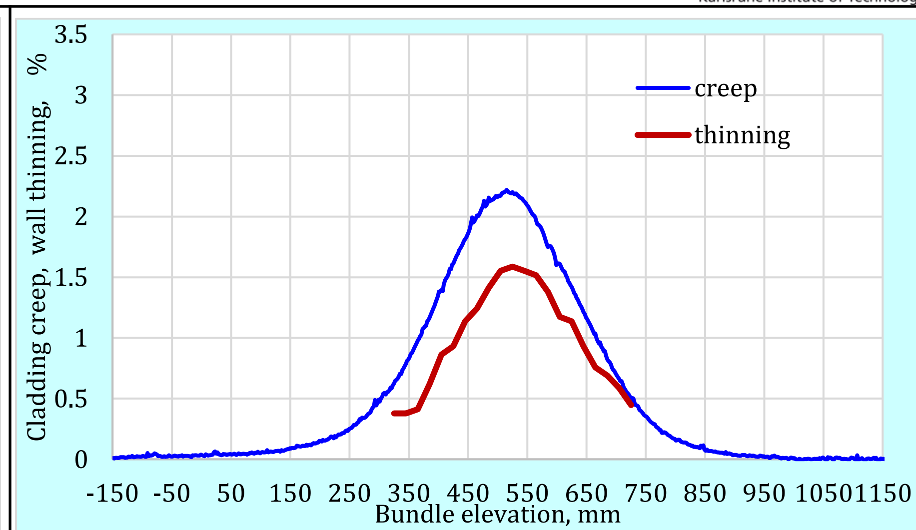
2D wall thinning during cladding creep (cross-section area change): calculated dependence of thinning on the increase in outer diameter



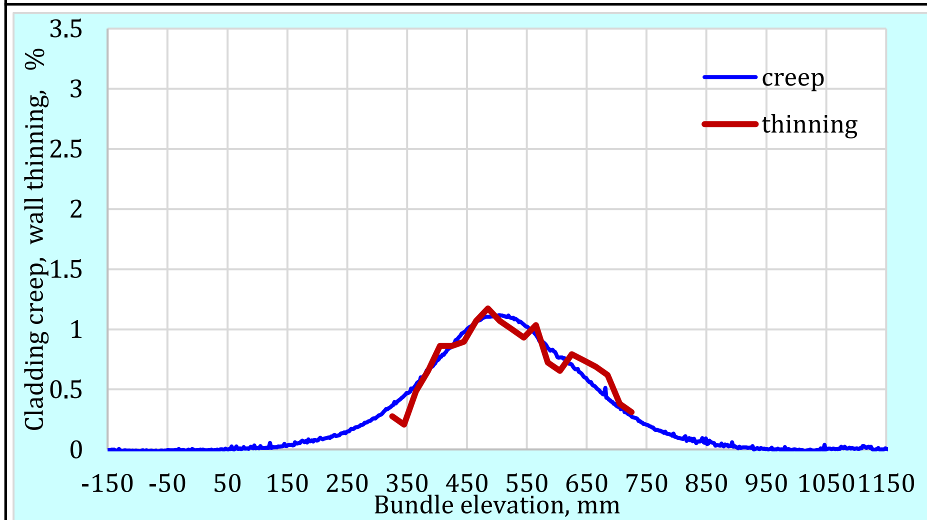
opt. ZIRLO: cladding creep, wall thinning



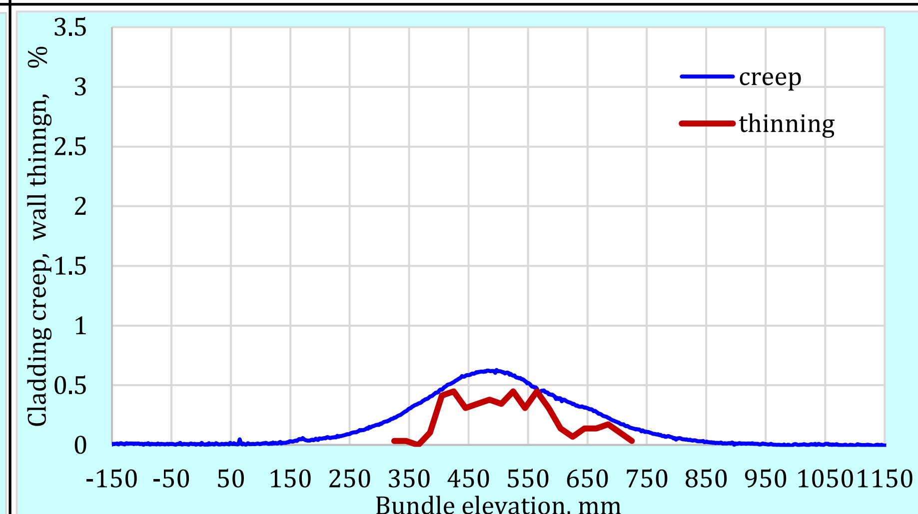
cladding 7: opt. ZIRLO, Pmax, Cmax



cladding 3: opt. ZIRLO, Pmax, Cmin

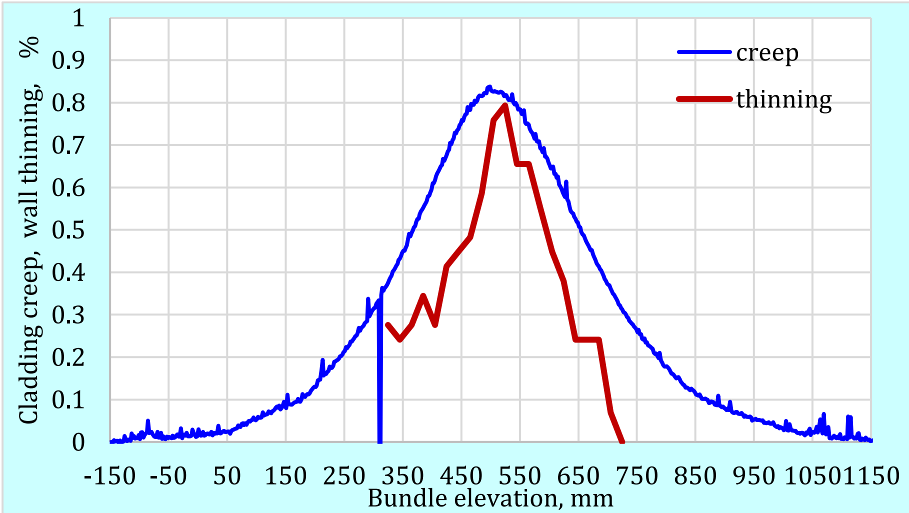


cladding 5: opt. ZIRLO, Pmin, Cmax

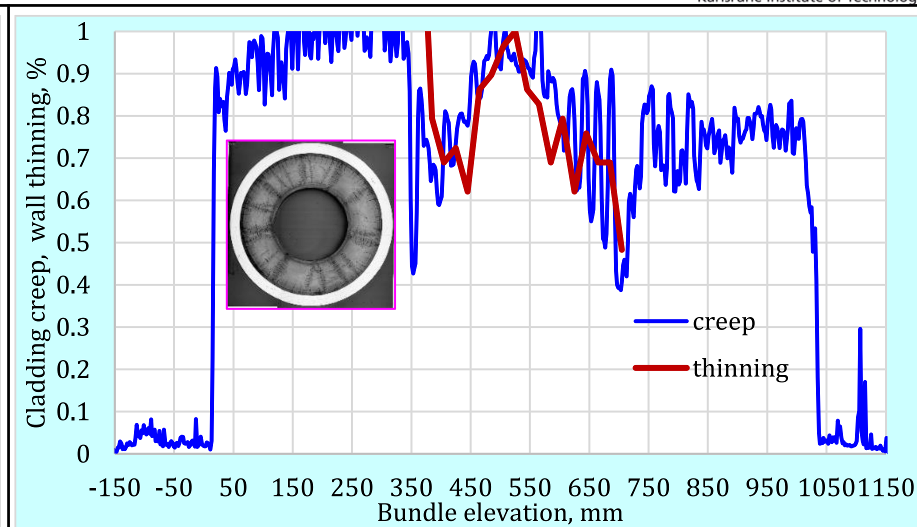


cladding 13: opt. ZIRLO, Pmin, Cmin

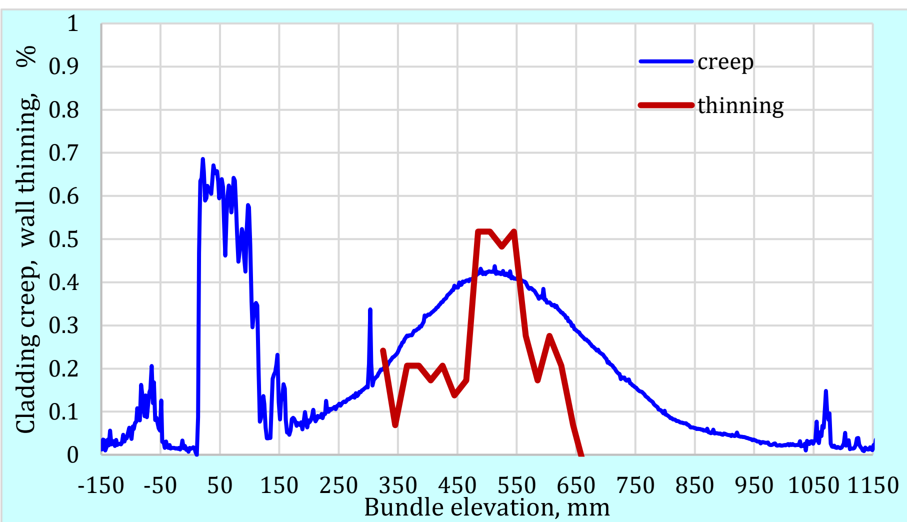
Zry-4: cladding creep, wall thinning



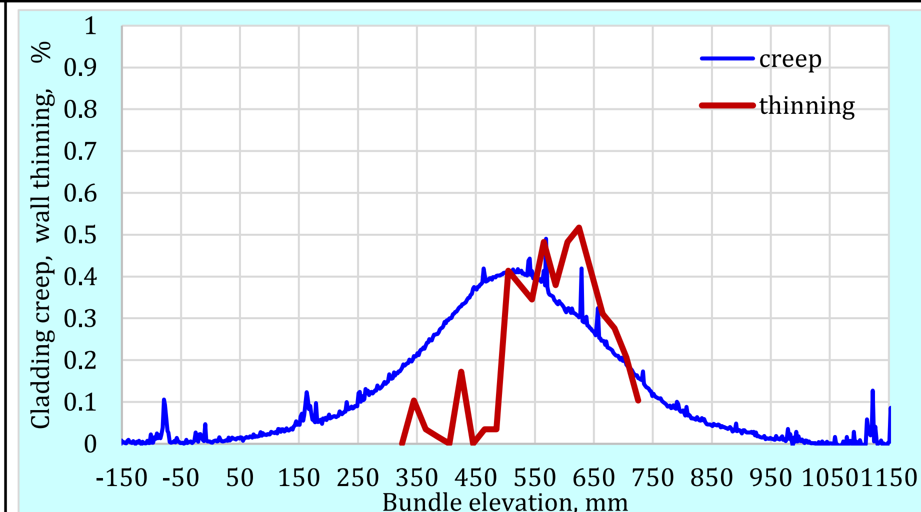
cladding 2: Zry-4, Pmax, Cmax



cladding 8: Zry-4, Pmax, Cmin;
contact of the cladding with the extended pellet

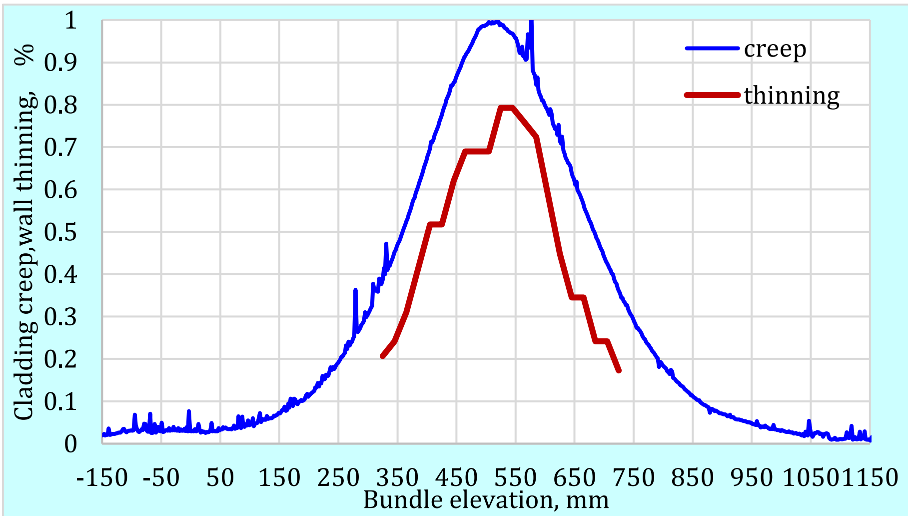


cladding 6: Zry-4, Pmin, Cmax

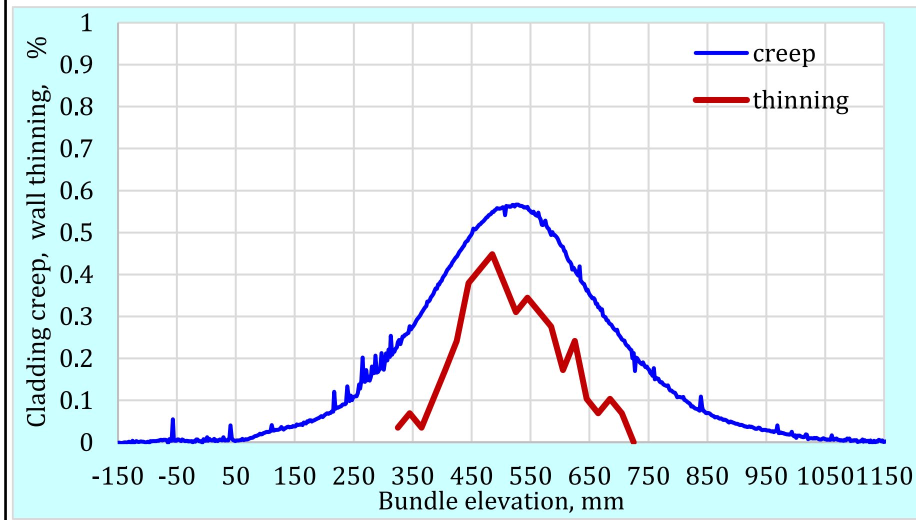


cladding 4: Zry-4, Pmin, Cmin

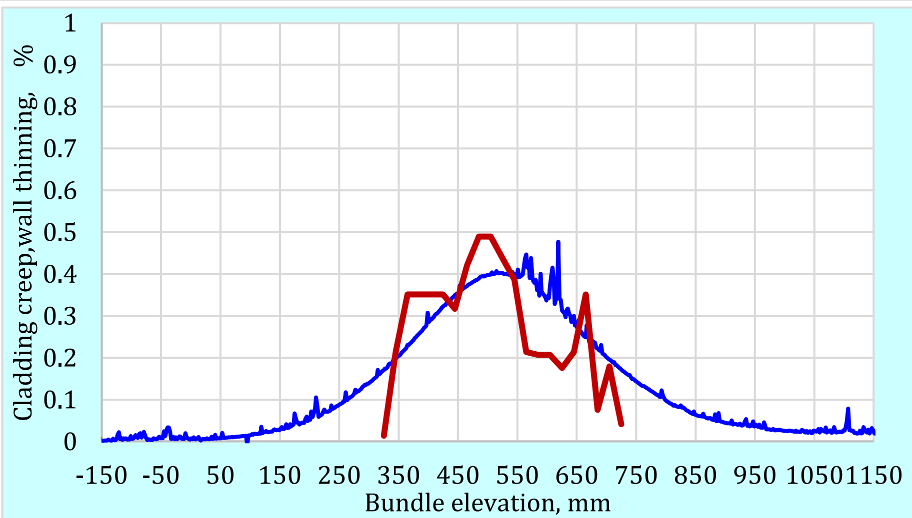
DX-D4 claddings: cladding creep, wall thinning



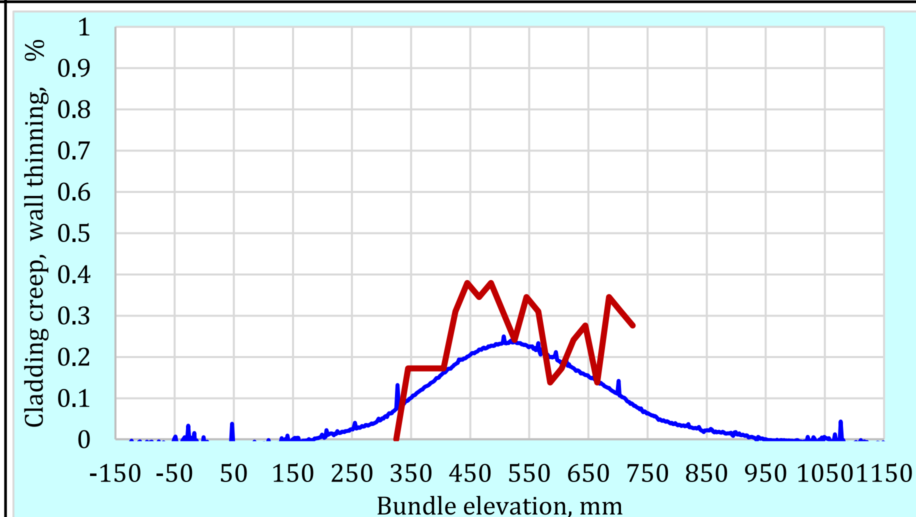
cladding 1: DX-D4, Pmax, Cmax



cladding 21: DX-D4, Pmax, Cmin

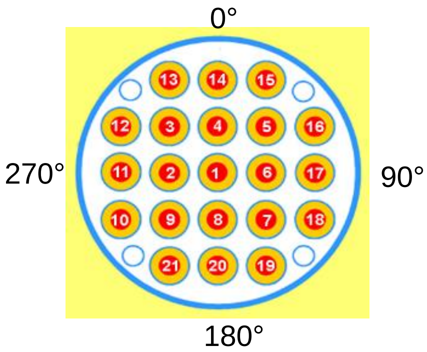


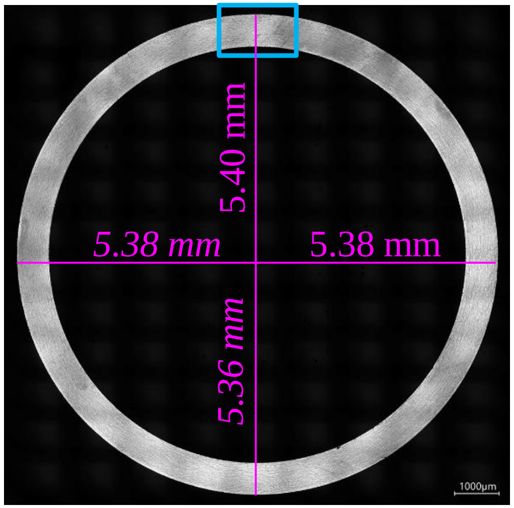
cladding 9: DX-D4, Pmin, Cmax



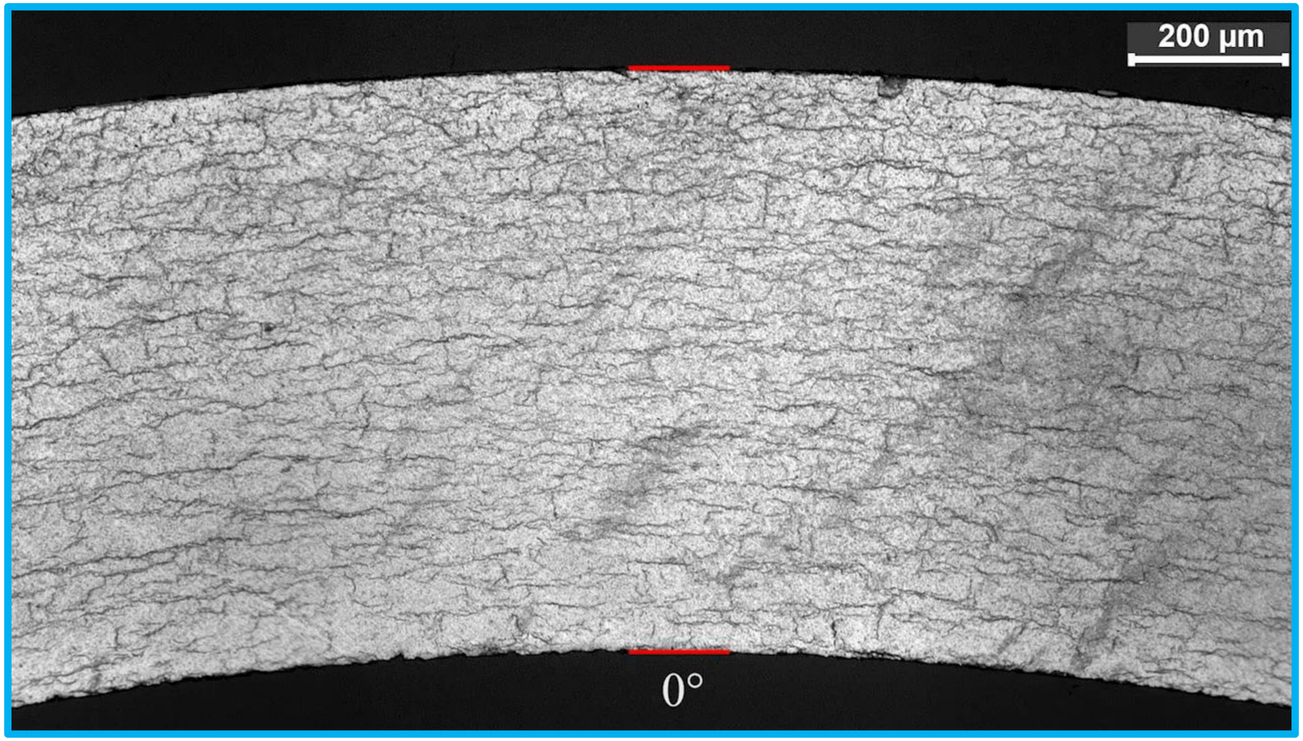
cladding 13: opt. ZIRLO, Pmin, Cmin

Post-test clad #11 (Zry-4, 146 bar, 300 wppm H), elevation 520 mm: metallographic observation of hydrides and cladding thinning





Link to movie:



- **Circumferential homogeneity of hydrides appearance;**
- **Not very radial symmetry for the wall thinning:**

angle	wall thickness, µm
0°	**721**
90°	718
180°	711
225°	708
270°	710

Post-test clad #10 (opt. ZIRLO, 146 bar, 300 wppm H), elevation <u>320 mm</u>: metallographic observation of hydrides and cladding thinning

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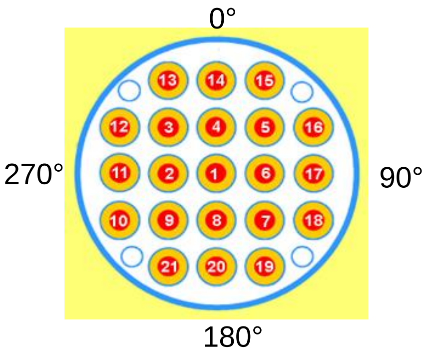
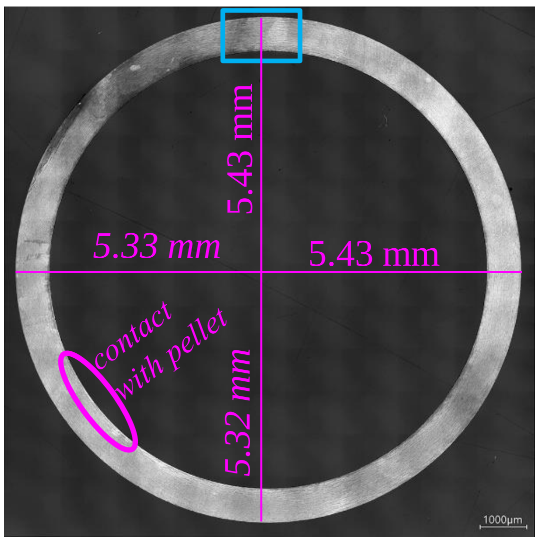
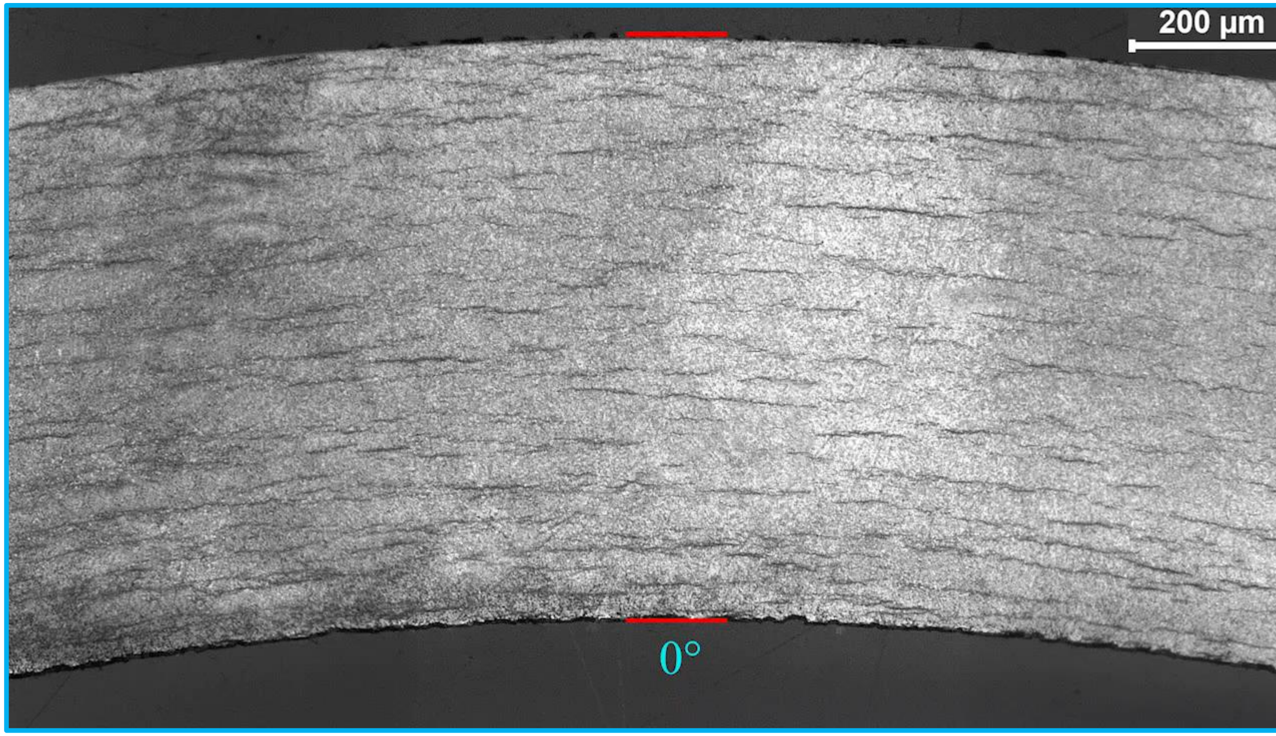
Link to movie:





Eccentricity



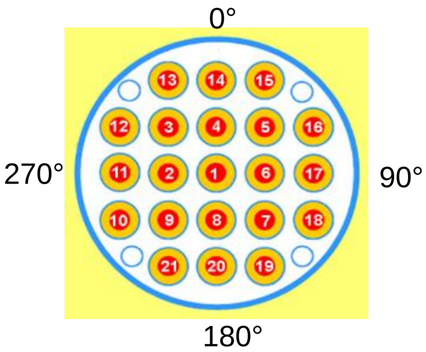
- **Circumferential homogeneity of hydrides appearance;**
- **Not very radial symmetry for the <u>wall thinning</u>:**

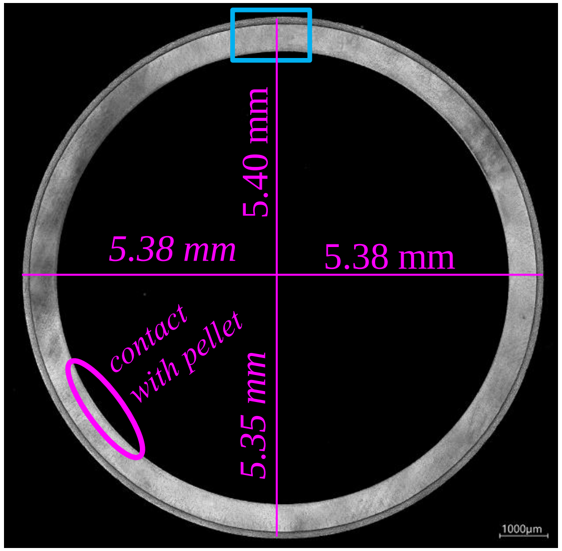
angle	wall thickness, µm
0°	721
90°	724
180°	719
225°	*716*
270°	*716*

Institute for Applied Materials

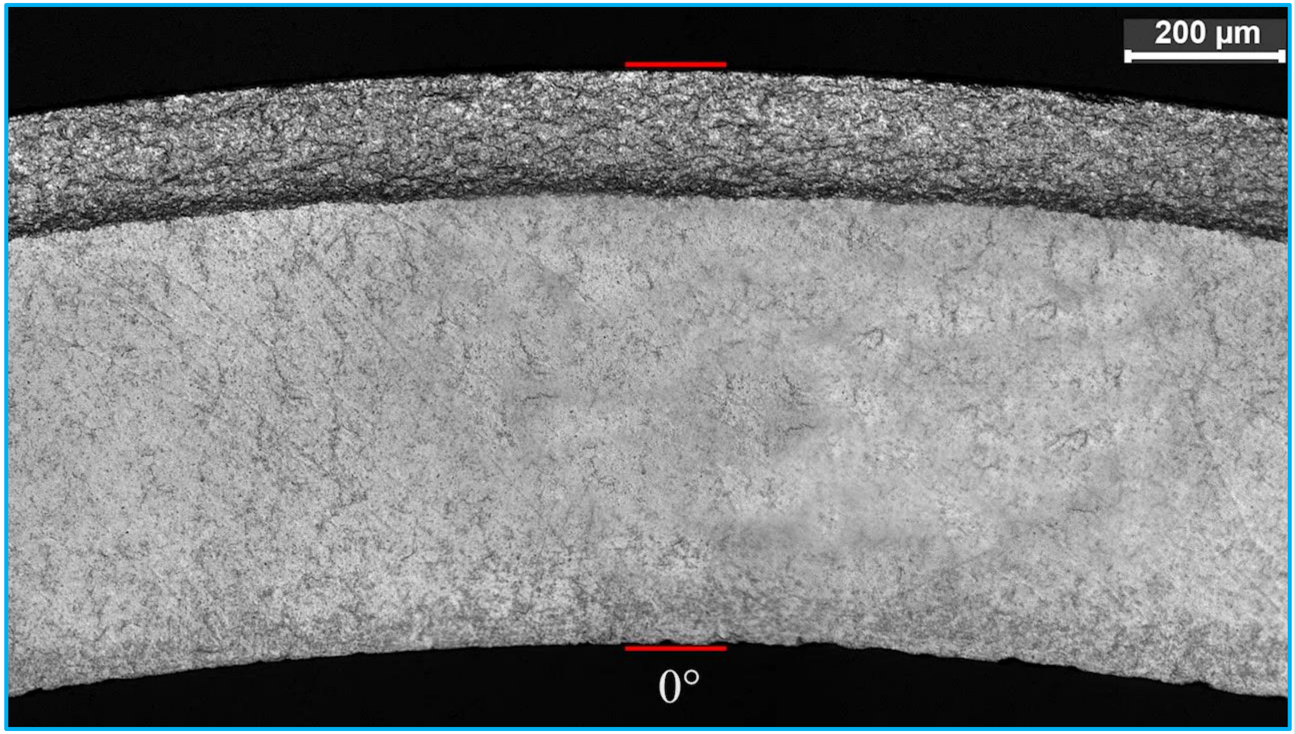
Post-test clad #1 (DX-D4, 146 bar, 300 wppm H), elevation 520 mm: metallographic observation of hydrides and cladding thinning

Link to movie:





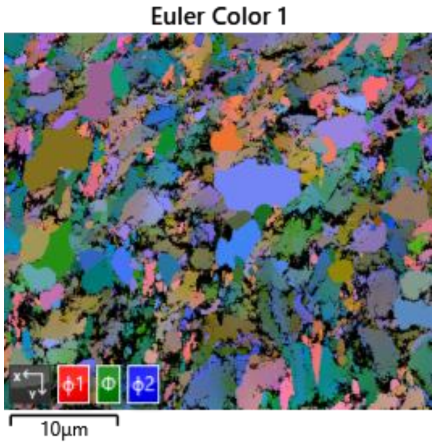
Eccentricity

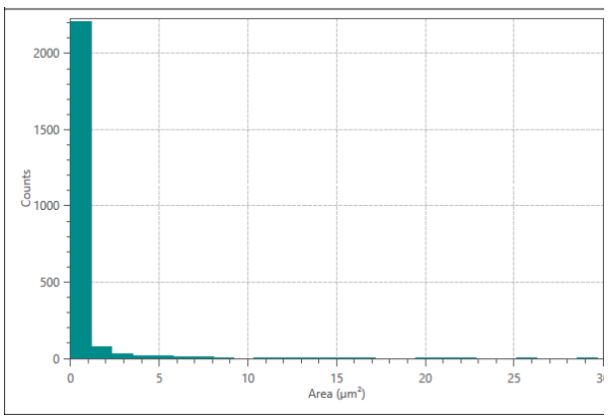


- **Circumferential homogeneity of hydrides appearance;**
- **Not very radial symmetry for the wall thinning:**

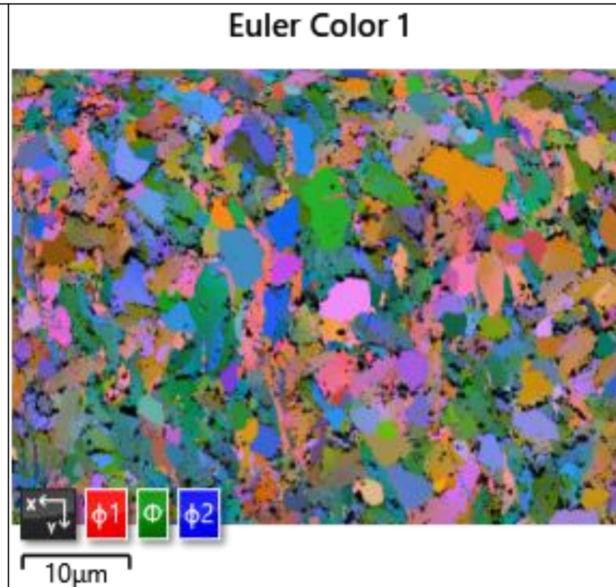
angle	wall thickness, µm
0°	716
90°	724
180°	720
225°	*712*
270°	*713*

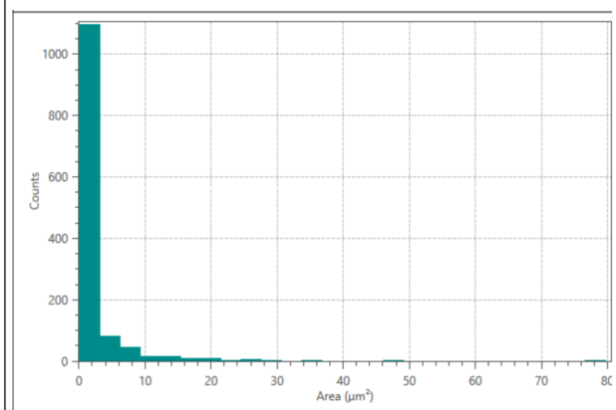
Grain growth during hydrogenation and following bundle test for Zry-4: fresh, hydrogenated, post-test samples



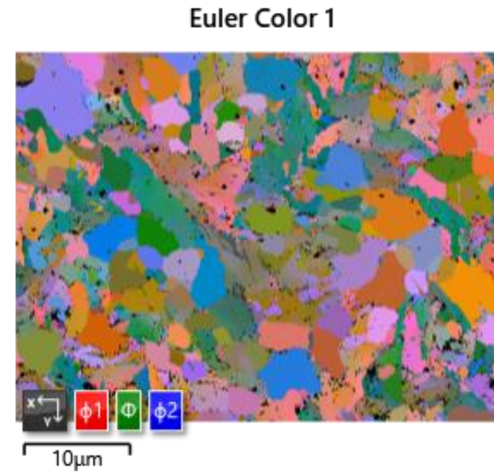


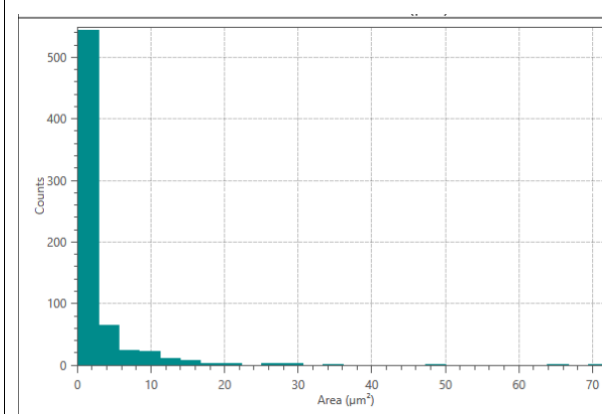
<u>as-received</u>: weight mean **5.74 ±1.67 µm²**, max 28.58 µm², grains 2387





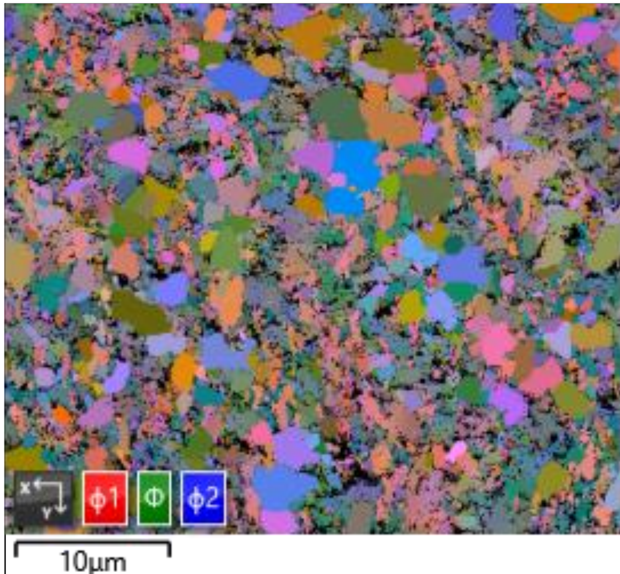
<u>300 wppm H</u>: weight mean **12.79±4.54 µm²**, max 76.68 µm², grains 1282

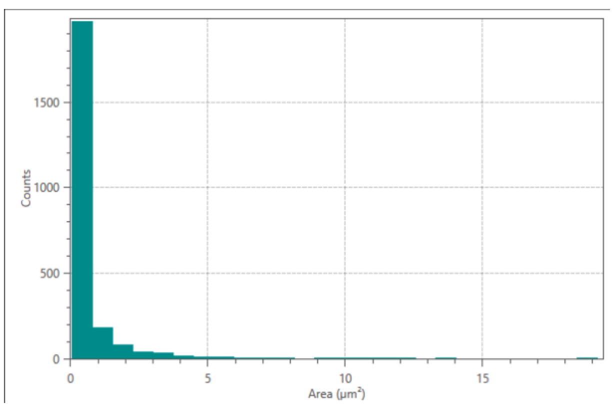




<u>bundle, 300 wppm H</u>: weight mean **16.02±5.66 µm²**, max 69.46 µm², grains 685

Grain growth during hydrogenation and following bundle test for opt. ZIRLO: fresh, hydrogenated, post-test samples



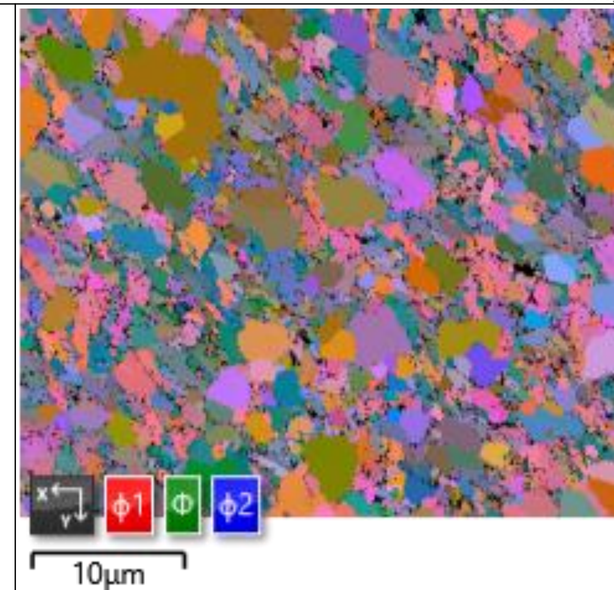


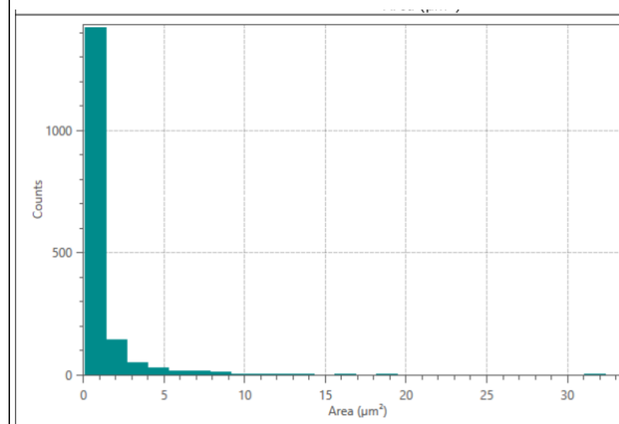
as-received: weight mean **2.92±1.16 µm²,** max 18.46 µm², grains 2363

not available

not available

300 wppm H





bundle, 300 wppm H: weight mean **4.33±1.72 µm²**, max 32.37 µm², grains 1696

Summary

- For the long-term SPIZWURZ bundle experiment, zirconium alloy tubes (opt. ZIRLO, Zry-4, DX-D4) were charged with hydrogen to 100 and 300 wppm in the special developed HOKI tube oven as homogeneously as possible along a length of 1.3 m.
- The hydrogenation was carried out at 450 °C by successively supplying fixed masses of hydrogen through the specially treated **inner** surface of claddings.
- After the hydrogen loading of the samples, the axial distribution of hydrogen was determined by laser scanning profilometry (calibrated by hot gas extraction).
- A long-term bundle test with 21 hydrogenated and pressurized cladding tubes began on 12.05.2023 and terminated on 17th January 2024. Two pressure set values were used: 106 and 146 bar. Due to small leaks in some of the rods, it was decided to maintain the pressure constant in all rods throughout the experiment (lasted 250 days) by periodically injecting the $Ar+O_2$ gas mixture refilled; oxygen was used for oxidation of the inner cladding surface to avoid hydrogen release from the cladding. The rod #18 was not pressurized and could be used as reference rod.
- The peak cladding temperature decreased in steps of ≈15 K from 400 to 165 °C (average cooling rate ≈0.9K/day). The maximal cooling rate during each temperature step was 6 K/h, step duration was about 10 h.

Summary (cont.)

- The post-test laser scanner measurements of the outer cladding diameter showed significant creep: radial deformation values are between 0.2 and 3.3% (diameter increase and the corresponding wall thinning).
- The largest creep of 3.3% was measured for opt. ZIRLO claddings hydrogenated to 300 wppm. The corresponding maximum creep value was 0.93% for Zry-4 and 1% for DX-D4.
- A clearly visible dependence of the degree of creep on the hydrogen concentration is observed for the opt. ZIRLO claddings: the creep of claddings hydrogenated to 300 ppm is 1.2-1.5 times higher than that of claddings hydrogenated to 100 ppm.
- A number of claddings show radially asymmetric wall thinning, which can be associated with the radial shift of the pellets from the central axis of the rod and the corresponding asymmetric heat supply along the circumference of the cladding.
- The metallographic investigations revealed a uniform distribution of hydrides throughout the entire cladding circumference for all three cladding types used. In the DX-D4 claddings, hydrogen primarily diffused toward the outer liner.
- The degree of hydride reorientation was significantly higher in the Zry-4 claddings compared to the opt. ZIRLO claddings.
- The difference in the behavior of the Zry-4 and opt. ZIRLO may be due, in part, to their different grain microstructures. Opt. ZIRLO claddings have a finer grain size than Zry-4. Moreover, although the temperatures during hydrogenation (450 °C) and the experiment itself (max. 400 °C) were relatively low, EBSD measurements showed grain growth from approx. 6 µm for the initial state to post-test 16 µm for Zry-4 (12 µm after hydrogenation), and from initial 3 µm to post-test 4.3 µm for opt. ZIRLO.

Thank you for your attention

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