Supplement of Saf. Nucl. Waste Disposal, 2, 25–25, 2023
https://doi.org/10.5194/sand-2-25-2023-supplement

Supplement of

Experimental investigations with neutron radiography of hydrogen effects by elastic stresses on cladding tubes under conditions similar to interim dry storage

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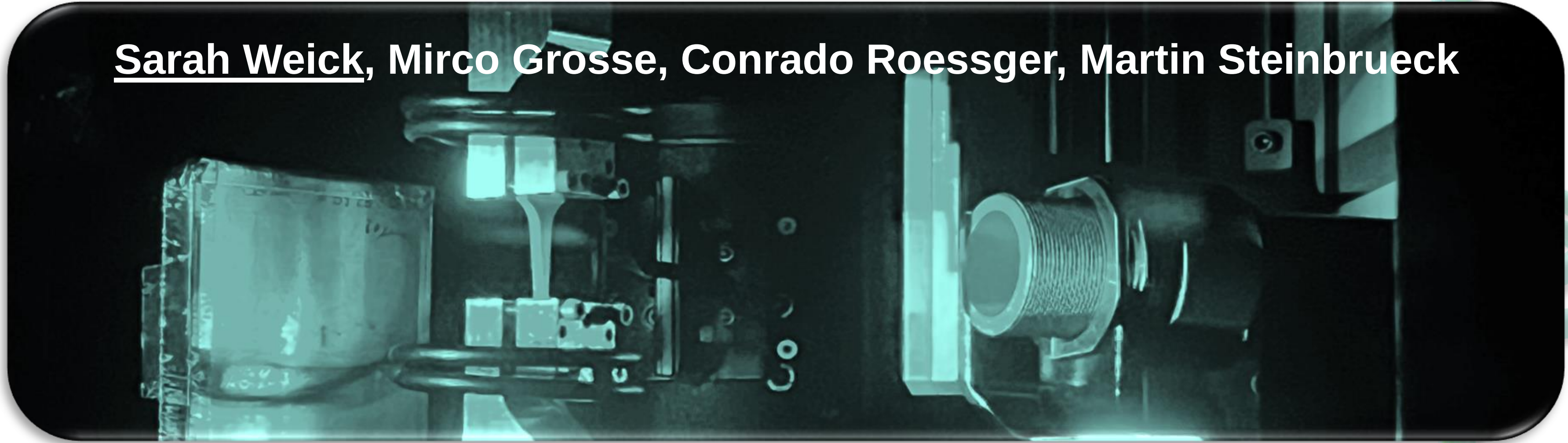

Interdisciplinary research symposium on the safety of nuclear disposal Practices (safeND)
13-15th September 2023, Berlin, Germany

Experimental investigations with neutron radiography of hydrogen effects by elastic stresses on cladding tubes under conditions similar to interim dry storage

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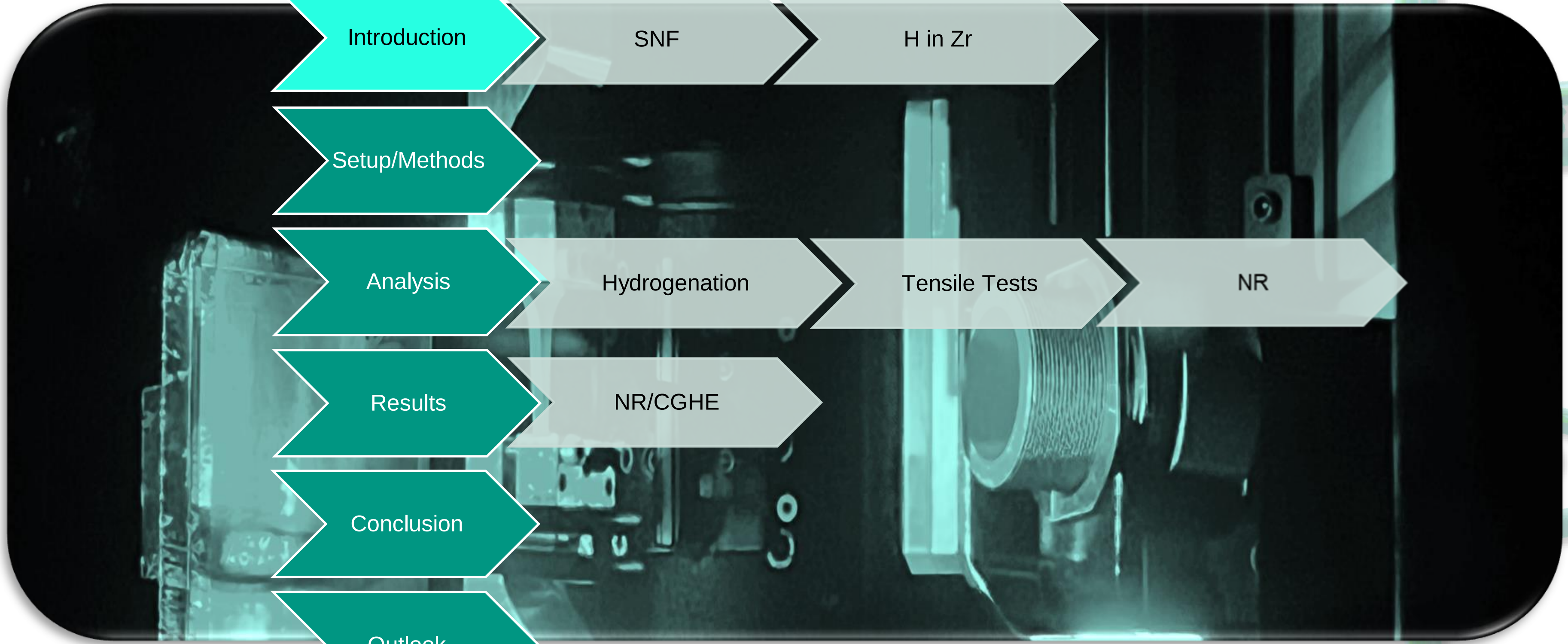
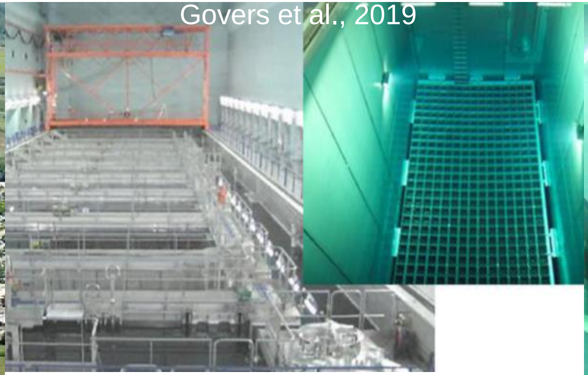
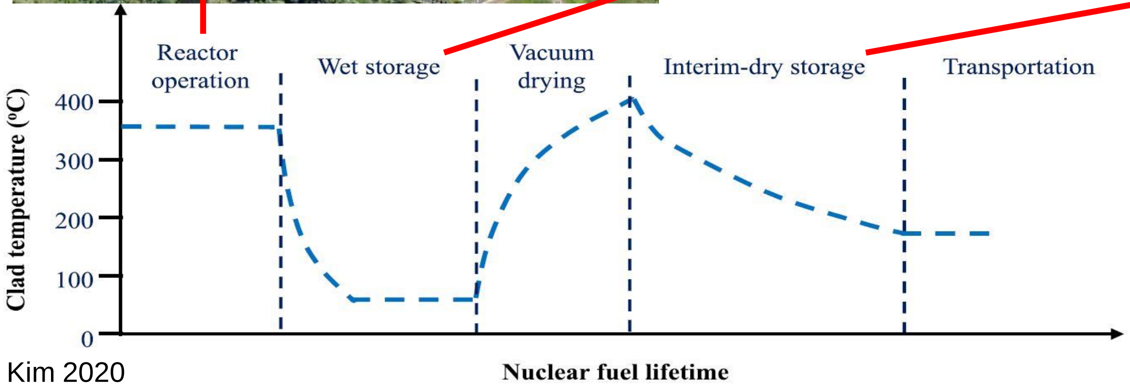


- Introduction
  - SNF
  - H in Zr
- Setup/Methods
- Analysis
  - Hydrogenation
  - Tensile Tests
  - NR
- Results
  - NR/CGHE
- Conclusion
- Outlook

Introduction – Spent Nuclear Fuel





Kim 2020

Introduction – Hydrogen embrittlement

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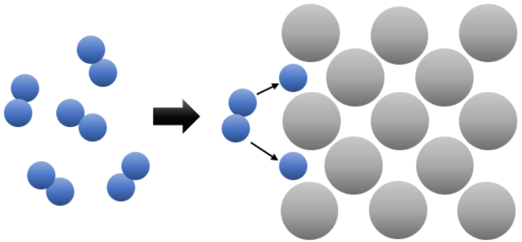
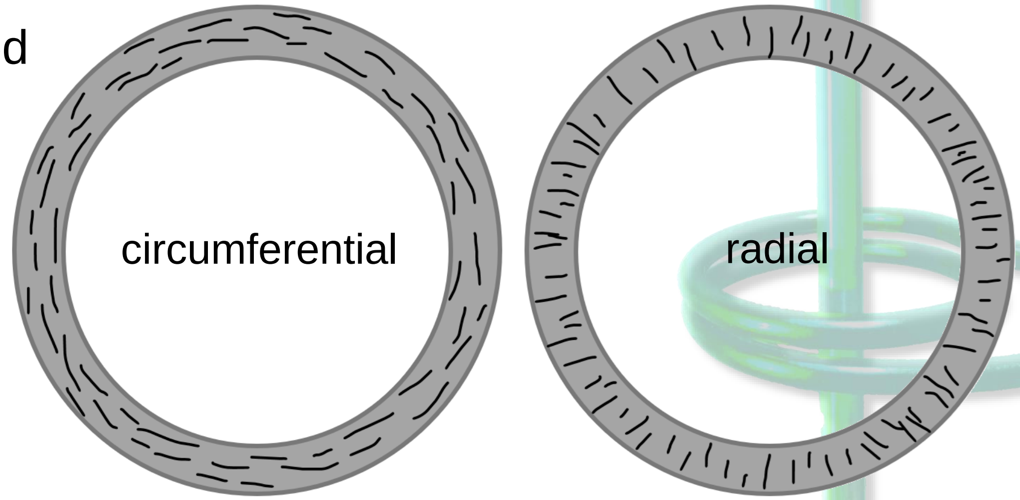
- H in cladding tubes
  - H uptake favoured by foreign atoms, alloying elements & textures
  - Mechanical strain & chemical activity → influence H diffusion

- Zr Hydrides
  - Circumferential or radially orientated
  - Reduce strength & ductility
  - Delayed hydride cracking (DHC)



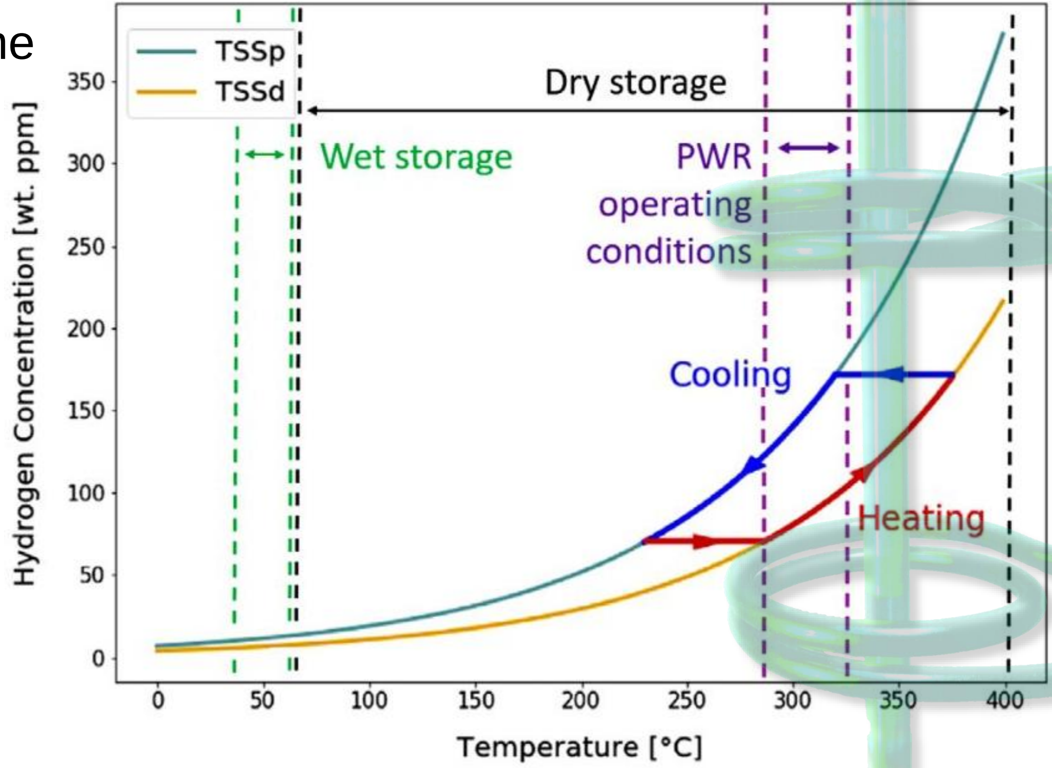


Introduction – H solubility

- H dissolution-precipitation scheme with modelled **TSS** (terminal solid solubility)

→ **TSSp**: precipitation terminal solubility limit

→ **TSSd**: dissolution terminal solubility limit



Kaufholz et al.2018, modified from Konarski 2021

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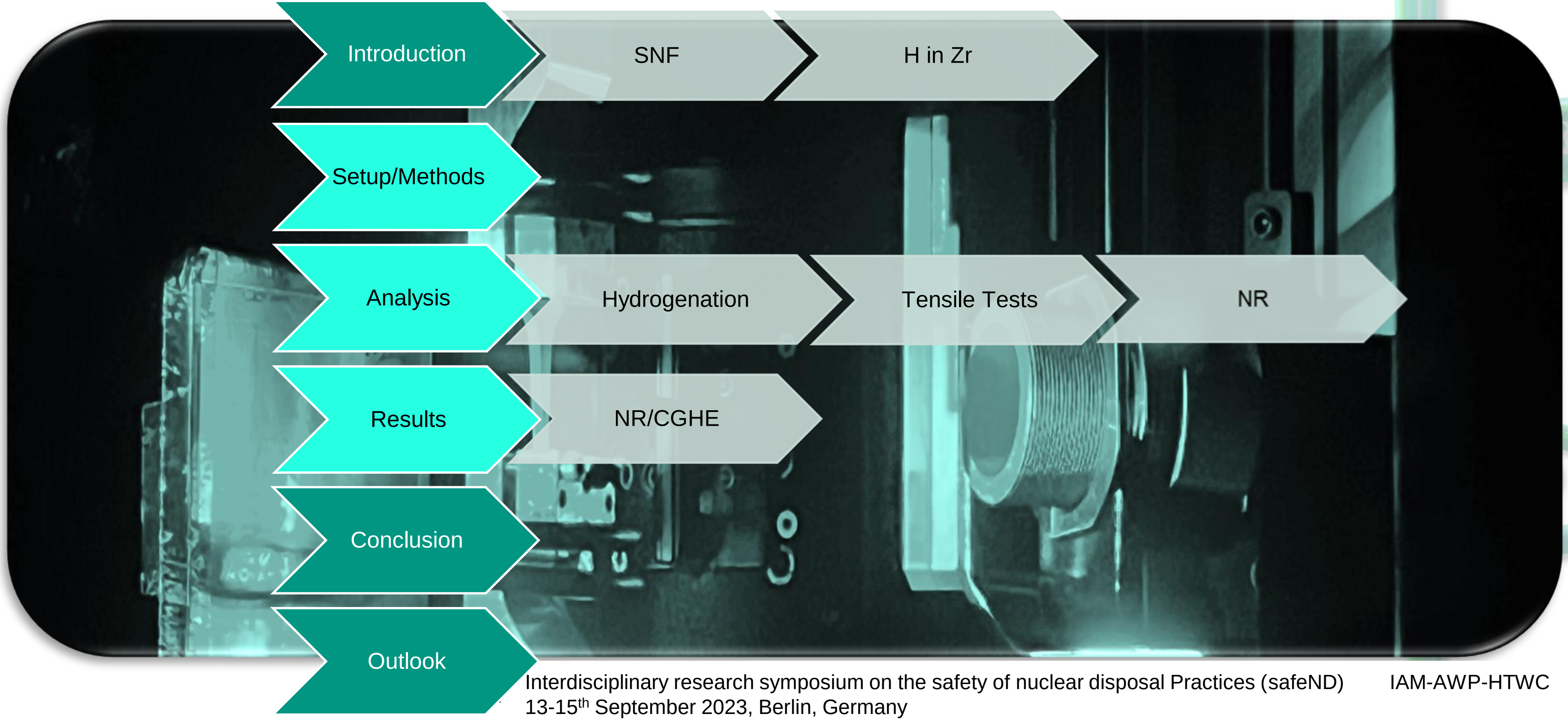
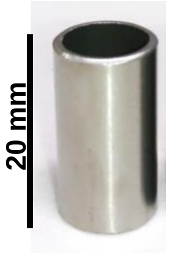
Experimental investigations with neutron radiography of hydrogen effects by elastic stresses on cladding tubes under conditions similar to interim dry storage

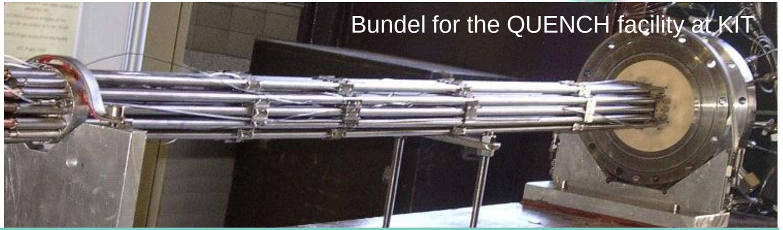


Experimental Setups

- Single Effect Experiments
  → Samples: cm - range
  → Influences of texture, grain size & elastic strain
  → Diffusion coefficients H

- QUENCH Bundle Test
  → Samples: m - range
  → Interim storage conditions (100-400°C; 70/96 MPa, 100/300 wt.ppm H)
  → Long-term 250 d

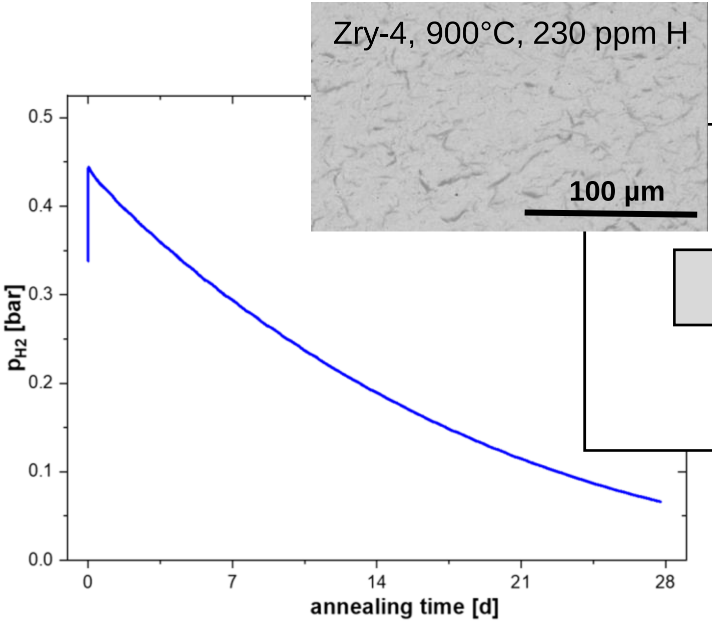


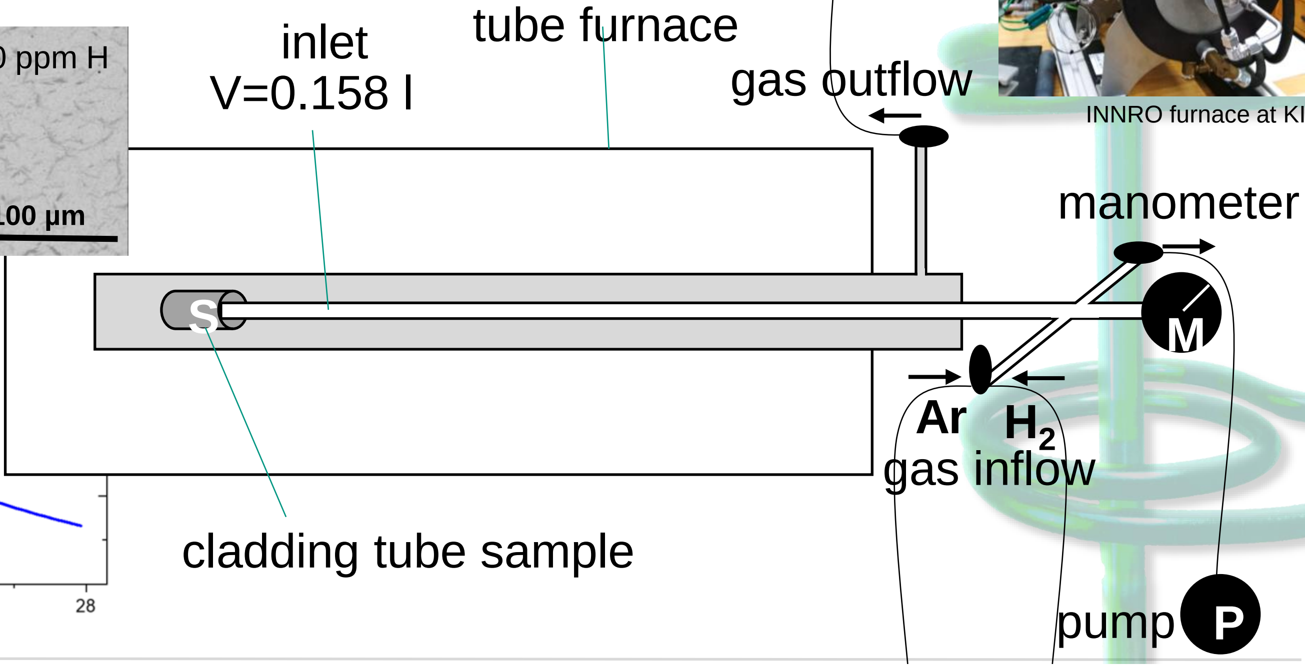

Bundel for the QUENCH facility at KIT

alloy	Sn [wt.%]	Fe [wt.%]	Cr [wt.%]	Nb [wt.%]	O [wt.%]
Zry-4, D4	1.3	0.2	0.1	-	0.13
Dx	0.5	0.5	0.2	-	0.14
Zirlo	0.9	0.1	-	0.9	-

Methods - Hydrogenation

- Annealing from gas phase with SICHA
- **SICHA** = Sieverts Chamber for Hydrogen Absorption

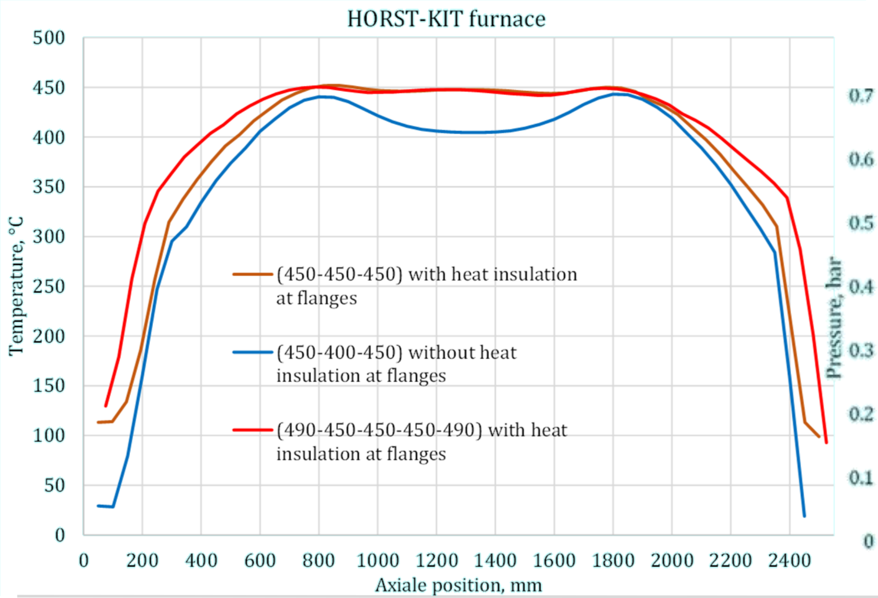


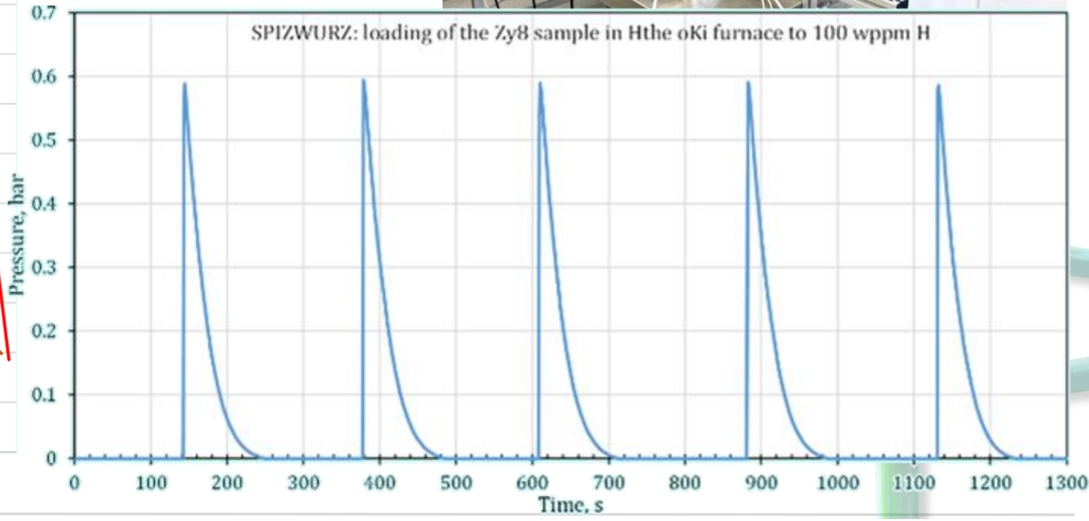


INNRO furnace at KIT

Methods - Hydrogenation

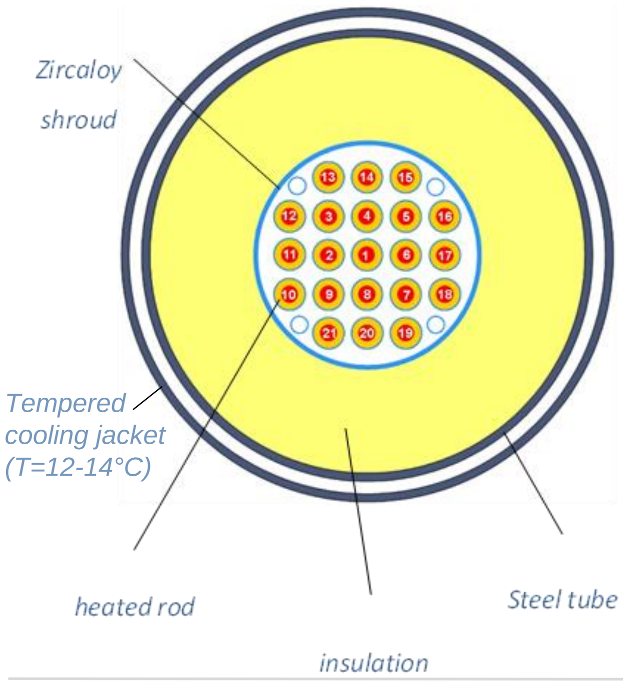
- 21 hydrided (100 & 300 wt.ppm) fuel rod simulators (l = 2.5 m)

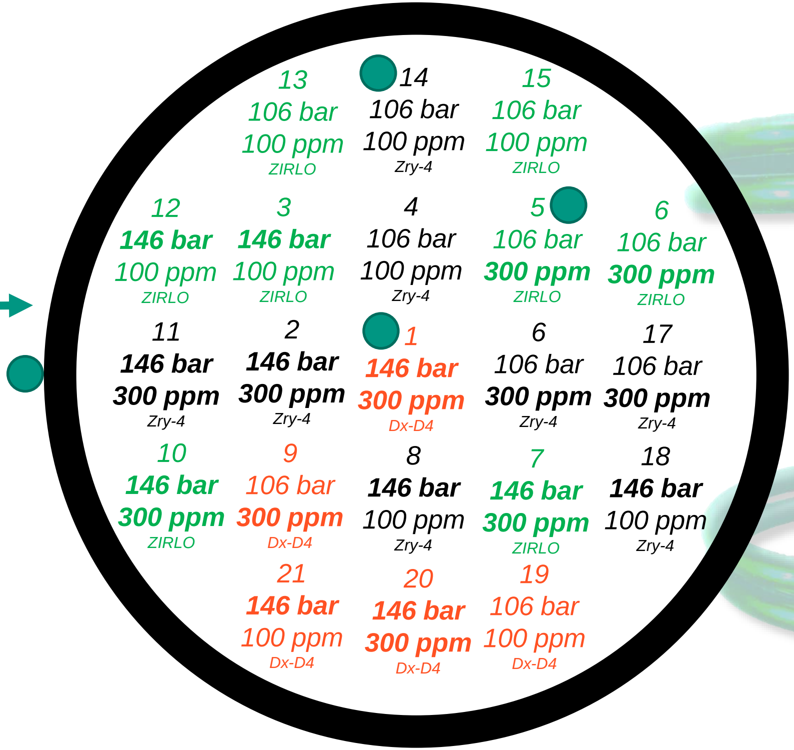




Methods – Dry Storage Simulation

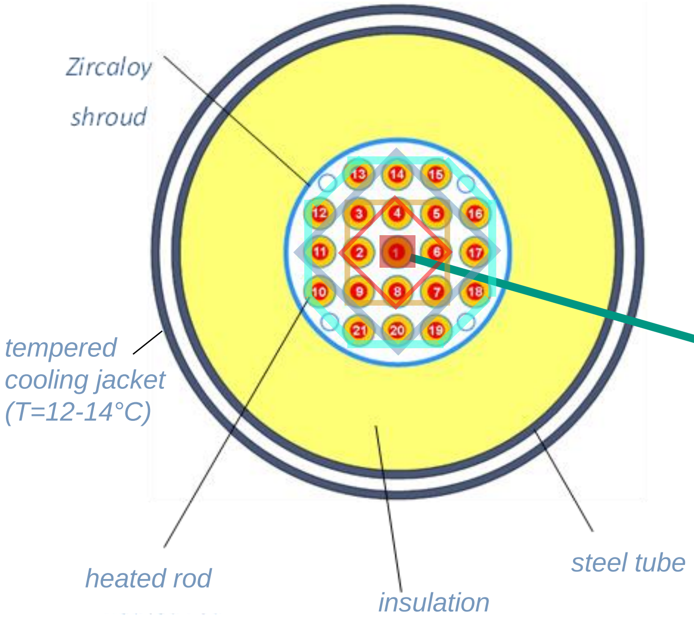
- 3 different cladding materials
- 2 different pressures

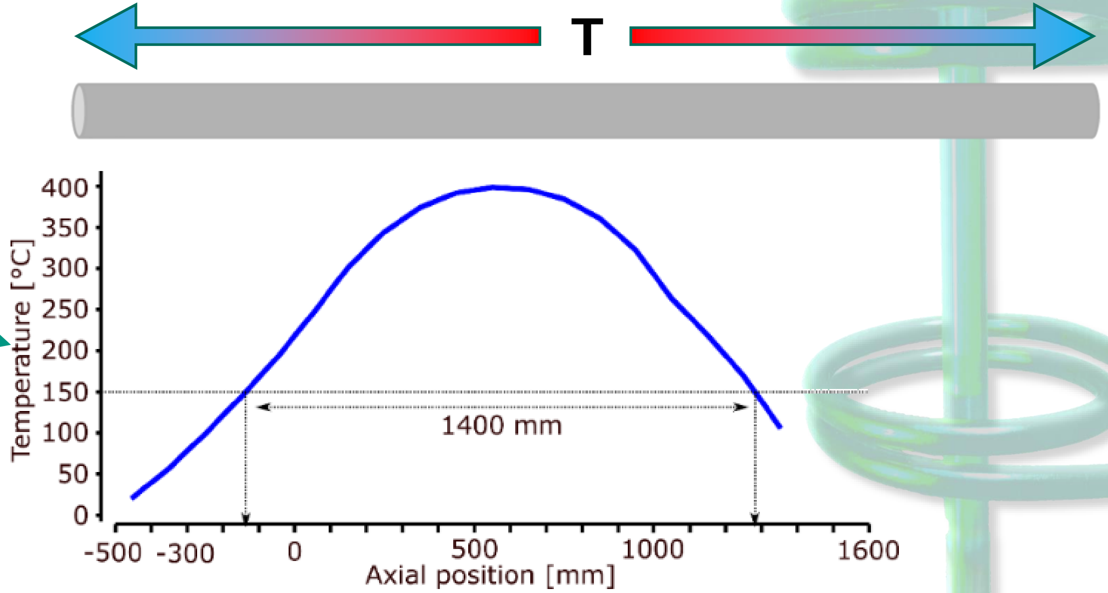




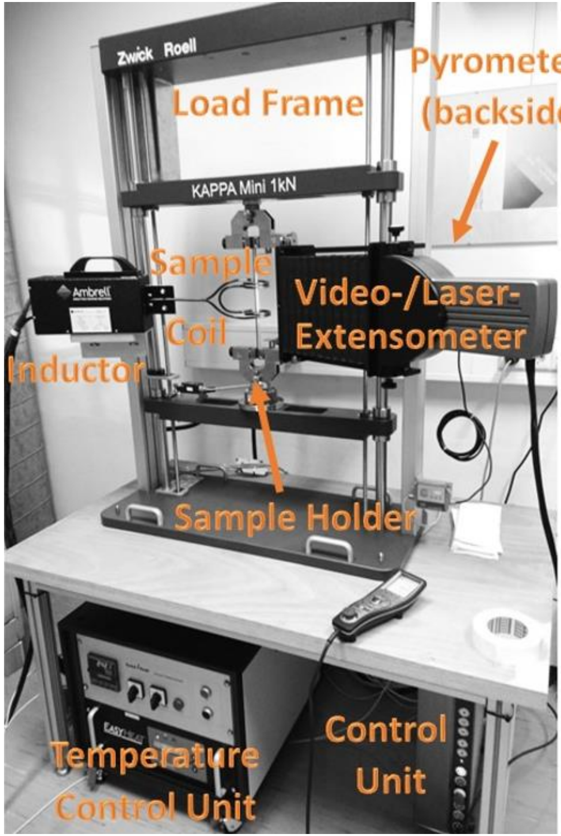
Methods – Dry Storage Simulation

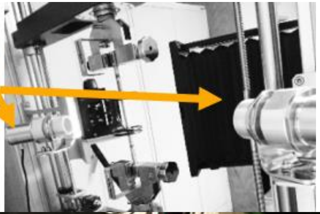
- Start-T: 400 °C (rod centre) - 100 °C (rod ends)
- Slow cooling: 7 K/w for 250 d
- Strain influences: thermal > plastic > cladding creep > elastic

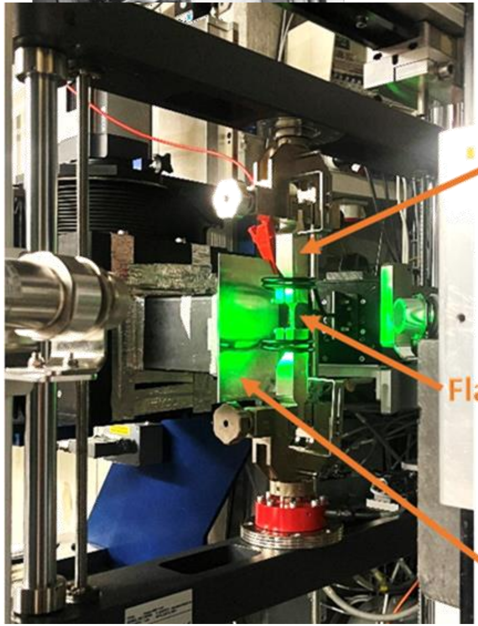




Methods – Tensile Tests





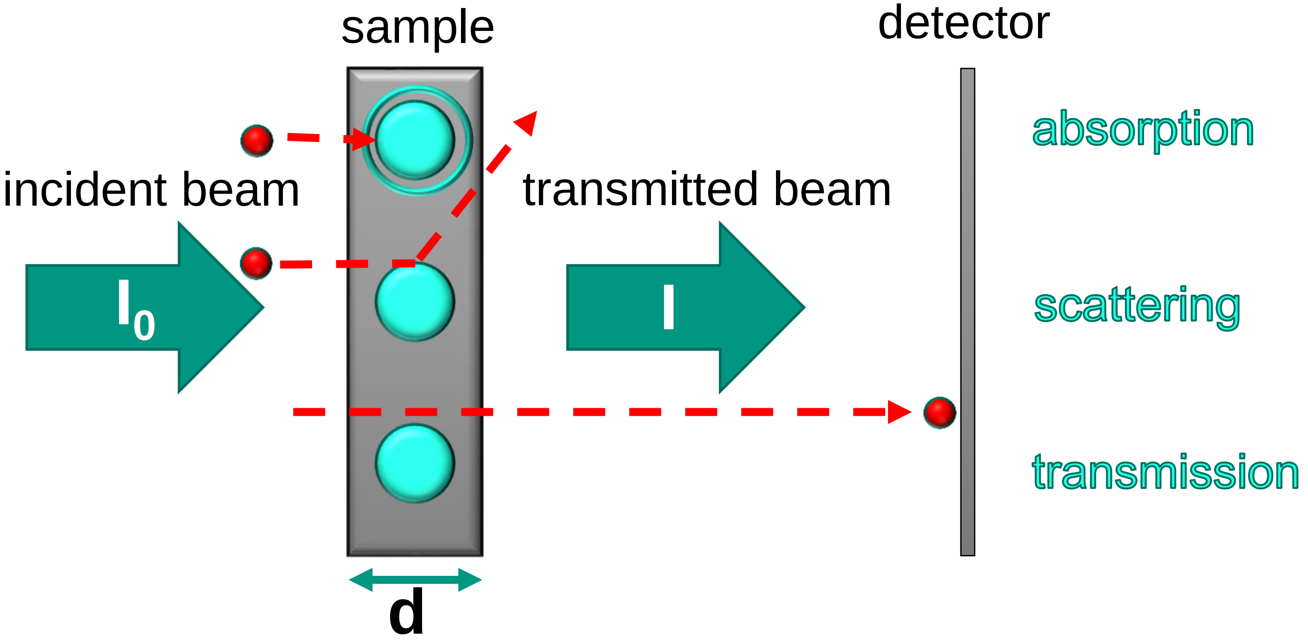


INCHAMEL = In-situ Neutron radiography CHAmber with MEchanical Load

- Tensile tests
- Inductive heating
- Contactless strain & temperature measurements
- Transportabel -> external neutron beamlines
- Iron free components; no long-term neutron activation

Analysis – Neutron Radiography

NR = Neutron Radiography



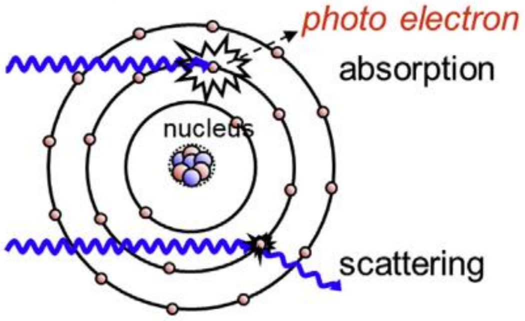
$$I = I_0 e^{-\sigma N d}$$

$$\Sigma = \sigma N$$

I: intensity
T: transmission
σ: microscopic neutron cross section
N: number density
d: sample thickness
Σ: macroscopic neutron cross section/ neutron attenuation coefficient

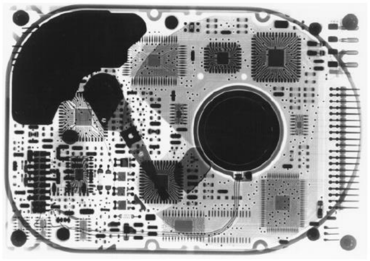
Analysis – Neutron Radiography

X-rays

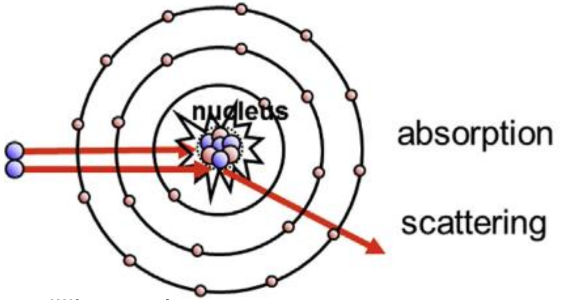


Attenuation coefficients with X-ray [cm?^{1}]

1a	2a	3b	4b	5b	6b	7b	8			1b	2b	3a	4a	5a	6a	7a	0
H 0.02																	He 0.02
Li 0.06	Be 0.22											B 0.28	C 0.27	N 0.11	O 0.16	F 0.14	Ne 0.17
Na 0.13	Mg 0.24											Al 0.38	Si 0.33	P 0.25	S 0.30	Cl 0.23	Ar 0.20
K 0.14	Ca 0.26	Sc 0.48	Ti 0.73	V 1.04	Cr 1.29	Mn 1.32	Fe 1.57	Co 1.78	Ni 1.96	Cu 1.97	Zn 1.64	Ga 1.42	Ge 1.33	As 1.50	Se 1.23	Br 0.90	Kr 0.73
Rb 0.47	Sr 0.86	Y 1.61	Zr 2.47	Nb 3.43	Mo 4.29	Tc 5.06	Ru 5.71	Rh 6.08	Pd 6.13	Ag 5.67	Cd 4.84	In 4.31	Sn 3.98	Sb 4.28	Te 4.06	I 3.45	Xe 2.53
Cs 1.42	Ba 2.73	La 5.04	Hf 19.70	Ta 25.47	W 30.49	Re 34.47	Os 37.92	Ir 39.01	Pt 38.61	Au 35.94	Hg 25.88	Tl 23.23	Pb 22.81	Bi 20.28	Po 20.22	At	Rn 9.77
Fr	Ra 11.80	Ac 24.47	Rf	Ha													
Lanthanides	Ce 5.79	Pr 6.23	Nd 6.46	Pm 7.33	Sm 7.68	Eu 5.66	Gd 8.69	Tb 9.46	Dy 10.17	Ho 10.91	Er 11.70	Tm 12.49	Yb 9.32	Lu 14.07			
Actinides	Th 28.95	Pa 39.65	U 49.08	Np	Pu	Am	Cm	Bk	Vf	Es	Fm	Md	No	Lr x-ray			

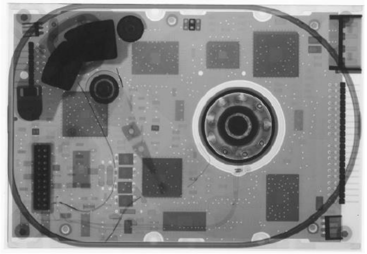


neutrons



Attenuation coefficients with neutrons [cm?^{1}]

1a	2a	3b	4b	5b	6b	7b	8			1b	2b	3a	4a	5a	6a	7a	0
H 3.44																	He 0.02
Li 3.30	Be 0.79											B 101.60	C 0.56	N 0.43	O 0.17	F 0.20	Ne 0.10
Na 0.09	Mg 0.15											Al 0.10	Si 0.11	P 0.12	S 0.06	Cl 1.33	Ar 0.03
K 0.06	Ca 0.08	Sc 2.00	Ti 0.60	V 0.72	Cr 0.54	Mn 1.21	Fe 1.19	Co 3.92	Ni 2.05	Cu 1.07	Zn 0.35	Ga 0.49	Ge 0.47	As 0.67	Se 0.73	Br 0.24	Kr 0.61
Rb 0.08	Sr 0.14	Y 0.27	Zr 0.29	Nb 0.40	Mo 0.52	Tc 1.76	Ru 0.58	Rh 10.88	Pd 0.78	Ag 4.04	Cd 115.11	In 7.58	Sn 0.21	Sb 0.30	Te 0.25	I 0.23	Xe 0.43
Cs 0.29	Ba 0.07	La 0.52	Hf 4.99	Ta 1.49	W 1.47	Re 6.85	Os 2.24	Ir 30.46	Pt 1.46	Au 6.23	Hg 16.21	Tl 0.47	Pb 0.38	Bi 0.27	Po	At	Rn
Fr	Ra 0.34	Ac	Rf	Ha													
*Lanthanides	Ce 0.14	Pr 0.41	Nd 1.87	Pm 5.72	Sm 171.47	Eu 94.58	Gd 1479.04	Tb 0.93	Dy 32.42	Ho 2.25	Er 5.48	Tm 3.53	Yb 1.40	Lu 2.75			
**Actinides	Th 0.59	Pa 8.46	U 0.82	Np 9.80	Pu 50.20	Am 2.86	Cm	Bk	Cf	Es	Fm	Md	No	Lr neut.			



Kardijilov et al. 2019

Lehmann 2012

Analysis – Neutron Radiography

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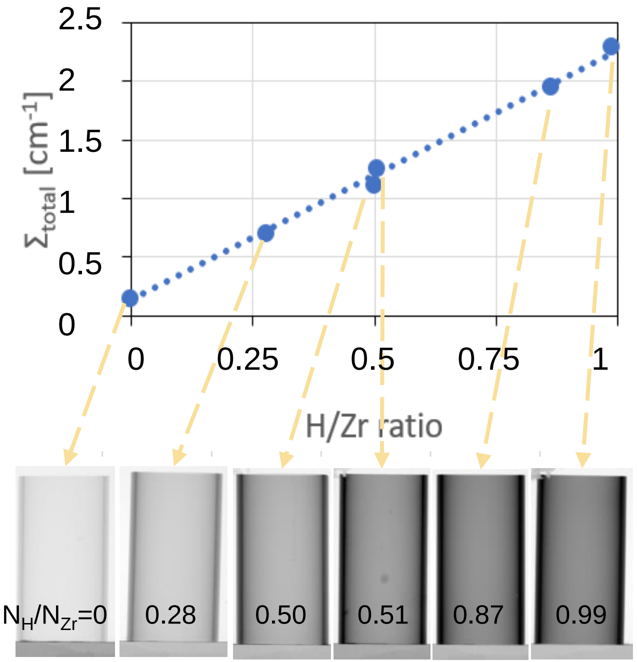
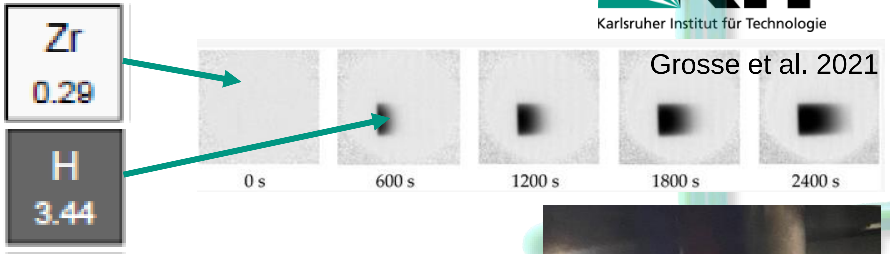
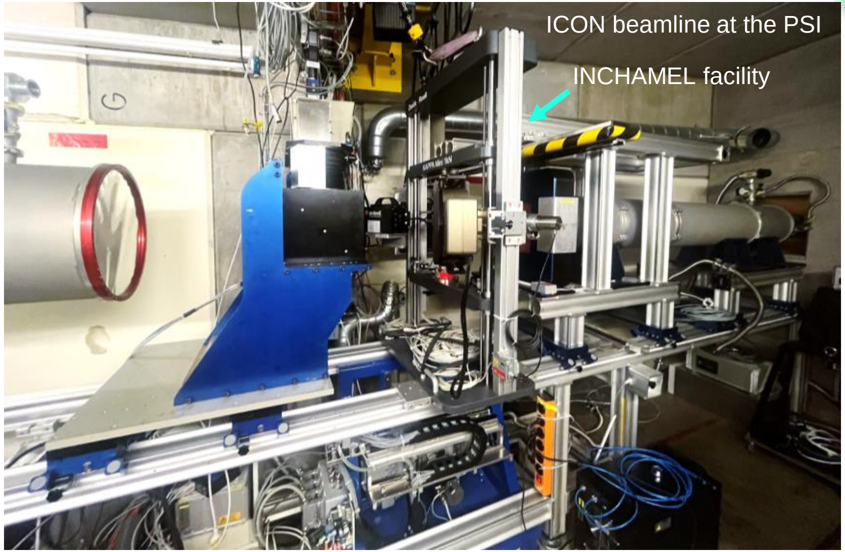
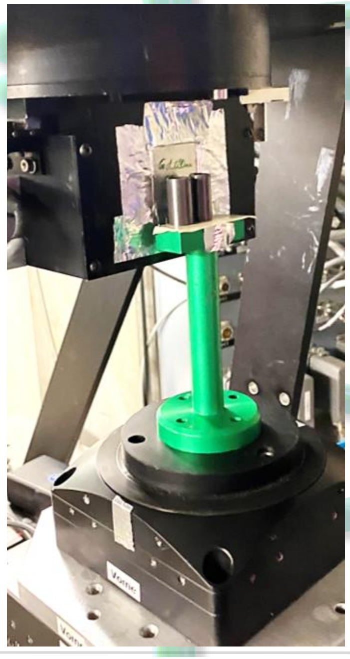
$$T = \frac{I}{I_0}$$







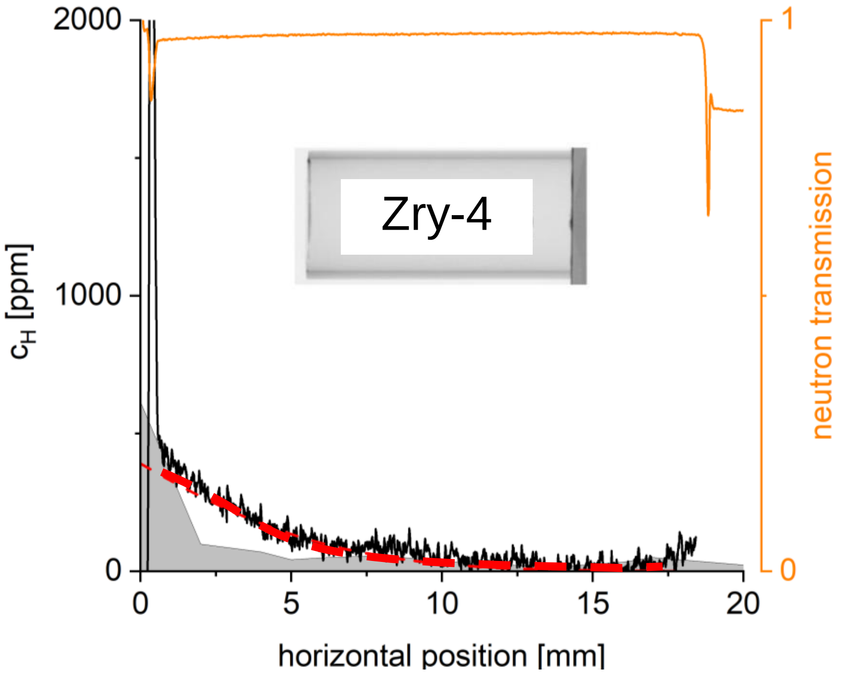


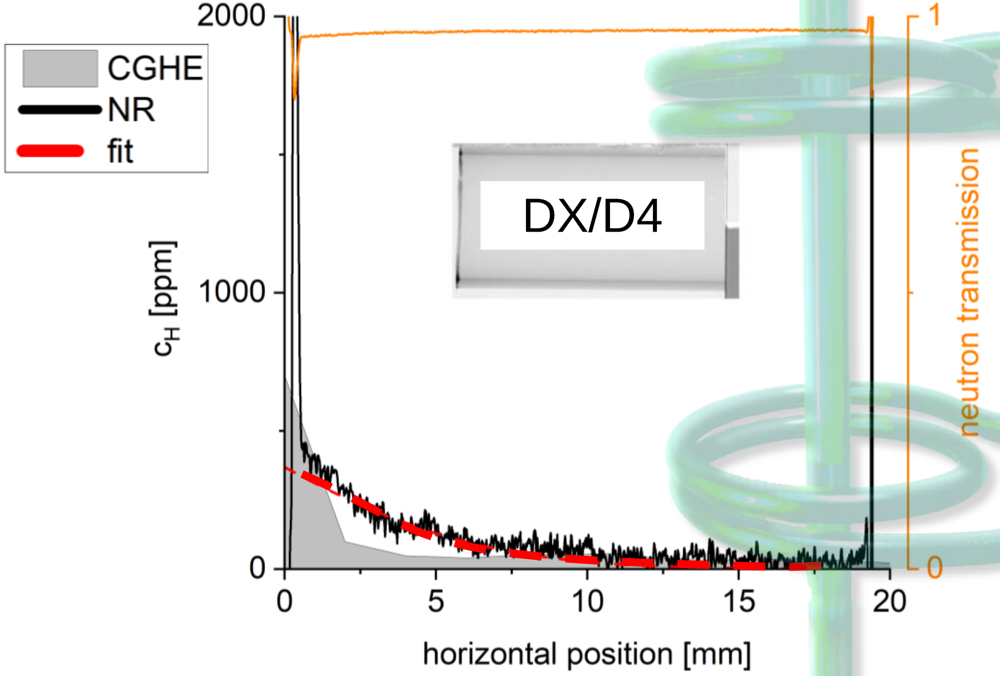
Sarah Weick

Results – Diffusion coefficients

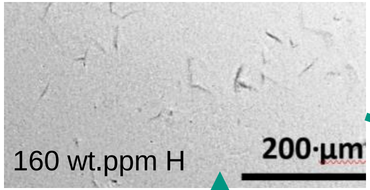
- ZrH_2 powder, Ar, 400°C, 3h

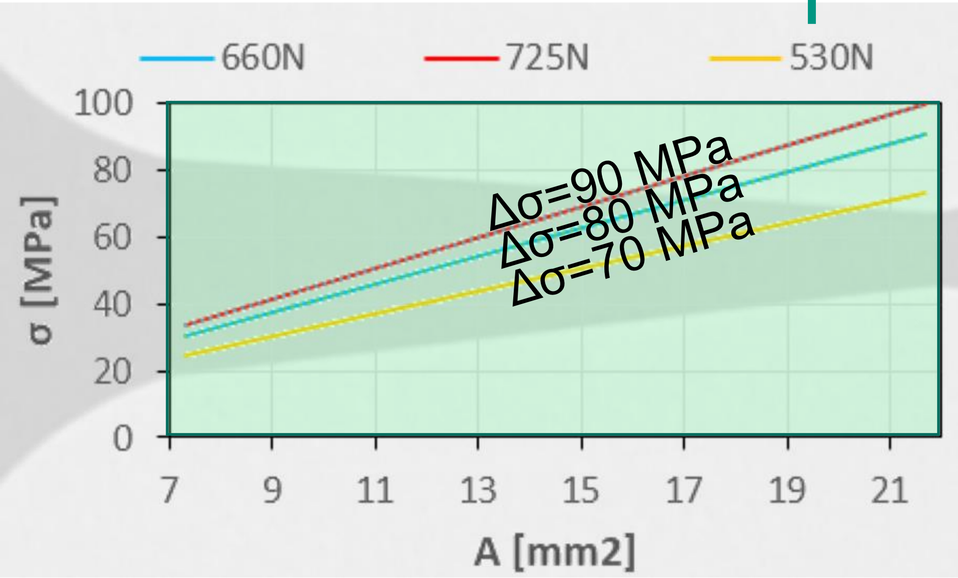
$$c(x,t) = c_0\left(1 - erf\left(\frac{x}{2\sqrt{Dt}}\right)\right) + c_i$$

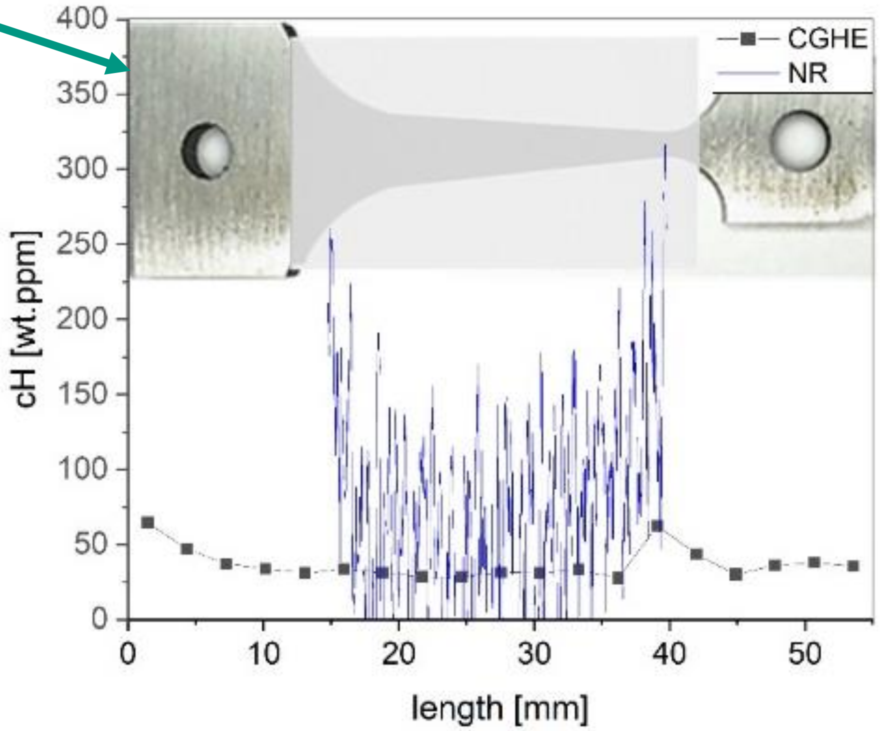




Results – Tensile Tests







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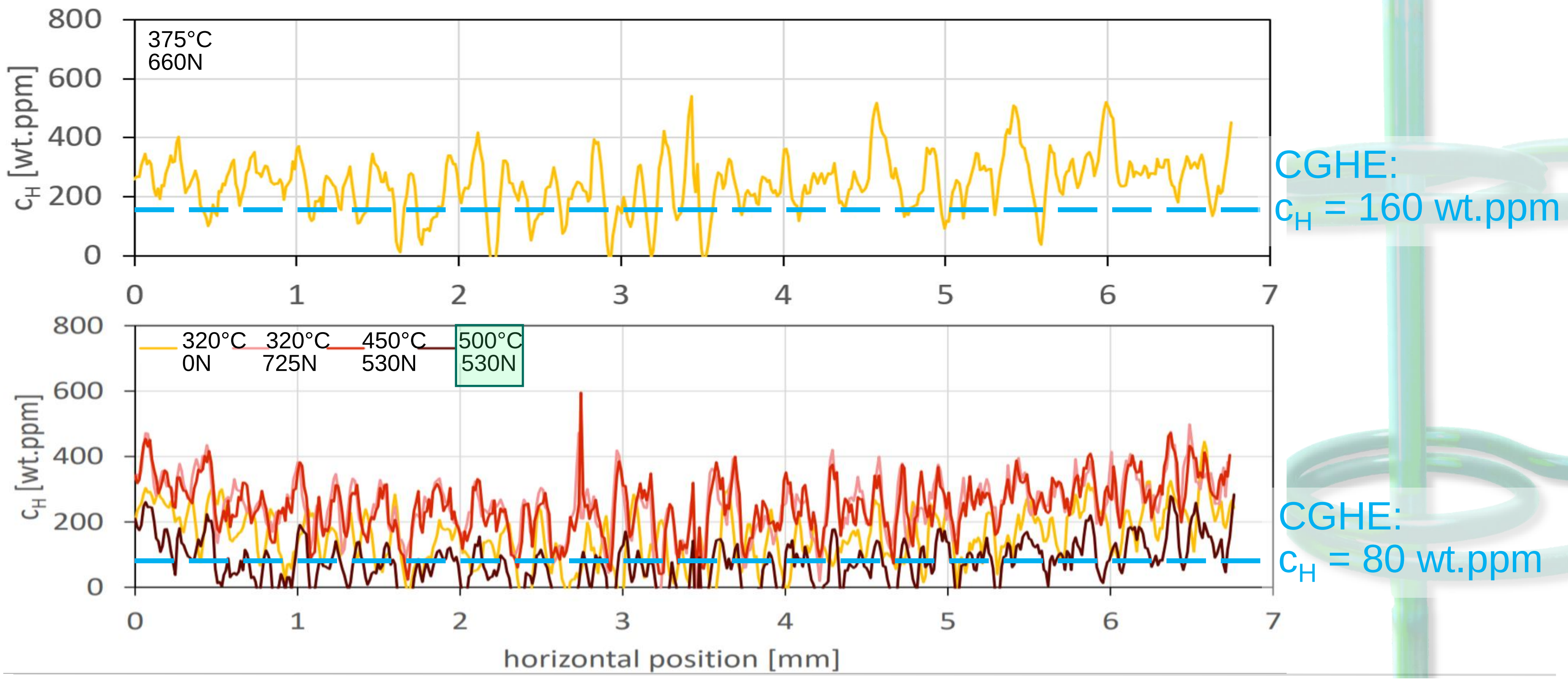

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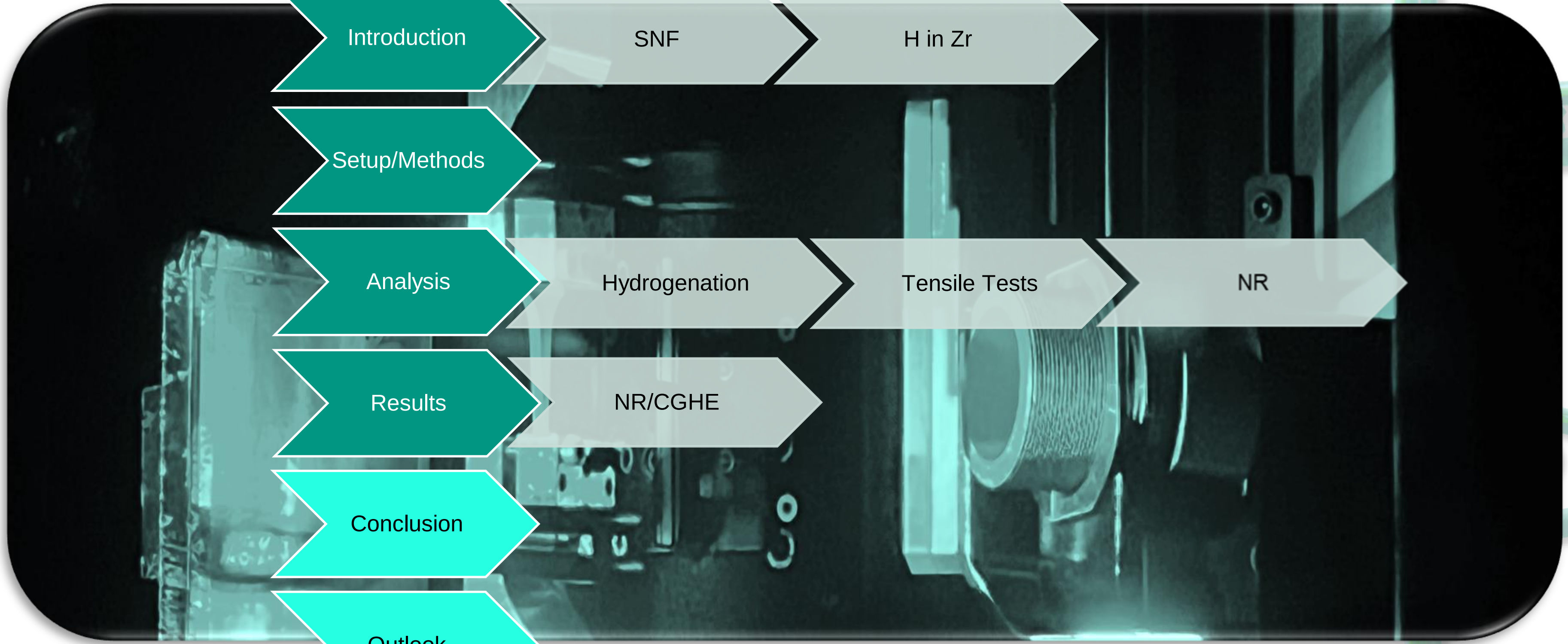

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Conclusion

- How to determine hydrogen effects by elastic stresses on cladding tubes under conditions similar to interim dry storage?

→ With single effect experiments in combination with a tensile testing machine observed by NR (ex-/in-situ)

→ With a long-term experiment imitating the slow cooling process and dry storage relevant p-T-conditions

→ With modelling

→ With SNF samples (pellet-cladding interactions)

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Outlook

- Single Effect Experiments

→ NR ex-situ with the INCHAMEL facility for investigations of the stress influence on H diffusion and solubility in Zr for longer time scales (weeks)

→ NR in-situ with the INCHAMEL facility for investigations of local stress induced hydrogen dissolution and precipitation processes

- QUENCH bundle test

→ NR ex-situ investigations of the simulation rods under the various p-T-conditions at the end of the test

→ metallographic investigations of the hydride precipitation direction (stress influenced) at the end of the test

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Acknowledgements

- the SPIZWURZ project (FKZ 1501609B) is funded by the Federal Ministry for the Environment, Nature Conservation, Nuclear Safety and Consumer Protection (BMUV)
- the INCHAMEL facility was funded by the HOVER program of the Helmholtz Association
- PSI for providing beamtime & Anders Kaestner, David Mannes & Matteo Busi for their assistance during the measurements /analysis
- GRS team
- chemical analysis group of Thomas Bergfeldt (IAM-AWP)
- QUENCH team & colleagues at KIT

Thank you!