Journal Pre-proof

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PII: S1738-5733(23)00435-7

DOI: https://doi.org/10.1016/j.net.2023.09.027

Reference: NET 2598

To appear in: *Nuclear Engineering and Technology*

Received Date: 24 July 2023

Revised Date: 13 September 2023

Accepted Date: 18 September 2023

[ICAPP 2023] The SPIZWURZ project – experimental investigations and modeling of the behavior of hydrogen in zirconium alloys under long-term dry storage conditions

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Abstract – In order to investigate the occurring processes during long-term dry storage of spent fuel assemblies, a joined project called SPIZWURZ, between the Karlsruhe Institute of Technology and the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), was started. Aim of the SPIZWURZ project is the determination and quantification of the influence of texture and elastic strain on diffusion and solubility of hydrogen in three different zirconium alloys used in western Europe during a long-term cooling transient (1 K/d) starting at 400 °C. The strain in the cladding of an irradiated spent fuel rod shall be measured. Models predicting the formation of radial oriented hydrides will be validated, improved, and implemented in the GRS fuel rod performance code TESPA-ROD. This paper describes the SPIZWURZ project and already obtained first results.

1. INTRODUCTION

The German waste management concept foresees the direct disposal of spent nuclear fuel (SNF) in a deep geological repository, available by 2050, at the best. SNF is stored in dry interim storage facilities using dual-purpose casks until then. Licenses for both, casks and facilities, will expire forty years after loading of the first assembly into the cask and emplacement of the first cask into the interim storage facility. However, considering the delay in the site selection process so far and the estimated duration for exploration, construction and commissioning of a final repository for high-level waste, a prolonged dry interim storage of SNF is inevitable [1]. Therefore, integrity of the cladding is of utter importance regarding the ultimately conditioning of the fuel assemblies for final disposal and safety assessments have to be performed evaluating the state of the fuel assemblies in the casks. In this framework, the long-term degradation of the toughness of the cladding materials by redistribution and reorientation of the hydrides in the claddings is a main topic. Because of the very long time scale - several decades - only modeling can provide the needed information about the state of the fuel rods. However, the modelling over such long time periods requires precise knowledge about the influence of parameters like temperature distribution, elastic stress, texture, or grain size on the solubility and the diffusivity of the hydrogen in the zirconium based cladding alloys.

The SPIZWURZ project is a collaborative project between the German institutions GRS and two sub-institutes of the Karlsruhe Institute of Technology (KIT). SPIZWURZ is the acronym for the German title “Spannungsinduzierte Wasserstoffumlagerung während Langzeit-Zwischen-lagerung” (stress induced hydrogen re-distribution during long-term intermediate storage). The cooperation seeks to determine parameters relevant for hydrogen diffusion and solubility and to validate and improve existing fuel performance codes by experimental data. Several single effect tests are combined with a large scale and long-term simulation test. The paper gives an update about experimental and modelling results obtained up to now and informs about further actions.

2. THE SPIZWURZ PROJECT

The SPIZWURZ project [2] includes several experiments at the KIT and a model development by the GRS. The results gained from these experiments will be used for code validations and verifications. Especially the slow cool-down of hydrogenated cladding samples as performed in the bundle experiment in the QUENCH facility will generate data of the hydrogen diffusion on macroscopic scales. The SPIZWURZ project can be divided into four topics:

i) A long-term test of a fuel rod simulator bundle (21 electrically heated fuel rod simulators with a length of about 2.2 m) at the KIT-QUENCH facility with a starting temperature of 400°C at the hottest position and a duration of 240 days with a cooling rate of 1 K/day.

ii) Separate effect tests with small samples to measure the diffusion rates at various temperatures in dependence on the texture and the mechanical stress state.

iii) Determination of the elastic and plastic strain of an irradiated spent fuel cladding tube.

iv) Validation and improvements of models based on the experimental data and implementation in fuel rod performance codes.

In the following, details to the topics are given.

2.1. The bundle test

The simulation bundle consists of 21 unirradiated fuel rod simulators with a length of 2.2 m. Three types of cladding materials are used: Zircaloy-4 (Zry-4), Optimized ZIRLO as well as D4/Dx Duplex consisting of a Zry-4 bulk (D4) and an outside liner with a reduced tin concentration (Dx). The chemical compositions are given in Table 1.

Each rod simulator is filled with ZrO_2 annular pellets, and electrically heated by one tungsten and two molybdenum heaters (connected in series) with diameters of 4.6 and 8.6 mm, respectively. The heater power of each rod can be measured individually. Furthermore, each rod is equipped with a pressure supply and the inner gas pressure can be adjusted individually. During the experiments, the fuel rod simulator bundle will be surrounded by a zirconium shroud. The volume between the rods will be filled with argon gas. To assure that the temperature of the outer cooling jacket remains independent from ambient temperature fluctuations, it will be cooled by fluid circulated with constant temperature of 10°C through the jacket gap.

A scheme of the bundle cross section is given in Fig. 1. The claddings were pre-hydrated to concentrations of 100 and 300 wt.ppm. The 2.2 m long tubes are hydrogenated from the inner side in a furnace dedicated for this application (Fig. 2). The cladding tubes are heated in this furnace to 450°C in an oxygen atmosphere outside. After a pre-annealing to dissolve the inner oxide layer, the tube is evacuated. Then, 55 cm^3 hydrogen gas is injected into the tube. To receive the aimed hydrogen concentrations of 100 or 300 wt.ppm, the tube must be refilled with hydrogen several times as shown in Fig. 3. If the aimed amount of hydrogen is absorbed, the annealing continued until the next morning (about 18 h) to get a homogeneous hydrogen concentration in Zry-4 and Opt. ZIRLO. In the Duplex material, the hydrogen becomes enriched in the liner and particularly at the liner/bulk interface. Additionally, an external oxide layer is produced preventing hydrogen release through the outer surface.

After their hydrogenation, the cladding tubes are instrumented and mounted to the bundle. The leak tightness is proofed. The long-term test starts by heating to the initial peak cladding temperature of 400°C. Once this temperature is reached, the cooling experiment with a cooling rate of 1 K/d for 8 months will start. A temperature gradient will occur in radial and in axial direction of the bundle. The radial difference at the beginning of the test is about 50 K between at the axial hottest position. A schematic temperature distribution at the beginning of the experiment for a central fuel rod (hottest rod) is shown in Fig. 4. The consequences of the temperature gradients are variations of the maximal temperatures between the different rods as well of their cooling rates, which is more or less proportional to the temperatures.

Material	Sn	Nb	Fe	Cr
Zry-4, D4	1.5	0.01	0.21	0.1
ZIRLO	1.0	1.0	0.11	0.005
Dx	0.5	0.01	0.5	0.2

Table 1 Concentrations of alloying elements of the cladding tube materials in wt.%, (Zr balance)

The chosen concentrations are below and above the solubility of hydrogen in the zirconium alloys (≈ 200 wppm) at the start temperature of 400 °C of the test. It means that in the claddings with the higher concentration circumferential hydrides formed whereas all hydrogen is in solution in the claddings loaded with the lower concentration. The rod simulators consist of heater rods surrounded by zirconia ring pellets. The gap is filled with krypton with inner pressures of 106 and 145 bar, corresponding with hoop stresses in the cladding of 68 and 93 MPa, respectively. The rods will be pressurized to their respective pressure (at 400°C) at the beginning of the experiment, the rods will then be sealed, and the pressure will reduce naturally with decreasing temperature during the experiment.

The variations in the cladding materials, hydrogen concentrations, inner pressures, initial temperatures and cooling rates results in the conduction of numerous single rod tests on laboratory scale in the QUENCH-SPIZWURZ bundle test. Tab. 2 gives an overview about the different test conditions of the materials used.

The test was started at May 16 2023 and will be conducted until January 11 2024. The test is accompanied by pre-test calculations made by GRS using the software packages ATHLET-CD and TESPA-ROD.

As results of the bundle test, detailed information about the axial distribution of hydrides in fuel rod claddings after cool-down are expected. Furthermore, the location as well as orientation of hydrides depending on the axial position will be analyzed.

Material	p_{min}, c_{min}	p_{min}, c_{max}	p_{max}, c_{min}	p_{max}, c_{max}
Zry-4	1, 1	1, 1	1, 1	1, 1
ZIRLO	2	1, 1	1, 1	1, 1
Dx/D4	1	1	1	1, 1

Table 2 Overview about the test conditions for the used alloys (red color indicates inner rod simulators with higher temperature, blue color peripheral tubes with lower temperature, hoop stresses: p_{min} = 68 MPa, p_{max} = 93 MPa, hydrogen concentrations: c_{min} = 100 ppm, c_{max} = 300 ppm)

2.2. Separate Effect Tests

Separate effect tests on laboratory scale are performed to determine the influence of structure parameters and the mechanical load on the solubility and the diffusion rate of hydrogen in nuclear fuel cladding alloys. This information is necessary for the development of models describing the fuel assembly performance during decades of dry storage.

The local amount and the distribution of the hydrogen were measured by means of neutron radiography [3].

The investigations comprise

- hydrogen diffusion measurements at different directions of zirconium single crystals,
- hydrogen diffusion measurements of nuclear fuel cladding tube segments with the required strong texture (c axis in hoop direction),
- hydrogen solubility and diffusion measurements of elastic strained zirconium.

As examples, Figs. 5 and 6 show first results of the investigations of the diffusion in axial and in hoop direction of cladding tubes, respectively. For the investigations in axial direction, 2 cm long tube segments standing in a ZrH_2 powder bed were annealed at temperatures between 325 and 450°C for different time scales. Fig. 5 compares the neutron radiographs of a Zry-4 and of a D4/Dx sample after 3 h annealing at 400°C, the measured neutron transmissions and the hydrogen concentration profiles calculated from the transmissions. Both materials show a similar behaviour. At the left edges (the positions where the samples were in contact with the ZrH_2 powder bed), the dark grey value indicates a very high hydrogen concentration. Obviously, a welding with the ZrH2 powder occurred. The similar hydrogen profiles give hints that for the DUPLEX material, the hydrogen transport in axial direction occurs mainly by diffusion in the D4 bulk and the Dx liner has less influence on the hydrogen diffusion.

In order to investigate the hydrogen diffusion in hoop direction of the cladding tube (corresponding with the crystallographic c direction), 4 mm long tube segments were cut. Then the oxide layer that formed at room temperature in air was removed at the cut by grinding. This procedure allowed the hydrogen to penetrate the sample only at the cut area. Fig. 6 shows the neutron radiograph of the Zry-4 sample annealed for 1 h at 450°C in axial direction.

The yellow bands mark the positions and directions where the neutron intensity distributions were measured beginning and ending at the cut position at the upper right side. The width of the bands indicates the integration range for the intensity measurements.

From these intensity distributions, the transmissions and the hydrogen concentration distributions along these bands are given in Fig. 6.

The radiograph underlines that the hydrogen does not only penetrates the sample at the crack flanks but also the whole part where the sample was in contact with the ZrH_2 powder bed. This problem was solved by hydrogen loading in hoop direction by Zircaloy wedge inserted into the cuts. The wedges were pre-loaded with hydrogen to very high concentrations of about 20000 wt.ppm. Analyses of the transfer of hydrogen from the wedges by means of hot gas extraction demonstrate the feasibility of this local hydrogen loading.

The concentration distribution along the hoop direction is visible in the diagram given in Fig. 6. The hydrogen uptake starts if the oxide layer that formed at room temperature in air dissolves by diffusion of the oxygen into the bulk. The onset of the hydrogen uptake start is not known. Therefore, no quantitative information as diffusion coefficients can be extracted from the results. In-situ neutron radiography experiments are planned. In such experiments, the beginning of the hydrogen uptake can be recorded.

Whereas the diffusion experiments with zirconium single crystals did not start yet, a special facility called INCHAMEL [4], was constructed for the in-situ neutron radiography investigations of the dependence of the diffusivity and the solubility of hydrogen in zirconium alloys on elastic stress and strain. It is a modified mechanical test facility. In order to modulate the neutron beam as less as possible, heating, temperature and strain measurement are contactless by inductive heating using Helmholtz coils, by pyrometers and video or laser extensometers.

The ICHAMEL facility is shown in Fig. 7. The special components for contactless heating and measurements of temperature and strain are marked. To reduce any material activation by scattered neutrons during the radiography measurements, ones made of aluminum alloys substitute steel components if possible.

The commissioning of the facility was conducted at the neutron radiography beamline ICON at the Swiss Neutron Source SINQ (Paul Scherrer Institute, Switzerland). First tests were successful. They demonstrate the feasibility of this facility. The analysis of the data is still ongoing and will be published elsewhere.

2.3. Determination of elastic and plastic strain of an irradiated spent fuel cladding tube

In spent nuclear fuel (SNF) with high average burnup, the initial gap between the fuel pellet and the cladding tube closes as a consequence of the in-reactor swelling of the pellet and the creep behavior of the cladding. A further volume expansion of the SNF pellet during dry storage is expected to be caused by the combined effects of a fuel lattice parameter expansion and a helium precipitation in the fuel matrix [5, 6].

These phenomena lead to chemical and mechanical interactions between the pellet and the cladding. In order to prevent cladding failure during dry storage of SNF, stress/strain criteria are set by law e.g. the maximum hoop stress during storage may not exceed 120 MPa and the maximum hoop strain may not exceed 1% in Germany [7].

The aim of the present study is to determine the stress state of irradiated fuel rod samples after about 30 years of storage with respect to the mechanical interaction between pellet and cladding. For this purpose, a Zry-4 cladding tube specimen in contact with UO_2 fuel was prepared and the diameter before and after removal of the fuel was measured. The difference in the cladding tube diameter is proportional to the elastic strain and allows to calculate the circumferential stress using the Young's modulus. Moreover, the experimental results were compared to ab initio calculated and modeled data.

The investigated cladding specimen in contact with UO_2 fuel was cut from a fuel rod segment that was irradiated in the pressurized water reactor of the Gösgen nuclear power plant in Switzerland. Irradiation was performed during four cycles until 1989. Since then, the fuel rod segment was stored. At the end of reactor operation, the fuel rod achieved an average burn-up of 50.4 GWd/tHM.

Dry cutting of the 8 mm length sample was performed slowly to limit heating of the sample using a low speed saw equipped with a diamond wafering blade (Isomet 11-1180, Buehler Ltd.). The external diameter of the segment was measured by means of a laser scan micrometer (Mitutoyo LSM-503) possessing a linearity of ± 1.0 µm and a repeatability of ± 0.11 µm. The laser scan micrometer shown in Fig. 8 was modified for remote handling and installed in a hot cell. Before use, the laser scan micrometer was calibrated using standards. Measurements were performed at five longitudinal and eight radial positions by moving the segment in a custom-made sample holder, designed to facilitate diameter measurements in a reproducible position as can be seen in Fig. 8. The specimen was scratched to obtain a zero position and moved longitudinally, by means of a micrometer screw; at steps of 1 µm. Up to six independent measurements were carried out at each position before and after removal of the SNF and a mean value calculated.

The SNF was removed by means of alkaline digestion in an autoclave using a mixture of $(NH_4)_2CO_3$ / H_2O_2 at room temperature within five days. After drying of the defueled cladding specimen, the sample was again characterized using the laser scan micrometer.

The selected Zircaloy-4 cladding sample in contact with UO_2 fuel was successfully characterized at five longitudinal and eight radial positions, using the laser scan micrometer, before and after removal of the SNF (see Fig. 9).

Preliminary results show an oval shaped cross-section with differences between the maximum and the minimum measured diameters of about 30 to 50 µm before removal of the SNF. Measurements performed after removal of the SNF reveal an average variation of the diameter of 3.3 µm. The accuracy of the measurements determined using standard bodies is 1.7 µm. However, the uncertainty in the diameter due to positioning of the pellet during the measurements is about 3.1 µm whereas the standard deviation of the multiple measurements per position is 2.7 µm.

Preliminary ab initio calculations show, the variation induced by the SNF removal is ≤ 20 µm. It is possible to affirm that variation of the diameter due to defueling is within the experimental uncertainties; however, it cannot be larger than about 6 µm (variation of the diameter + effect of uncertainty of positioning).

2.4. Validation and improvements of models based on the experimental date and implementation in fuel rod performance codes

As part of the SPIZWURZ project GRS proposes a benchmark [8] to compare existing fuel rod simulation codes against the cool-down bundle test (see section “The bundle test”) regarding the prediction of hydrogen diffusion within nuclear fuel rod claddings [9]. The experiment is designed to study hydrogen behavior in fuel rod claddings within boundary conditions close to the dry storage of spent nuclear fuel. The benchmark includes three phases:

- The preparation phase is dedicated to code developers using the simulated boundary conditions to test their hydrogen models in advance.
- Task I is the blind benchmark, where basic design features, the cladding pre-treatment and the temperature conditions of the bundle experiment are provided.
- Task II is conducted after the end of the experiment, when the analysis of the hydrogen species and distribution is completed.

The benchmark is still open for participation and will comprise a set of typical parameters used for analyzing the cladding behavior and is intended as support for the experiment as well as a fundament for future code model development [9].
For the simulation, the fuel rod simulator (Fig. 10 a)) with the thermo-couples in the positions shown in Fig. 10 b) will be divided in nine axial zones (Fig. 10 c)). Once the experiment is terminated each fuel rod will be analyzed. The axial distribution of hydrogen within the cladding will be determined by neutron radiography and hot vacuum gas extraction at a minimum of five axial positions. The radial distribution as well as the orientation of the hydrides will be determined using cross-sectional micrographs at selected positions. This data will be available for the third phase of the Benchmark.
In the following, the cladding's thermo-mechanical behavior is predicted by the GRS fuel rod code TESPA-ROD based on the information available for the preparation phase. This includes the experimental setup and the thermo-hydraulic boundary conditions calculated by ATHLET-CD. The behavior of the hydrogen within the cladding is presented for Rod 8 (see Fig. 1), a Zry-4 rod with high pressure and low hydrogen content. The initial hydrogen distribution of the fuel rod is assumed to be a constant value of 100 wppm over the full axial length. TESPA-ROD predicts that diffusion processes of concentration- and temperature-driven diffusion will result in a rearrangement of hydrogen within the cladding tube. Due to the gradients in temperature, the temperature-driven diffusion (Soret-effect) leads to an accumulation of hydrogen in the colder parts of the fuel rod, which is shown in the Fig. 10. In total, approximately 6 wt.ppm hydrogen diffuses from the center to both directions.
It becomes apparent, that the local maxima and minimum hydrogen concentration is reached after approximately 120 days. Currently point the temperature decreased below 150°C and is too low to allow significant diffusion. Furthermore, diffusion is only visible in a 1400 mm wide central region of the fuel rod, since the temperature beyond is at the start of the experiment already lower than 150°C (Fig. 4, Fig. 11).
In every axial zone, the hydrogen behavior can be described as a function of dissolved hydrogen (in solid solution) or precipitated hydrogen (as radial or circumferential hydrides) with time [10].
Fig. 12 shows the evolution of hydrogen behavior over time in the central axial zone 5 (Fig. 10 c)). The violet line shows the total amount of hydrogen, which starts at 100 wt.ppm and decreases over time. The red line shows the hydrogen in solid solution, which starts to decrease after approximately 50 days due to reduction of temperature. Due to slow cooling, the simulation predicts a concentration of dissolved hydrogen close to the terminal solid solubility for dissolution (TSSd). The green graph is the sum of the precipitated hydrides. Initially, hydrides with radial and circumferential orientation precipitate simultaneously. TESPA-ROD uses threshold stresses for the prediction of hydride reorientation. Below 90 MPa circumferential and above 110 MPa radial precipitation is assumed. Between theses thresholds, the orientation ratio of the hydrides varies based on linear interpolation. The latter case is shown between 50 days and 95 days, the share of both orientations increases (Fig. 12). Since the rod temperature decreases the inner gas pressure decreases as well, which causes a reduction of the cladding hoop stress below the lower threshold. After 95 days, the share of radial hydrides stays constant, and only circumferential hydrides are formed.

3. CONCLUSIONS

The SPIZWURZ project includes several experiments at KIT as well as model development at GRS. The results gained from these experiments will be used for code validation and verification. Especially the slow cool-down of hydrided cladding samples as performed in the bundle experiment in the QUENCH facility will generate hydrogen diffusion data on macroscopic scale.

The preparation of the large-scale bundle test including the hydrogen loading of the cladding tubes is finished and the test has started in May 2023.

For the separate effect tests, the equipment and procedures are installed and optimized. First results of the separate effect test were obtained.

Measurements involving irradiated spent fuel have shown that the elastic responds of the diameter due to defueling is within the experimental uncertainties, however, it cannot be larger than about 6 µm.

The current simulation shows the dynamics of hydride diffusion and precipitation during a slow cool-down of gas pressured cladding tubes in the QUENCH facility. The diffusion is forced by the temperature difference (Soret effect) over the cladding length and retards at axial zones with low temperatures. The total effect of the axial diffusion is with approximately 6 wt.ppm rather small. Within the central axial zone, the hydrogen precipitates after temperature drops below the terminal solid solubility for hydrogen. Since the cladding hoop stresses are higher than the lower threshold for reorientation, radial and circumferential hydrides precipitate first. After falling below this threshold due to the decrease of gas pressure, only circumferential hydrides precipitate.

The proposed benchmark will be organized along side the three major steps in the SPIZWURZ bundle experiment, which are described above. With this benchmark, we aim to simulate the hydrogen diffusion processes in the fuel rod claddings during the bundle experiment. This benchmark consists out of three phases: the (optional) preparation phase, the task I (“blind benchmark”) and the task II (“open benchmark”).

The GRS will evaluate and compare the results of the benchmark participants as well as facilitate the publication of a compiled report. The benchmark progress as well as the SPIZWURZ status and latest results will be presented in the scope of the SEDS workshops at GRS Garching.

ACKNOWLEDGEMENTS

The SPIZWURZ project (FKZ RS1586A/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.

The authors highly appreciate the technical support of the KIT workshops during this study.

The authors thank the Paul Scherrer Institute (PSI) for providing beam-time at the ICON facility and Anders Kaestner and David Mannes for their assistance during the measurements and the analysis.

Further, the chemical analysis group of Thomas Bergfeldt (IAM-AWP), the KIT’s QUENCH group and all colleagues is thanked.

NOMENCLATURE AND ACRONYMS

SPIZWURZ	German project title "*Sp*annungs*i*nduzierte *W*asserstoff*u*mlage*r*ung während Langzeit-*Z*wischenlagerung" (stress induced hydrogen re-distribution during long term intermediate storage)
KIT	Karlsruhe Institute of Technology
GRS	Gesellschaft für Anlagen- und Reaktorsicherheit
INCHAMEL	In-situ neutron radiography chamber for experiments with mechanical load

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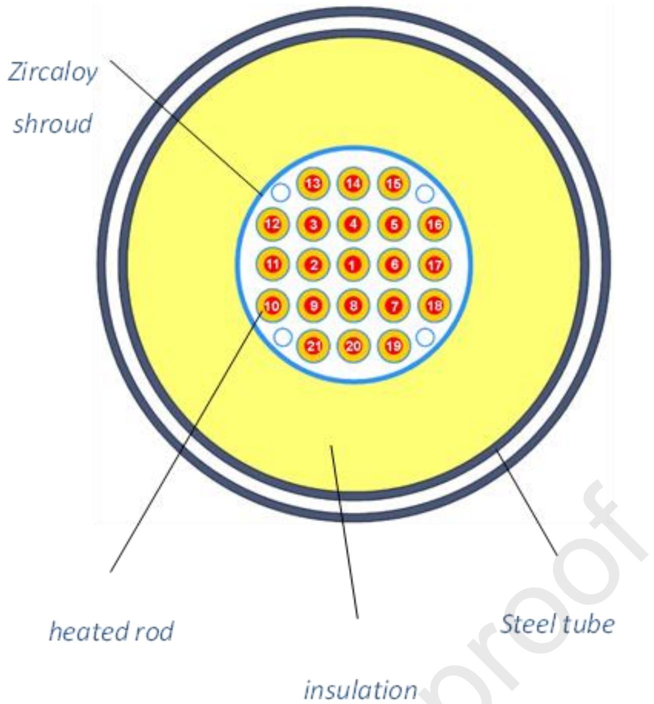

Fig. 1. Schematic illustration of the fuel rod bundle within the QUENCH facility.

Fig. 2 HoKi furnace dedicated for hydrogenation of 2.5 m long cladding tubes.

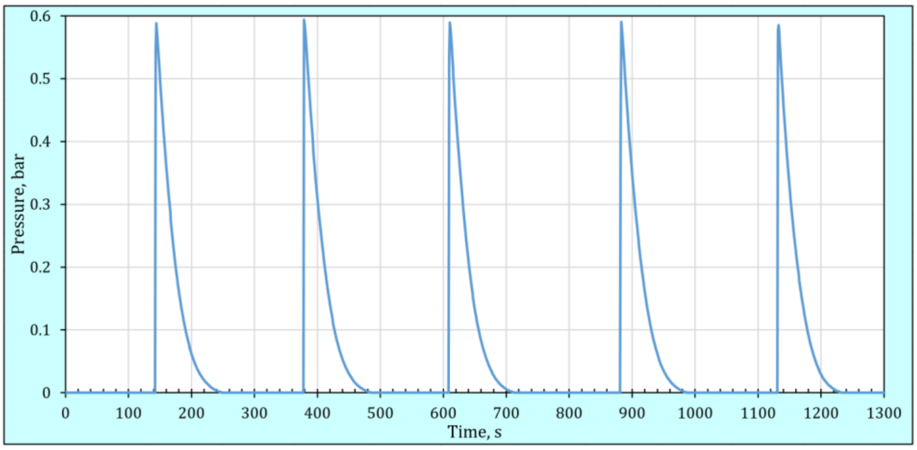

Fig. 3 Hydrogen pressure inside a tube during its hydrogenation in the HOKI furnace.

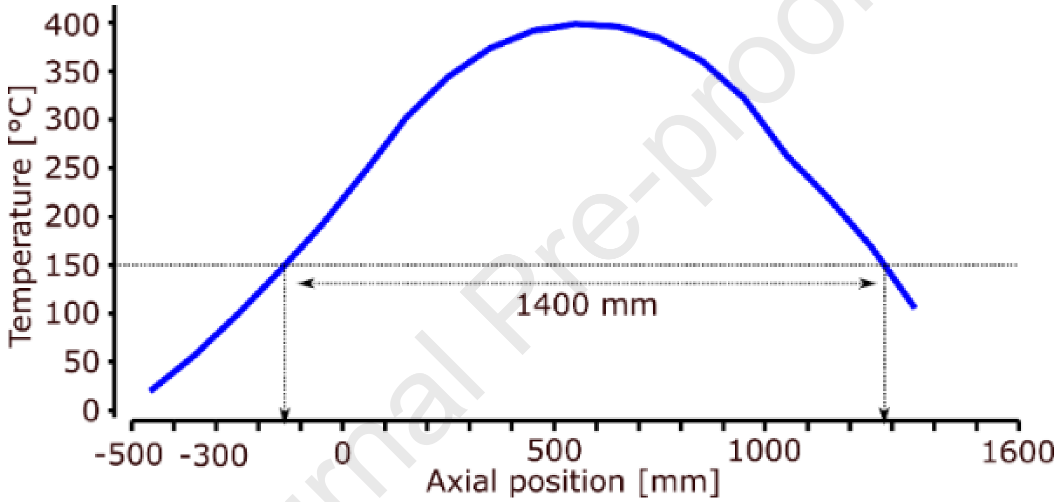

Fig. 4. Axial temperature distribution over the central rod at the experiment start predicted by ATHLET-CD.

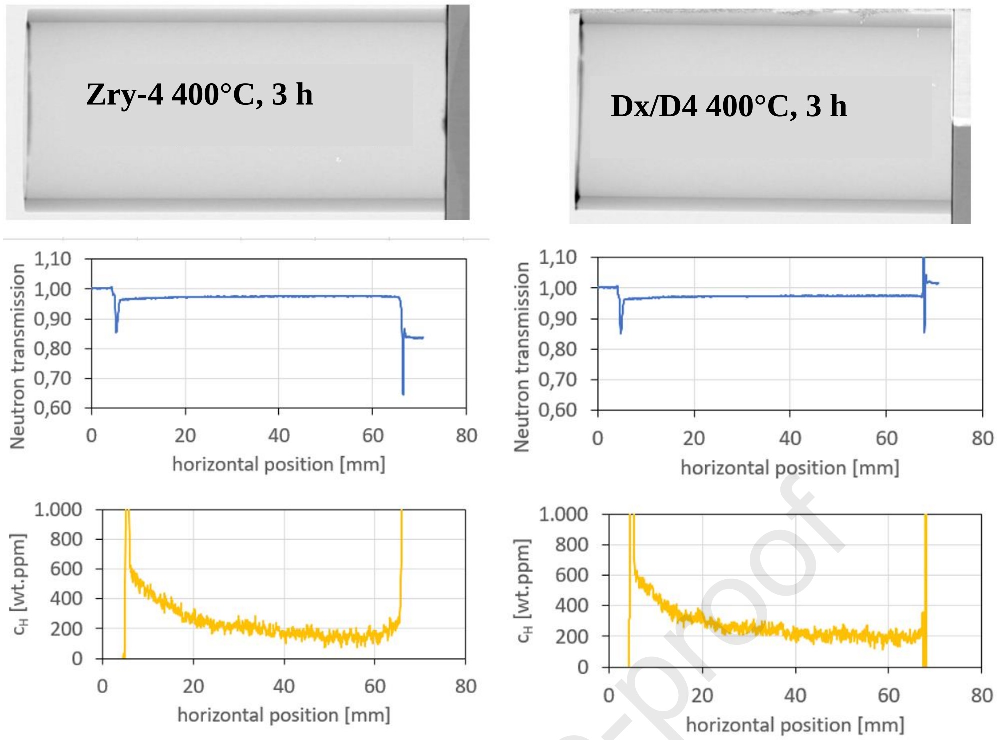

Fig. 5 Neutron radiographs and the axial distributions of the neutron transmission and the hydrogen concentration for Zry-4 and Dx/D4 DUPLEX after 3 h of annealing at 400°C with contact to the ZrH_2 powder bed only at the left side

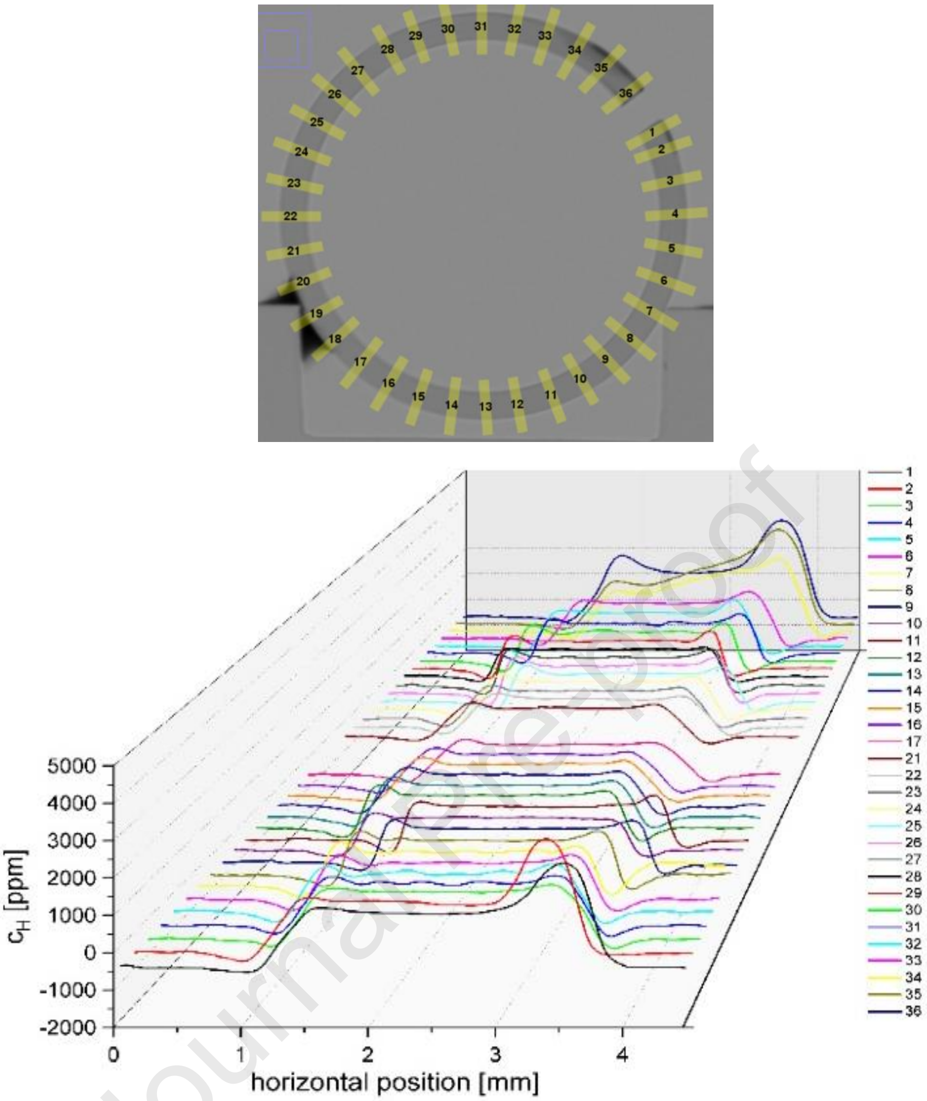

Fig. 6 Neutron radiographs of the Zry-4 sample annealed for 1 h at 450°C. The diagram shows the hydrogen distributions calculated from the intensity distributions along the marked bands for the investigation of the hydrogen diffusion in hoop direction

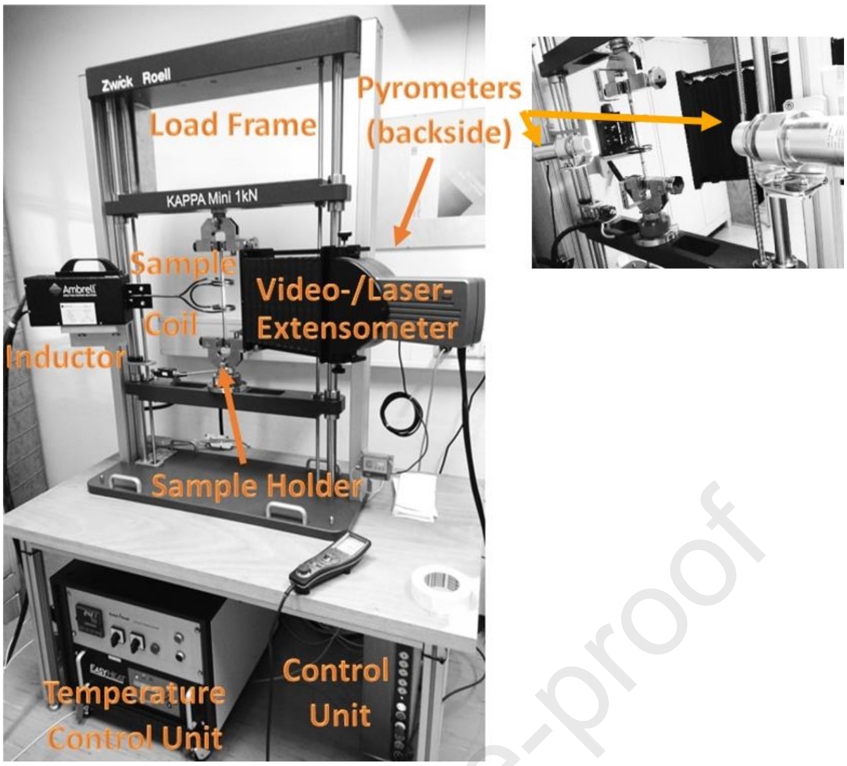

Fig. 7 The INCHAMEL facility at the IAM-AWP with inserted 300 mm long Zircaloy-4 cladding tube sample [3]

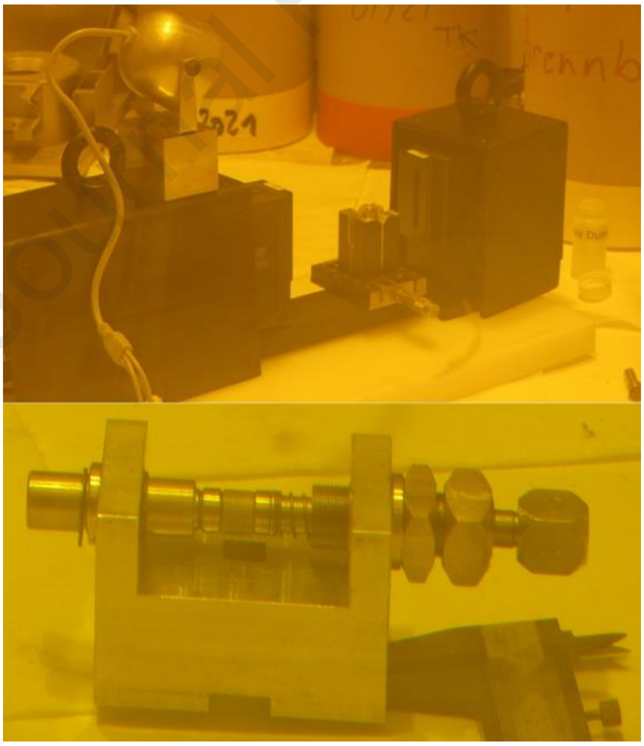

Fig. 8 The upper picture shows the modified laser scan micrometer inside a hot cell and the lower picture depicts the custom-made sample holder

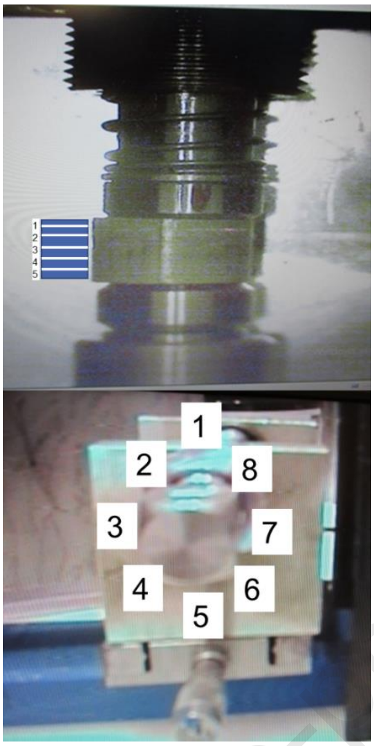

Fig. 9 The picture shows the Zircaloy specimen in the sample holder and the five longitudinal and eight radial positions used for the measurements

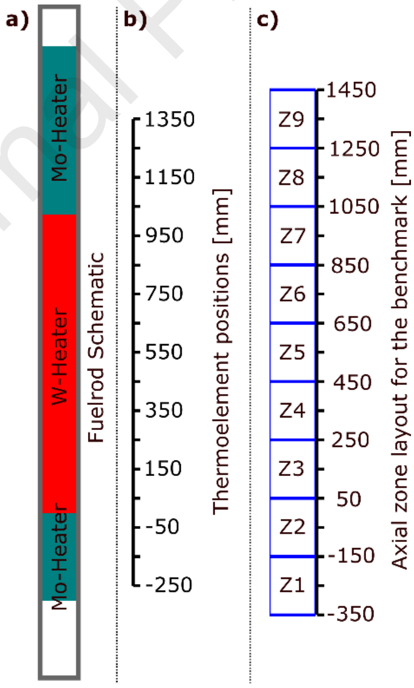

Fig. 10 a) Schematic illustration of a fuel rod simulator with heater. b) Axial positions of the thermo-couples. c) Axial zone layout for the benchmark

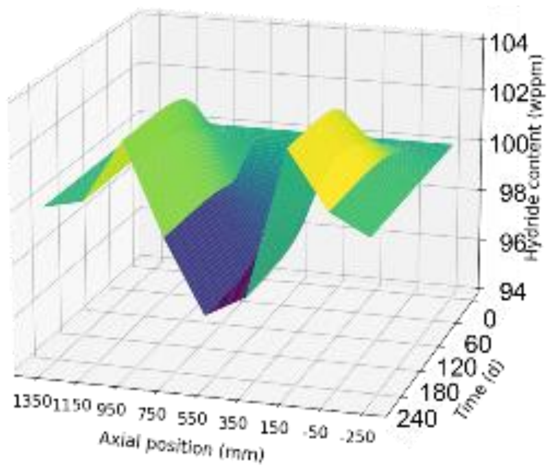

Fig. 11 Axial distribution of hydrogen concentration in cladding over time

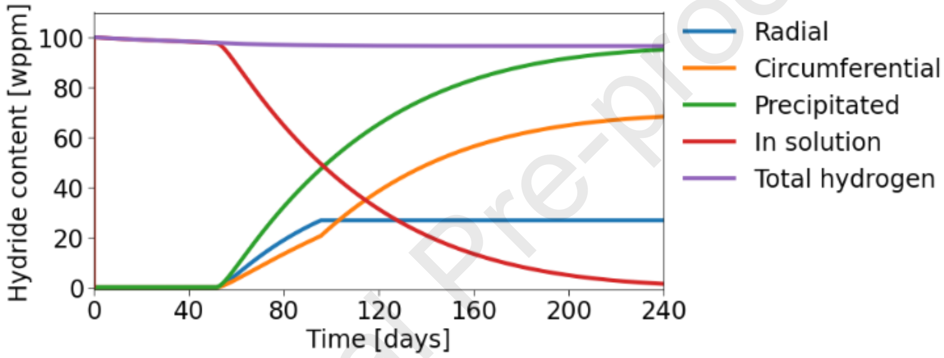

Fig. 12 Hydrogen and hydride behaviour over time in axial zone 5

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: