

**SEDS-Proceedings
of the 7th GRS Workshop**

**Safety of Extended
Dry Storage of Spent
Nuclear Fuel**

**Garching,
24th – 26th May 2023**

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Florian Rowold
Oliver Bartos
Klemens Hummelsheim

January 2024

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Keywords

Ageing Management, Cladding, Dry Storage, Inventory, Safety of the extended dry Storage of spent nuclear Fuel, Spent Fuel

Introduction

The Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH hosts its 7th workshop “Safety of Extended Dry Storage of Spent Nuclear Fuel (SEDS)” in Garching b. München. For the second time after the COVID-19 pandemic, the workshop was provided as an in-person event. The event attracted great attention as the program was filled with presentations from 18 institutes from 7 countries and attended by 45 experts. For Germany, the broad range of experts was represented by universities and research organizations, technical support organizations, fuel vendors, and the Federal Ministry for Economic Affairs and Energy. It started with a warm welcome of the chairpersons on the 24th of May 2023 followed by a broad overview of the research activities of the GRS and IAEA. With 15 presentations in the scientifically oriented agenda of the workshop, a wide variety of current research projects was reflected. The topics comprised material behaviour of claddings and sealings, simulation approaches for thermal cask evaluations and thermo-mechanical fuel rod performance during dry storage in common CASTOR® dry storage casks.

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1 Agenda

7th GRS Workshop on the Safety of Extended Dry Storage of Spent Nuclear Fuel

Chairs: Florian Rowold, Klemens Hummelsheim, Oliver Bartos

Location: Boltzmannstraße 14, 85748 Garching bei München

Wednesday, 24th May 2023

Time	Title	Speaker/Organization
12:00	Snacks & Drinks	
13:00	Welcoming and Workshop Overview	Chairs
13:20	Current and future GRS Dry Storage Research Activities	F. Rowold, GRS
13:50	IAEA Activities related to Extended Storage of Fuel	L. McManniman, IAEA
14:20	Coffee break	
14:40	1. Finding Critical Rods for Dry Storage Analyses	P. Konarski, PSI
15:10	2. Extended Storage of Spent Nuclear Fuel in Casks - Inventory Assessment using Fuel Rod Performance Codes	G. Spykman, TÜV Nord
15:40	Coffee Break	
16:00	3. Evaluating Spent Fuel Safety in Transportation/ Handling Accidents - A Statistical Approach	C. Aguado, CIEMAT
16:30	Experimental and numerical Validation of the Radionuclide Inventory in irradiated High Burn-Up UOX and (U, Pu)OX pressurized Water Reactor Fuel Claddings	T. König, KIT
17:00	Coffee Break & End of Session	
18:00	Buffet @ GRS in Garching	

Thursday, 25th May 2023

Time	Title	Speaker/Organization
9:00	Coffee & Tea	
9:45	Belgian SNF Ageing Management Program Strategy	C. Dupuit, Synatom
10:15	BGZ's Research Programme - Update and Overview	M. Stuke, BGZ
10:55	Coffee Break	
11:15	Cooling Rate Impact on Hydride Reorientation Embrittlement	J. Jonnet, EDF
11:45	Influence of Solid Solution Hydrogen on Zircaloy-4 Softening	F. Fagnoni, PSI
12:15	Lunch Break	
13:15	Muography for the Inspection of Dual-Purpose Storage Casks - Status Quo, Perspectives and GRS Activities	T. Braunroth, GRS
13:45	Radiation-based non-invasive Monitoring Methods for Transport and Storage Casks	S. Eisenhofer, HSZG
14:30	Coffee Break	
15:00	BGZ Research Activities on Muon Radiography	J. Niedermeier, BGZ
15:30	4.Update on the GRS Interim Storage Project BREZL-II	D. Nahm, GRS
16:00	Coffee Break & End of Session	
19:00	Dinner @ Hofbräukeller in München	

Friday, 26th May 2023

Time	Title	Speaker/Organization
9:00	Snacks & Drinks	
9:30	Development of an AGR Fuel Drying Rig	M. Morales, NNL
10:00	Cohesive Zone Modelling Approach on irradiated Claddings subjected to long-term Dry Interim Storage	M. Gaddampally, BAM
10:30	Coffee Break	
10:50	Update of the Experimental Work in the Framework of the SPIZWURZ Project	M. Große, KIT
11:20	Update of Neutron Radiography Experiments in the Framework of the SPIZWURZ Project	S. Weick, KIT
11:50	Summary and Discussion of the Workshop	All
12:15	End of Workshop	

2 Titles of the Lectures of the Authors

1. Current and future GRS Dry Storage Research Activities; Florian Rowold, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS)
2. IAEA Activities related to Extended Storage of Fuel; Laura McManniman, International Atomic Energy Agency (IAEA)
3. Finding Critical Rods for Dry Storage Analyses; Piotr Konarski, Paul Scherer Institut (PSI)
4. Extended Storage of Spent Nuclear Fuel in Casks -Inventory Assessment using Fuel Rod Performance Codes; Gerold Spykman, Technischer Überwachungsverein (TÜV Nord)
5. Evaluating Spent Fuel Safety in Transportation/ Handling Accidents - A Statistical Approach; Carlos Aguado, Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT)
6. Experimental and numerical Validation of the Radionuclide Inventory in irradiated High Burn-Up UOX and (U, Pu)OX pressurized Water Reactor Fuel Claddings; Tobias König, Karlsruher Institut für Technologie (KIT)
7. Belgian SNF Ageing Management Program Strategy; Charles Dupuit, Synatom
8. BGZ's Research Programme - Update and Overview; Maik Stuke, Gesellschaft für Zwischenlagerung (BGZ)
9. Cooling Rate Impact on Hydride Reorientation Embrittlement; Jerome Jonnet, Électricité de France (EDF)
10. Influence of Solid Solution Hydrogen on Zircaloy-4 Softening; Francesco Fagnoni, Paul Scherer Institut (PSI)
11. Muography for the Inspection of Dual-Purpose Storage Casks - Status Quo, Perspectives and GRS Activities; Thomas Braunroth, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS)
12. Radiation-based non-invasive Monitoring Methods for Transport and Storage Casks; Suzanne Eisenhofer, Hochschule Zittau/Görlitz (HSZG)
13. BGZ Research Activities on Muon Radiography; Julia Niedermeier, Gesellschaft für Zwischenlagerung (BGZ)
14. Update on the GRS Interim Storage Project BREZL-II; Daniel Nahm, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS)
15. Development of an AGR Fuel Drying Rig; Marissa D. Morales Moctezuma, National Nuclear Laboratory (NNL)

16. Cohesive Zone Modelling Approach on irradiated Claddings subjected to long-term Dry Interim Storage; Mohan Reddy Gaddampally, Bundesanstalt für Materialforschung und -prüfung (BAM)
17. Update of the Experimental Work in the Framework of the SPIZWURZ Project; Mirko Große, Karlsruher Institut für Technologie (KIT)
18. Update of Neutron Radiography Experiments in the Framework of the SPIZWURZ Project; Sarah Weick, Karlsruher Institut für Technologie (KIT)

2.1 Current and future GRS Dry Storage Research Activities

*Florian Rowold, Margarita Tzivaki, Kai Simbruner, Oliver Bartos,
Klemens Hummelsheim*

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), Germany

Due to the restart and further delays in the Repository Site Selection process in Germany, the extended storage of spent nuclear fuel is becoming increasingly important. Since the 2010s, GRS has been conducting various research projects on the topic of extended dry storage of spent nuclear fuel. The topics range from spent fuel behavior to hydrogen behavior, muon imaging for sealed casks, source term and radiological analysis, and the evaluation of load scenarios for spent fuel assemblies (see Figure 1). Spent fuel behavior was and is addressed within the BREZL projects. Hydrogen behavior is being examined in the SPIZWURZ project in collaboration with the Karlsruhe Institute for Technology (KIT).

Regarding the load scenarios for spent fuel assemblies after or during extended storage periods, the objective of the research project is to apply finite-element analysis to investigate transport or handling scenarios that result in additional loads for the fuel assemblies. In the first step, an extensive literature review was performed to identify useful information on stresses, accelerations, or transfer functions stemming from measurements and experimental setups, such as the transport and handling tests performed by the Sandia National Labs or different drop tests performed by GNS, BAM, or ENSA.

Another field of interest for GRS is the characterization of the spent fuel inventory and the evaluation of its radiological impact. A project to modernize the computational methods for source term analysis and dose rate determination was recently initiated. It builds on internationally used and validated software like MCNP and SCALE and employs a graphical user interface to facilitate knowledge transfer. All tools and libraries will be adapted to the current state of science and technology.

Besides the storage of spent fuel from light water reactors, spent fuel from research and prototype reactors has recently gained more attention due to the emerging longer storage periods. Gap analyses, similar to those performed for PWR and BWR fuels, also need to be conducted for the variety of fuel types from the more unusual reactors in Germany, such as the AVR and THTR high-temperature reactors or the multi-purpose research reactors (e.g., FRM-II or BER-II).

2.2 IAEA Activities related to Extended Storage of Fuel

Laura McManniman, Amparo González Espartero, Christoph Gastl

International Atomic Energy Agency (IAEA), Austria

N/A

2.3 Finding Critical Rods for Dry Storage Analyses

Piotr Konarski, Alexey Cherezov, Cedric Cozzo, Grigori Khvostov, Hakim Ferroukhi,

Paul Scherrer Institut (PSI), Switzerland

According to the Swiss nuclear fuel management policy, used nuclear fuel is planned to be disposed in a deep underground repository. Prior to final disposal, used nuclear fuel is stored at reactor sites and in a centralized dry storage facility. Since the operation dates of the final repository are unknown, extended periods of interim storage have to be considered. A research program to investigate the fuel rod integrity during long-term dry storage has been launched at Paul Scherrer Institute. In the context of the project, fuel rod performance simulations are carried out with the code Falcon.

Dry storage cannot be fully supported with experiments due to its long duration. Therefore, safety assessment relies largely on numerical calculations. Millions of rods have been irradiated in the Swiss reactors over decades and simulating all of them with state-of-the-art fuel performance codes would be extremely time-consuming. Therefore, Principal Component Analysis (PCA) and clustering algorithms have been applied to identify the most critical rods and reduce the number of computationally expensive fuel performance simulations. The data for PCA comes from the Studsvik Scandpower 3D core simulation code package CASMO5/SIMULATE3 and the identified rods are simulated with the fuel code Falcon and the hydrogen behavior code HYPE, which allows studying hydrogen-related degradation mechanisms based on the fuel performance results.

2.4 Extended Storage of Spent Nuclear Fuel in Casks -Inventory Assessment using Fuel Rod Performance Codes

Gerold Spykman

Technischer Überwachungsverein Nord Ensys (TÜV NORD EnSys), Germany

The dry storage of spent nuclear fuel in casks in Germany is limited to forty years' period by the license. Since a final repository will be not available in the next decades the licenses has to extend to longer period for dry storage in casks. In today's storage period the fuel verification for dry storage performance base on a few conservative chosen parameters, e.g. stress and strain calculated with conservative values / models for the cladding geometry, oxide layer thickness, inner rod pressure, cladding creep rate and cladding temperatures to ensure cladding integrity for a 40 years storage time. This verification yield on covering values of the parameter. In a prolonged storage period, other effects e.g. further fission gas release will increase the inner rod pressure e.g. alpha decay in MOX fuel, hydride precipitation, hydride reorientation, rod free volume changes due to creep and fuel swelling, solid body pressing of the pellet on the cladding due to pellet swelling and some more that may be identified in future.

For the assessment of the fuel performance in a prolonged storage period, a more realistic approach should be considered. In our investigations, we are using a probabilistic assessment instead of a conservative and covering approach for the most burdened rods only. Therefore, we implemented an integral calculation of the life span including in pile performance, wet storage on site and dry storage in cask up to approximately 100 years.

Further, we are determining the appropriate distributions for data und model parameters and variate all parameters as far as possible to evaluate the importance and to define probabilistic limits for all items of interest in a kind of an appropriate quantiles. The assessment will cover all fuel rods in a cask loading.

In this presentation, we show briefly the prerequisites for a probabilistic assessment, discuss the data sources and the modeling of the boundary conditions. Finally we will show some first results.

2.5 Evaluating Spent Fuel Safety in Transportation/ Handling Accidents - A Statistical Approach

Carlos Aguado, Francisco Feria, Luis E. Herranz

Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT), Spain

Nuclear fuel cladding serves as a crucial barrier to prevent the release of nuclear material to the environment during the reactor stage and subsequent storage until the final disposal of spent nuclear fuel. The potential radiological impact of a postulated accident during the transport of spent fuel depends on the number of failed rods resulting from the mechanical loading that the fuel rods might undergo.

Under the frame of the EJP EURAD project, a statistical methodology has been structured to quantify the number of failed rods in a postulated accident scenario during the transport of spent nuclear fuel. This methodology is based on the fuel performance code FRAPCON-4.0 coupled with the statistical toolbox DAKOTA. This paper describes the methodology and displays the results of its application to a number of accident scenarios.

Through the application of the methodology, it has been highlighted that the potential irradiation damage annealing of the spent fuel affects the simulated rod performance and it is heavily affected by the failure criterion posed in the methodology. The modelling of the annealing with FRAPCON-4.0 has been studied and its effect on the determination of the number of rods failed has been quantified through a sensitivity analysis. The major challenges and needs to consolidate the proposed methodology have been identified and discussed.

This work has been conducted under the frame of EJP EURAD project (Grant agreement ID: 847593) of HORIZON-2020.

2.6 Experimental and numerical Validation of the Radionuclide Inventory in irradiated High Burn-Up UOX and (U, Pu)OX pressurized Water Reactor Fuel Claddings

*Tobias König, Ron Dagan, Michel Herm, Volker Metz,
Arndt Walschburger, Horst Geckeis*

Karlsruher Institut für Technologie (KIT), Germany

Currently, spent nuclear fuel (SNF) assemblies discharged from German power reactors are stored in dual-purpose casks (DPC) in interim dry storage facilities and kept there until their conditioning for final disposal. Licenses for both, the DPCs and interim dry storage facilities, will expire after forty years consecutive to the loading of the cask and the first emplacement of a DPC in the storage location. Due to considerable delays in the site selection process and the resulting absence of a final repository in Germany so far, a prolongation of interim dry storage of a timespan of about 100 years is inevitable. Regarding this long timespan, the integrity of the SNF cladding is of significant importance in respect of the latter conditioning of the fuel assemblies for final disposal.

With increasing burn-up, the gap between the nuclear fuel pellet and Zircaloy cladding tube closes under the formation of an interaction layer leading to chemical and mechanical interactions between pellet and cladding. Within this fuel-cladding interface, possible cladding degrading elements can accumulate and induce pitting or stress corrosion processes. In addition, the cladding is exposed to alpha irradiation damage induced by the decaying actinides present at the periphery of the fuel pellet. Precise knowledge of the radionuclide content in SNF and the cladding is of utter importance for a thorough reevaluation of all safety relevant aspects regarding the long-term behaviour of the casks and their inventory. Moreover, knowledge of the SNF radionuclide inventory is important in safety assessments for final disposal, in order to scrutinise the individual radionuclide source terms in case of ground water intrusion into the repository and a consecutive cladding failure. However, uncertainties in calculated radionuclide inventories in SNF can be about 2 % for uranium and plutonium, 7 % for fission products and up to 11 % for minor actinides [1]. Uncertainties of activation products, present in the claddings, can be much higher since they depend solely on the precursor content within the material [1].

Within our contribution, we provide radionuclide inventory data of Zircaloy-4 claddings taken from a high burn-up UO_x (50.4 GWd/t_{HM}) and an (U, Pu)O_x (38.0 GWd/t_{HM}) fuel, previously irradiated in commercial pressurised water reactors during the 1980s.

Furthermore, the obtained experimental data is compared to results received by MCNP/CINDER and webKORIGEN calculations in order to validate the performance of calculation codes against radiochemical analyses. For activation products, such as ^{14}C , ^{36}Cl or ^{125}Sb , a good agreement between experimental analysis and numerical calculations is apparent. However, in case of fission products and actinides, the radiochemical determined activities differ significant from values obtained by MCNP/CINDER and webKORIGEN calculations.

References

- [1] Spahiu, K., State of the Knowledge (SoK) Report, European Joint Programme on Radioactive Waste Management (EURAD) (2021).

2.7 Belgian SNF Ageing Management Program Strategy

Charles. Dupuit,

Synatom, Belgium

Synatom is a unique Belgian company that plays a key role in the nuclear fuel cycle. At the front end of the cycle, it supplies Belgian nuclear operators with enriched uranium. At the back end of the cycle, SYNATOM takes all the action needed to make spent nuclear fuel assemblies safe, until their transfer to public body in charge with their final disposal.

Synatom also provides highly specific financial services. It is responsible for covering:

- the costs of decommissioning nuclear power plants;
- the costs associated with managing spent fuel assemblies until their final transfer to ONDRAF/NIRAS.

Over the years Synatom built up considerable financial provisions. In the frame of its missions, Synatom designed a plausible industrial scenario for Spent Fuel management, based on ONDRAF/NIRAS own schedule, to evaluate the required back-end provisions.

It implies interim storage of Spent Nuclear Fuel Assemblies for a duration of about 80-100 years, before their reconditioning in canisters. Synatom is in the process to set up a Spent Fuel Ageing Management Program, to ensure that the aging effects on the Spent Fuel during its storage in dry facilities do not jeopardize its structural functional integrity.

The AMP will follow the guidance on the Managing Aging Processes in Storage created by United State Nuclear Regulatory Commission Regulation (NUREG-2214). It relies on released public data and documents, such as NRC ISG-11 and NUREG-2224, to establish storage criteria and assessment of ageing phenomena of major importance.

The ageing itself will be followed via a surrogate program, mainly based on DOE High Burnup Demonstration Program as surrogate, and complementary research.

As such, Synatom has a keen interest to follow latest “trends” related to spent fuel behaviour in dry storage, as well as building connections with key research players.

2.8 BGZ's Research Programme - Update and Overview

Maik Stuke

Gesellschaft für Zwischenlagerung (BGZ), Germany

During this presentation, we will provide an overview of our current research on high-level radioactive waste stored in the interim storage facilities of BGZ. We will be focusing specifically on the light water reactor fuel assemblies, with a particular emphasis on the challenges that arise during extended storage periods.

BGZ has identified research needs to ensure that the protection goals for storage times beyond the current 40-year limit are met. Our research strategy is protection goal-oriented and is based on the timeline resulting from the expiration of storage licenses, the commissioning of a repository, and the actual duration of the dry interim storage.

The BGZ's research program [1] outlines the necessary research needed to address identified knowledge gaps related to inventories, casks, and storage buildings in relation to the extended storage periods.

We will present updates and an overview of ongoing research projects related to casks and inventories. Specifically, we will report on the progress of the projects MSTOR – Metal Seals During Long-Term Storage, and BGZ's dose rate and temperature measurement programme OBSERVE as well as on our further computational benchmarking activities and the Long-term Experimental Dry storage Analysis (LEDA) project for inventories. The results and insights gained from these research endeavours will aid in achieving the protection goals for extended storage periods of high-level radioactive waste.

2.9 Cooling Rate Impact on Hydride Reorientation Embrittlement,

*Jerôme Jonnet, Antoine Ambard, Nathanaël Mozzani,
Ludovic Idoux, Davide Costa, Marc Ton-That*

Électricité de France (EDF), France

Context of the study

It is well-known that Hydride Reorientation (HRO) that occurs under internal pressure during cooling might generate some embrittlement of the cladding. Reorientation is done by setting an internally pressurized tube in a furnace at high temperature (typically 400°C) and let it cool down under stress. Because of experimental constraints the cooling rate effect has been hardly worked out. It is usually higher than the cooling rate in the case of dry storage.

Experimental setup for HRO heat treatment

A specific device has been developed to investigate the cooling rate effect. It consists in using four nozzles to inject compressed air (up to 5 bar) and located at the mid-plane of the tube (left-hand side of Fig. 1). A significant axial thermal gradient is expected during the tube cooling. This axial gradient is deduced from the temperature measurements by use of six thermocouples located along two opposite generatrices. The temperatures measured during cooling of two tubes are shown on Fig. 2. This setup allows a net variation of cooling rate along a short segment as shown on the right-hand side of Fig. 1.

Ring tensile test results and analysis

Following the HRO heat treatment, rings are cut and ring tensile tests are performed. For each test, a stress-displacement curve is obtained. The area below the curve is homogeneous to an energy density, here the Strain Energy Density (SED). The SED is chosen in the present study to assess the residual ductility of the hydrided and reoriented tubes. For an as-fabricated Zy-4 cladding at room temperature, the reference SED is about 0.6 J/mm². Above 0.15 J/mm² obtained with ring tensile tests, the material is considered as ductile. The calculated relative SED for the tested rings located at the thermocouple positions is plotted on Figure 3. The results show that:

At low cooling rate (typically a few °C/mn), the material is brittle. Decreasing further the cooling rate does not generate further degradation.

At high cooling rates, the material recovers its ductility.

The variation of ductility recovery is a continuous function of the cooling rate. No threshold is evidenced.

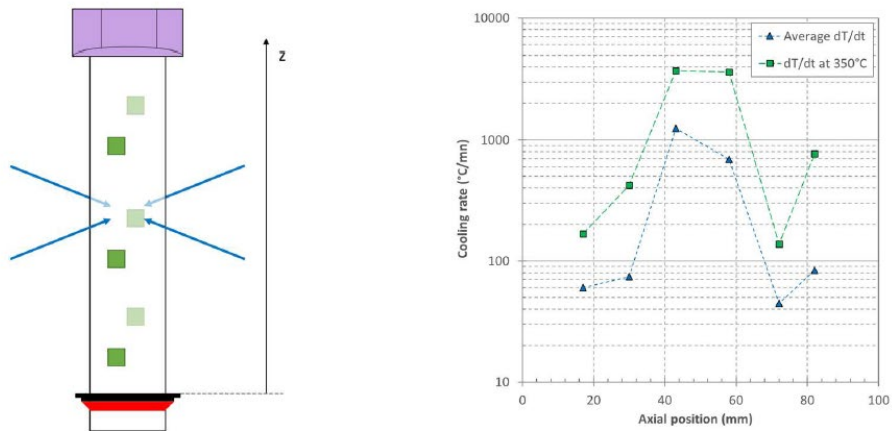


Fig. 1 Experimental setup (left-hand side) and average and instantaneous cooling rates calculated for the reoriented tube cold-down with injected air at 3 bar (right-hand side)

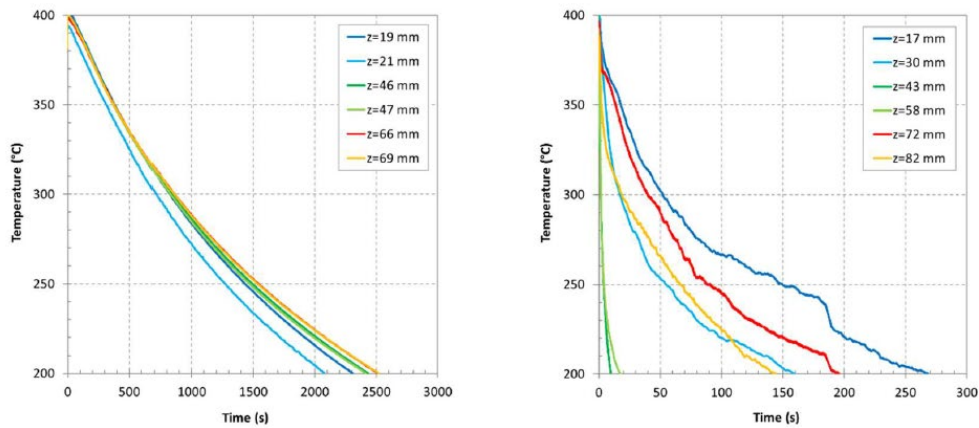


Fig. 2 Cooling as measured by thermocouples for the standard - no injected air - HRO heat treatment (left-hand side) and with injected air at 3 bar (right-hand side)

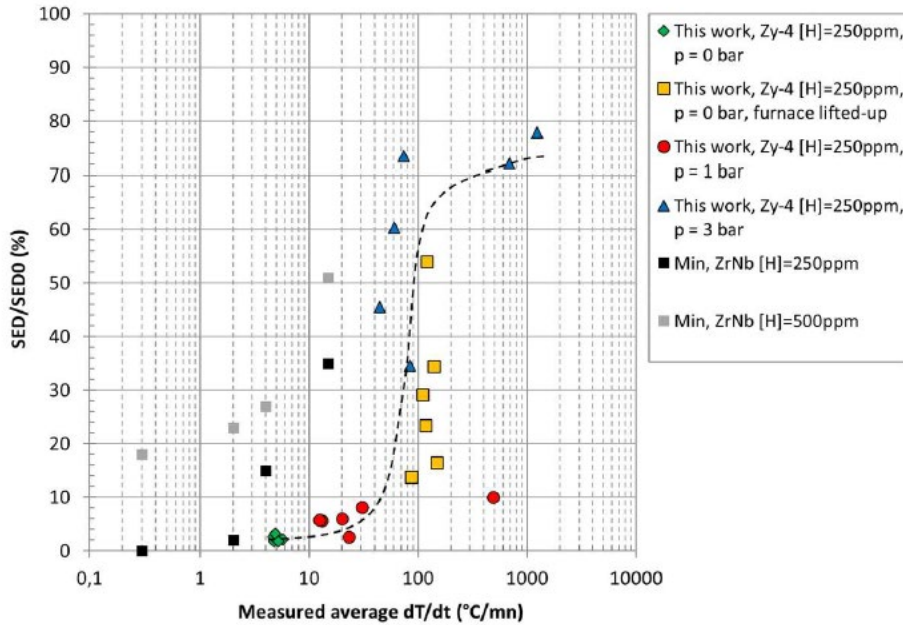


Fig. 3 Relative Strain Energy Density of tested rings as a function of the average cooling rate after HRO heat treatment Influence of Solid Solution Hydrogen on Zircaloy-4 Softening

The cooling rates deduced by the thermocouple measurements on the tube outer surface shown on Fig. 2 are probably overestimated for the inner surface due to the thermal inertia of the oil used to pressurize the tubes at the test temperature. Consequently, the precipitation rate of hydrides located close to the tube inner surface is probably overestimated too. Nevertheless, a tendency is observed with the recovery of ductility through SED calculations for cooling rates above 100°C/mn. These results agree with the literature (internal references), obtained in other laboratories with similar HRO heat treatment conditions and on different cladding materials (irradiated or unirradiated). As for the results obtained by Min et al. [1] reported on Fig. 3 with RTT based on HRO heat treatment performed on rings, a partial but significant recovery of ductility is observed for low cooling rates, between 4 and 15°C/mn. These results differ from the present work but, more importantly, they agree with the beneficial effect of increasing the cooling rate. The results show that the back-end fuel cycle operational cooling rates are in the range where the material is embrittled if [H], hoop stresses and temperature are in the ad hoc ranges. Increasing the cooling rates should be beneficial to the ductility recovery of the cladding.

References

- [1] S.-J. Min, M.-S. Kim and K.-T. Kim, Cooling rate- and hydrogen content-dependent hydride reorientation and mechanical property degradation of Zr–Nb alloy claddings. *Journal of Nuclear Materials*, Vol. 441, pp. 306-314, 2013.

2.10 Influence of Solid Solution Hydrogen on Zircaloy-4 Softening

Francesco Fagnoni^{1,2}, Ralph Spolenak², Johannes Bertsch¹, Liliana I. Duarte¹*

¹ *Nuclear Energy and Safety, Laboratory for Nuclear Materials Paul Scherrer Institut, (PSI), Switzerland,*

² *Laboratory for Nanometallurgy, Department of Materials, ETH Zurich,, Switzerland*

This study investigates the effects of hydrogen presence and thermal history on the mechanical properties of Zry-4, a zirconium alloy commonly used in nuclear applications. Tensile tests, three-point bending tests, and micro-hardness measurements were conducted on Zry-4 samples at approximately 300 °C in the presence of hydrogen. The results demonstrate that free hydrogen lowers the yield point and hardness of the zirconium alloy. The minimum yield point and hardness is observed when the majority of hydrogen is in solid solution. Furthermore, the thermal history of the samples plays a significant role in determining their mechanical properties. Upon cooling, a substantial hardness reduction of 7.8 % is observed compared to the material tested upon heating, which is attributed to the increase in solid solution hydrogen. These findings provide valuable insights for assessing the performance of zirconium alloys in conditions relevant for prolonged storage of spent nuclear fuel.

2.11 Muography for the Inspection of Dual-Purpose Storage Casks - Status Quo, Perspectives and GRS Activities

Thomas Braunroth

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), Germany

Muography [1], i.e., the non-invasive imaging with atmospheric muons, has received an increasing interest in the R&D community and is already used in several fields of applications on a routine basis. Atmospheric muons are characterized by relativistic energies which leads to large ranges in matter, allowing them to traverse even dense and extended materials such as volcanoes or pyramids. In addition, muons show a scattering behavior that is correlated to the effective atomic number of the traversed material, which makes them a suitable tool for the identification of High-Z materials in an otherwise low/medium Z environment. Besides archaeological and geological applications, muography is also applied in the nuclear field where it is used for the identification of nuclear materials, the scanning of reactors [2], and others. In addition, many recent studies investigate the applicability of muography to image Dual-Purpose Storage Casks and their interiors (fuel assemblies). These studies are motivated by a high-demand for a non-invasive imaging technology and a foreseeable extended interim-storage period in many countries.

This presentation provides an overview of muography and its general applications. It focuses on the application of muography for the non-invasive imaging of Dual-Purpose Casks, for which a short summary of the status quo based on recent works is provided. The presentation stresses that progresses require efforts in four interconnected fields: Detector development, numerical studies, image reconstruction (algorithms) as well as test- and field-measurements. It is therefore true to say that the application of muons for imaging purposes is a true multidisciplinary effort, requiring different competences and a high degree of communication between the associated fields.

At the end of the presentation, recent GRS activities, e. g. feasibility studies showing the applicability to identify missing fuel assemblies and missing individual fuel rods are summarized (see also Fig. 1) [3]. This summary is completed by a short outlook on upcoming GRS activities in this field.

References

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- [2] S. Procureur *et al.* – *3D imaging of a nuclear reactor using muography measurements* published in: *Sciences Advances* **9**, eabq8431 (2023), Doi: 10.1126/sciadv.abq8431
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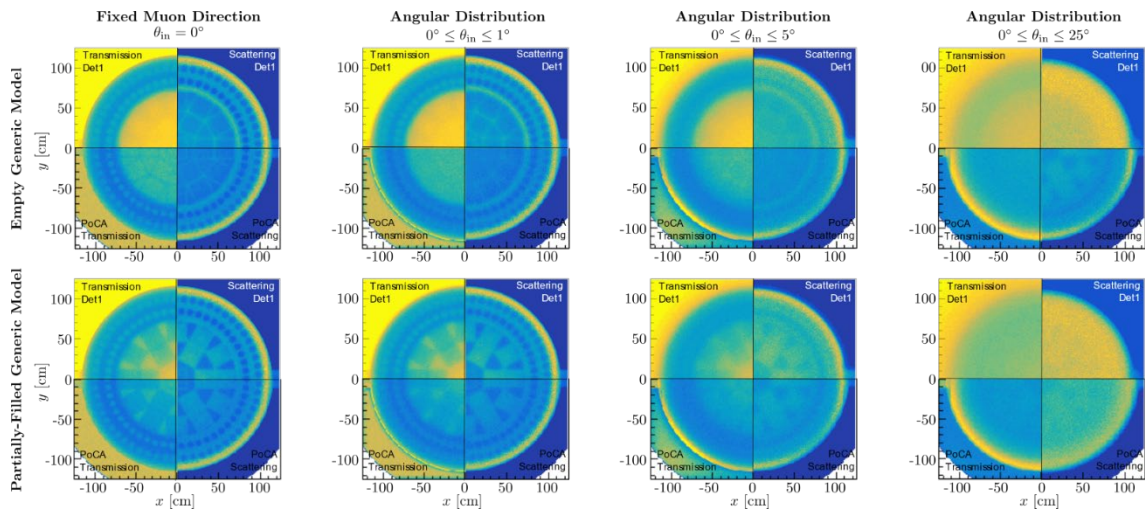


Fig. 1 Projection images using atmospheric images showing an empty (top) and partially filled (missing central fuel assembly, bottom) generic model using different information (transmission probability, scattering property) associated to different position information (Detector position information and Point-of-Closest Approach (PoCA)). The effect of different angular acceptances on the image quality is shown from left to right.

2.12 Radiation-based non-invasive Monitoring Methods for Transport and Storage Casks

*Suzanne Eisenhofer¹, Sebastian Reinicke¹, Sören Alt¹, Sebastian Kobelt²,
Michael Wagner², Uwe Hampel²*

*¹ Institute for Process Technology, Process Automation and Measurement Technology,
Zittau/Görlitz University of Applied Sciences, Germany,*

² Institute of Power Engineering, Technische Universität Dresden, Germany

This study investigates the non-invasive monitoring methods of muon tomography and gamma and neutron flux measurement for assessing the state of inventory of CASTOR interim storage casks. Following the conclusions of the precursor study, these two radiation-based methods were deemed the most successful for evaluating the condition of spent fuel. This work focuses on the latter flux distribution approach. Both Monte Carlo simulations and a fully designed and constructed automated measurement system are employed to obtain the radiative flux spectra outside the cask wall. Partnering with EWN, experimental data will be taken this year at the interim storage facility ZLN using two CLYC sensors that can efficiently differentiate between neutron and gamma flux distributions. The open-source Monte Carlo particle transport program FLUKA is utilised and fine-tuned to simulate and gather the flux distribution. As interim storage casks are designed for shielding, variance reduction techniques such as region biasing are used to balance appropriate statistical error and CPU time. The intention is for experimental and simulated data to corroborate and validate one another, and to determine adequate measurement time and the detection sensitivity of the measurement system. With the simulation method's substantiation, realistic inhomogeneities and hypothetical fuel relocation setups can be simulated and spectra obtained. This will provide an indication of what measured flux distributions can be expected. With a more informed understanding of the state of the inventory, appropriate measures and preparation for future handling can be organised with a higher degree of safety.

2.13 **BGZ Research Activities on Muon Radiography**

Julia Niedermeier

Gesellschaft für Zwischenlagerung (BGZ), Germany

Cosmic muons provide a non-destructive imaging method for nuclear fuel in sealed dry storage casks. By analyzing the scattering data of muons after traversing a cask, information about the cask's interior can be inferred via the effective scattering angles, which depend on the atomic number and densities of the materials the muons interact with. BGZ pursues a comprehensive research program on muon radiography or tomography, encompassing both theoretical and experimental work. We will present and discuss some recent results in this talk.

Our latest theoretical work investigates the impact of modeling assumptions and simplifications on the effective scattering angle. We discuss four GEANT4 cask models of a CASTOR® V/19 with different degrees of simplifications, including the complete omission of individual components. Our results indicate the importance of considering the level of simplification used in the model to ensure accurate and reliable results for the scattering angle distribution.

Additionally, an international collaboration involving INFN Padova, the University of Padova, the University of Brescia, the Gesellschaft für Zwischenlagerung (BGZ), Forschungszentrum Jülich, and the European Commission, Directorate General for Energy, Luxembourg, conducted a feasibility study on the possible use of muon tomography as re-verification method for selfshielding casks. The experiment in the dry spent fuel storage at Grafenrheinfeld employed drift tube detectors encircling a CASTOR® V/19 cask to capture the trajectories of atmospheric muons traversing the volume. The acquired data is analyzed with a multi-point measurement setup consisting of three different positions to differentiate fuel assemblies from dummy fuel assemblies. Even though the analysis of the data is ongoing precluding any definitive conclusions at this time, we will present some recent developments. This project is funded by the Federal Ministry for the Environment, Nature Conservation, Nuclear Safety and Consumer Protection (BMUV), Germany, under the funding code 02W6279.

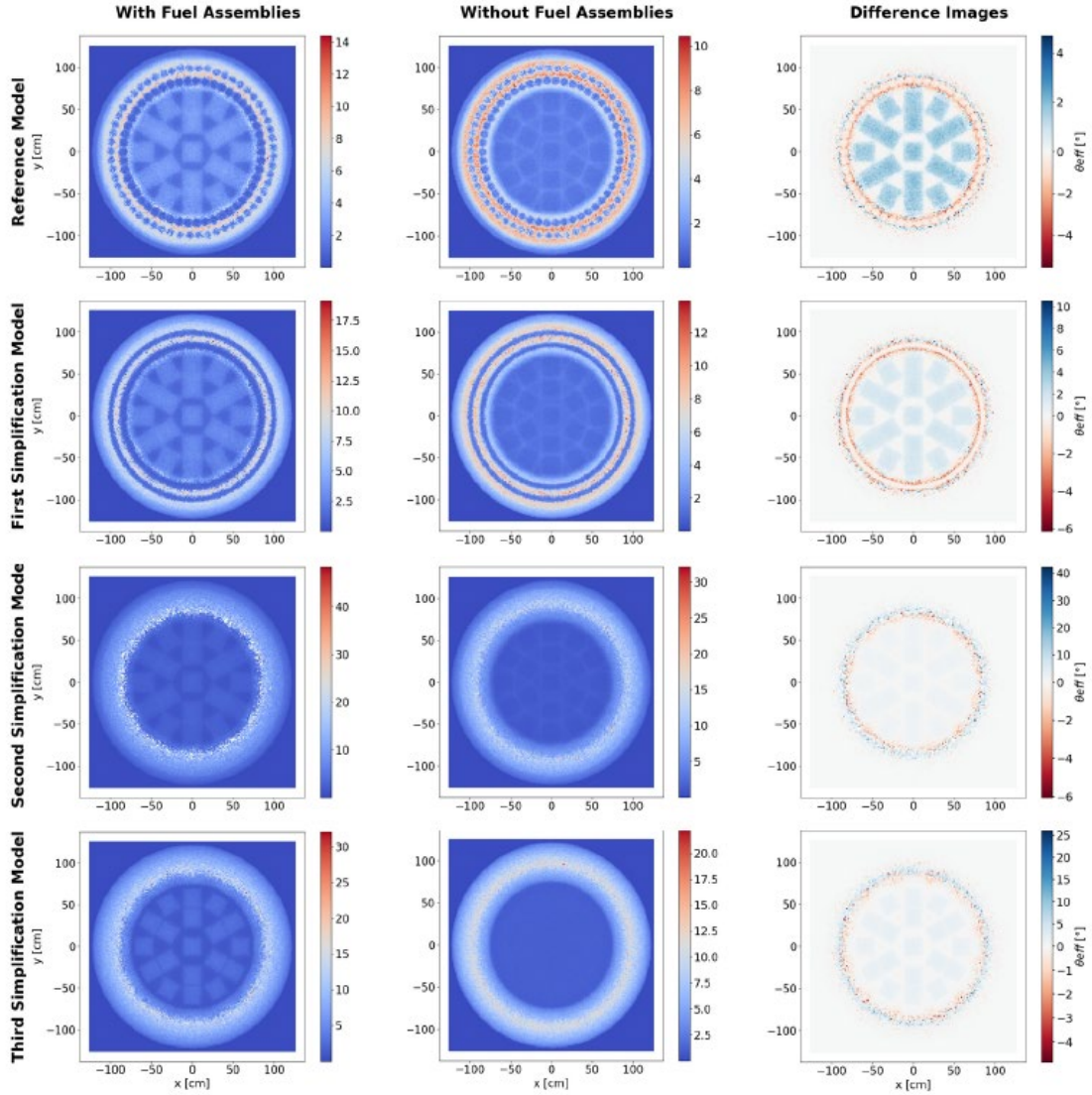


Fig. 1 The figure displays scattering images for all models, with and without fuel assemblies, as well as difference scattering images. The difference images are generated by subtracting the scattering angles with fuel assemblies from those without, for each model. The color code represents the mean of the effective scattering angle distribution (in $[\circ]$) for each pixel, with the x- and y-positions indicating the muon position after traversing the incoming detector. These images were generated through simulation of the reference model (first row), as well as the first, second, and third simplification models (row 2 to 4, respectively) both with (first column) and without fuel assemblies (second column). The difference scatter plots in the third column highlight the variations in scattering angles between simulations with and without fuel assemblies.

2.14 Update on the GRS Interim Storage Project BREZL-II

Daniel Nahm, Felix Boldt, Matthias Küntzel, Jonathan Sappl

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), Germany

Extensive knowledge of the thermo-mechanical behaviour of fuel rods is essential for providing safety predictions regarding the extended dry storage of spent nuclear fuel in storage and transportation casks. To give accurate predictions, it is necessary to consider the entire lifespan of the fuel rod, which includes in-reactor irradiation, wet storage in fuel ponds, loading to and drying inside the casks, and subsequent long-term dry storage. Throughout its lifecycle, the fuel rod experiences significant changes in environmental conditions, including pressure gradients, varying heat loads, and ongoing radioactive decay of the spent fuel. To predict fuel rod behaviour during storage, the GRS (Gesellschaft für Anlagen- und Reaktorsicherheit) utilizes a simulation chain comprised of MOTIVE as burnup code, COBRA-SFS as thermo-hydraulic code, and TESP-ROD as fuel rod code. COBRA-SFS, a validated simulation tool, is employed to calculate thermo-hydraulic boundary conditions for fuel rod analysis during cask-based dry storage. It is coupled with the GRS fuel rod code TESP-ROD to predict thermo-mechanical parameters of individual fuel rods. In the scope of the BREZL projects the GRS conducts research on the fuel rod behaviour during storage.

The project BREZL-II is focused on advancing the hydride modelling capabilities of TESP-ROD, expanding the GRS storage and transportation cask model repertoire, and improving the general accessibility of the calculation chain by a graphical user interface (GUI). Furthermore, detailed uncertainty and sensitivity analyses of both the temperature field and the mechanical simulations are conducted.

To expand the GRS cask repertoire, a generic boiling water reactor storage and transportation cask model was developed. In a homogenous cask loading scenario with 40 kW the best estimate, i.e. not necessarily conservative model revealed a peak cladding temperature of 282 °C (see Fig 1).

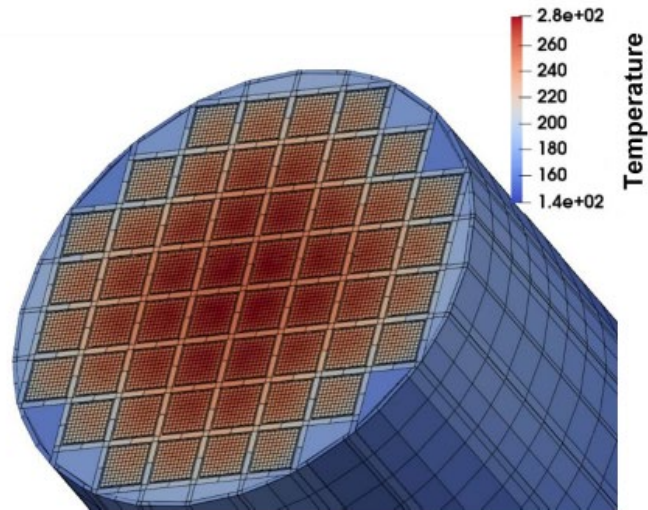


Fig. 1 Three-dimensional visualization of the temperature field in the BWR vessel model using ParaView

For a better understanding of the model, detailed uncertainty and sensitivity analyses were performed by varying 23 model parameters using a Monte Carlo based approach. In this analysis, the ambient temperature, the basket gap, the decay heat, the thermal conductivity of the basket and the external heat exchange coefficient were found to be most influential on the peak cladding temperature (see Fig. 2). Interestingly, the influence of the varied parameters on the temperature depends much on the position analysed in the cask. A potential offset of the fuel assembly basket, for instance, showed neglectable impact to the peak cladding temperature, while in contrast it showed strong impact to the cask surface temperature.

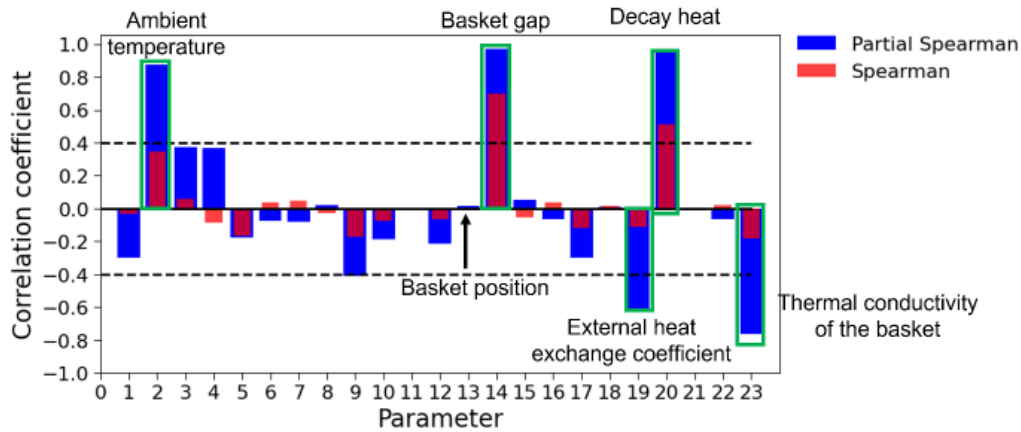


Fig. 2 Sensitivity analysis of the generic SWR cask model regarding the peak cladding temperature

A graphical user interface (GUI) was developed to streamline the simulation of an interim storage scenario. Currently the GUI allows for the configuration of PWR and BWR storage scenarios and adjust simulation parameters such as individual decay heat of the fuel assemblies, the axial resolution of the models, or the ambient temperature. Additional features such as the configuration of TESPA-ROD inputs are planned for future activities.

Considering the GRS capabilities for the simulation the fuel rod behaviour during storage, the cladding model of the fuel rod code TESPA-ROD was extended. The previous model allowed segmenting the cladding in axial direction only. While hydrogen could diffuse in axial direction, no resolution of the hydrogen concentration within one axial level was possible. In BREZL-II the TESPA-ROD cladding model was expanded to subdivide each axial level into several radial zones. Hydrogen diffusion is enabled between these zones. This allows for a more reliable description of the hydrogen behaviour in the fuel rod cladding during storage and can be used to simulate a “liner”-effect as well.

With the developments within the BREZL-II project, GRS extends its calculation chain for the analysis of spent nuclear fuel during long-term dry storage. However, there are several phenomena with potential impact on the integrity of a fuel rod during storage, which are still not yet fully understood. Examples thereof are fuel swelling during dry storage, reorientation process of hydrides, or delayed hydride cracking. These processes might reduce the damage threshold of the cladding significantly

and therefore impact the safety margins for fuel rods during storage. Hence further investigations are required to accurately implement such phenomena in fuel rod performance codes, to allow for an improved long-term prediction of the fuel rod behaviour during storage.

2.15 Development of an AGR Fuel Drying Rig

Marissa D. Morales Moctezuma, James B. Goode

National Nuclear Laboratory (NNL), UK

Until 2018, spent AGR fuels were reprocessed at Sellafield in the Thermal Oxide Reprocessing Plant (THORP). The Nuclear Decommissioning Authority (NDA) spent fuel management strategy 4 has now been updated to consolidate all spent AGR fuel from the EDF AGR stations in the THORP Receipt and Storage (TRS) pond at the Sellafield site, and interim store all oxide fuels pending a future decision on whether to classify the fuel as waste for disposal in a Geological Disposal Facility (GDF) [1]. This has led to an increased interest in fuel drying either as a potential contingency option if pond storage is found not to be viable in the long term or ahead of direct disposal if the fuel becomes waste. There is significant international experience of drying and dry storage of spent nuclear fuel; however, this has focussed on zirconium alloy clad and Light Water Reactor (LWR) fuel. The majority of spent fuel in the United Kingdom is from the second generation Advanced Gas-cooled Reactors (AGRs) and has properties which raise questions over how effective drying may be. This work describes NNL's experimental programme, on behalf of the NDA and Sellafield Ltd, to investigate the drying behaviour of irradiated AGR fuel, with the objective to provide data to support long-term drying research. This work will mainly focus on the preparations considered for the design of the Experimental AGR Drying Rig (EADR). Two nominally identical versions of EADR were built, one for installation in the Windscale hot cell facilities and another one installed at NNL's Workington Rig Hall. EADR's active version will initially be used to dry ~50 irradiated AGR fuel pins, which are to be examined as part of a study of fuel condition after long-term pond storage. A proportion of the fuel pins under investigation are believed to have failed in the mid 1980's due to a combination of previous storage conditions (e.g. absence of corrosion inhibitor in pond) and post-irradiation effects caused by chromium depletion from the grain boundary in the cladding. These are believed to be the only failed AGR fuel pins in pond storage and therefore it is expected that unique data will be collected during the active drying trials. The non-active replica of EADR has been designed to carry out a series of development trials in advance of the active programme. It will be used to provide an understanding of the rig behaviour under controlled conditions so that data collected on real fuel pins in later work can be correctly interpreted. Tests carried out in EADR's non-active version will use a number of simulant mock pins with machined microdefects and tungsten carbide pellets with similar thermal properties (albeit without

decay heat) to UO₂ as well as samples that have been investigated in detail at the University of Leeds [2]. The work presented here details the state-of-the-art design of the drying rig built to be deployed in an existing hot cell facility in NNL, some of the testing that is due to be carried out with the rig in order to provide insights into the measurement of water inside fuel pins, drying behaviour and characteristics indicating the extent of drying. Keywords: Irradiated AGR fuel, drying behaviour.

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2.16 Cohesive Zone Modelling Approach on irradiated Claddings subjected to long-term Dry Interim Storage

Mohan Reddy Gaddampally, Uwe Zencker, Holger Völzke

Bundesanstalt für Materialforschung und -prüfung (BAM), Germany

Germany Long-term dry interim storage may adversely affect the mechanical properties of spent fuel rods, possibly resulting in a reduced resilience during handling or transport after storage. Pre-storage drying and the early stage of interim storage can subject the cladding to higher temperatures and higher pressure induced tensile hoop stresses than those associated with in-reactor operation and pool storage. Under these conditions, radial hydrides may precipitate in zirconium-based alloys (Zircaloy) during slow cooling, which may result in embrittlement of the cladding material and eventually a sudden failure of cladding under additional mechanical loads. Especially long, continuous radial hydride structures and low temperature can cause severe embrittlement of claddings and finally failure by fracture even at small deformations. The focus of the presented research is on the development of appropriate numerical methods for predicting the mechanical behaviour and identification of limiting conditions to prevent brittle fracture of Zircaloy claddings. An iterative inverse analysis method is used for deriving the elastic-plastic material properties in the hoop direction of a ring-shaped sample. A modelling approach based on cohesive zones is explained which can reproduce the propagation of cracks initiated at radial hydrides in the zirconium matrix. The developed methods are applied to defueled samples of cladding alloy ZIRLO®, which were subjected to a thermo-mechanical treatment to reorient existing circumferential hydrides to radial hydrides. A selected sample showing sudden load drops during a quasi-static ring compression test is analysed by means of fracture mechanics for illustrative purposes.

2.17 Update of the Experimental Work in the Framework of the SPIZWURZ Project

*Mirko Große, F. Boldt, Michael Herm, Daniel Nahm,
Conrado Roessger, Juri Stuckert, Sarah Weick*

Karlsruher Institut für Technologie (KIT), Germany

The SPIZWURZ project is a cooperation between the German institutions GRS and two sub-institutes of the Karlsruhe Institute of Technology KIT. Several single effect tests are combined with a large scale long-term simulation test. The SPIZWURZ project includes several experiments at the KIT as well as model developments at the 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 on the QUENCH facility will generate hydrogen diffusion data on macroscopic scale. The SPIZWURZ project 1 can be divided into four topics:

A long-term test of a fuel rod simulator bundle (21 electrical heated fuel rod simulators with a length of about 2.5 m) at the KIT-QUENCH facility with a starting temperature of 400°C at the hottest position and a duration of 250 days with a cooling rate of 1 K/day.

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

Determination of the elastic and plastic strain of an original spent fuel cladding tube. 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 on the QUENCH facility will generate hydrogen diffusion and solubility data on macroscopic scale.

Validation and improvements of models describing hydrogen diffusion and solubility in cladding alloys based on the experimental data and implementation in fuel rod performance codes.

The preparation of the large-scale bundle test including the hydrogen loading of the cladding tubes is finished and the test started at May 16th 2023 and will be end at

January 11th 2024. Information about the hydrating process of the fuel rod cladding tubes as well as first online measurements will be given.

For the separate effect tests the equipment and procedures are installed and optimized. First results of the separate effect test were obtained. Details are given in S. Weick et al., Update of neutron radiography experiments in the framework of the SPIZWURZ project. Zirconium single crystals are delivered by MaTeck. First results of the pre-characterization are given. Measurements of real spent fuel have shown that the elastic response 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.

2.18 Update of Neutron Radiography Experiments in the Framework of the SPIZWURZ Project

Sarah Weick, Mirco Grosse, Conrado Roessger, Martin Steinbrueck

Karlsruher Institut für Technologie (KIT), Germany

Internal and external stresses affect zirconium based cladding tubes throughout their slow cooling during interim dry storage. Generally, hydrogen in solid solution follows gradients in temperature, concentration, and stress. Consequently, hydrogen moves from higher to lower temperatures and from lower to higher stresses due to the thermodynamic more favourable conditions. The influence of an applied elastic tensile stress on the hydrogen solubility and diffusion is investigated as a part of the SPIZWURZ project. This project was initiated as a cooperation between the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) and the Karlsruhe Institute of Technology (KIT) in Germany. In the project the flow and the chemical potential of hydrogen in cladding tube materials shall be determined under conditions of long-term interim dry storage of spent fuel elements in transport and storage casks. The INCHAMEL (**In**-situ Neutron Radiography **Chamber** for tests under **Mechanical Load**) facility is a modified mobile tensile testing machine that is used elastically strain zirconium samples under defined temperatures of up to 500 °C while applying neutron radiography. Because of the very low neutron cross section of zirconium, the metal is nearly invisible for neutrons and the contrarily behaving hydrogen that scatters neutrons strongly, appears as dark contrast in neutron images. The samples have different cross section profiles, which amplifies the tensile stress and thus facilitates the observation of hydrogen diffusion. This paper describes neutron radiography experiments in the framework of the SPIZWURZ project under different temperature-time conditions with hydrogenated tensile zirconium samples that simulate stress conditions of cladding tubes during interim dry storage.

**Gesellschaft für Anlagen-
und Reaktorsicherheit
(GRS) gGmbH**

Schwertnergasse 1
50667 Köln

Telefon +49 221 2068-0

Telefax +49 221 2068-888

Boltzmannstraße 14

85748 Garching b. München

Telefon +49 89 32004-0

Telefax +49 89 32004-300

Kurfürstendamm 200

10719 Berlin

Telefon +49 30 88589-0

Telefax +49 30 88589-111

Theodor-Heuss-Straße 4

38122 Braunschweig

Telefon +49 531 8012-0

Telefax +49 531 8012-200

www.grs.de