BRITTLE FAILURE OF SPENT FUEL CLADDINGS UNDER LONG-TERM DRY INTERIM STORAGE CONDITIONS – PRELIMINARY ANALYSIS

U. ZENCKER, K. SIMBRUNER, H. VÖLZKE
Bundesanstalt für Materialforschung und -prüfung (BAM)
Berlin, Germany
Email: uwe.zencker@bam.de

Abstract

The evaluation of cladding integrity is a major issue to be demonstrated in Germany for extended interim storage periods up to 100 years and subsequent transportation considering operational and accidental conditions with respect to reactor operation, cask drying and dry interim storage. The chemical reaction between the zirconium fuel cladding and the cooling water in water-cooled reactors produces hydrogen and zirconium oxide. Hydrogen diffuses into the cladding and precipitates as zirconium hydrides when the solubility limit is reached, preferably oriented in hoop direction. At high temperatures during vacuum drying procedures, the hydrides can dissolve. Over a succeeding period of slow cooling with existing hoop stress the hydrides precipitate again, but partly reoriented along the radial direction of the cladding. This change of microstructure in combination with a decreasing temperature (0.5–2 K/year) during (extended) interim storage and additional mechanical load by handling procedures or under accident conditions could lead to a potential cladding embrittlement and consequently increased failure probability. The current research project BRUZL (Fracture mechanical analysis of spent fuel claddings during long-term dry interim storage) has been launched by BAM to investigate potential spontaneous brittle failure of spent fuel claddings at small deformation under long-term dry interim storage conditions. Based on the key thesis that radial hydrides may be considered as sharp cracks, BAM plans Ring Compression Tests (RCT) with unirradiated cladding samples with representative hydride distribution (including hydride reorientation), numerical simulation of the RCT, calculation of fracture toughness, and identification of failure criteria. Without hydride reorientation, samples under RCT conditions show large plastic deformation with gradually decreasing force at the end of the test indicating ductile failure. Contrary, with hydride reorientation, spontaneous failure with abruptly decreasing force at very small deformation and low temperature is possible dependent on hydrogen content and mechanical load during hydride reorientation.

1. INTRODUCTION

Spent fuel cladding is a thin-walled tube that acts as the first containment barrier of the fuel pellets and fission products. During the fuel cycle, the cladding is subjected to considerable stresses, mainly along the hoop direction due to pellet swelling and fission gases. Therefore, cladding integrity is a major issue.

Zirconium alloys are widely used as cladding materials mainly due to their low cross section absorption of neutrons, mechanical strength and corrosion resistance at operational conditions. During operation in water-cooled reactors, the chemical reaction between zirconium and the cooling water produces hydrogen and zirconium oxide. Hydrogen diffuses into the cladding. When saturation is reached, hydrogen precipitates inside the cladding as zirconium hydrides which appear as thin platelets. These hydrides are aligned in hoop direction because of the microstructure formed during manufacturing of the cladding [1].

In Germany the dry interim storage of spent fuel is preferred. After operation in the reactor and a subsequent period in a holding basin, spent fuel elements are stored in dual purpose transport and storage casks where they are vacuum-dried. The drying procedure may result in a significantly increased temperature up to 370°C and high internal pressure of the fuel rods leading to considerable stress in hoop direction inside the cladding tube. As a result, zirconium hydrides can dissolve again. During the slow cooling process with ongoing hoop stress, hydrides re-precipitate partly in radial direction [2].

It has been shown that hydrides can reduce the ductility of zirconium alloys, an effect also known as hydride embrittlement [1]. A spontaneous failure of the cladding at lower stresses compared with the behaviour of non-hydrided material would be the result. Brittle failure primarily occurs at low temperatures with hydride platelets being aligned perpendicularly to the load the cladding is subjected to. The susceptibility to embrittlement depends on multiple aspects like the total amount of hydrogen being absorbed and the temperature history combined with hoop stress during the drying process. In addition, the distribution of

hydrides, their locations, and their sizes in respect of the cladding geometry play a part. Fig. 1 illustrates a typical hydride distribution in a cladding tube. Near the outer edge there are hydrides of various sizes and orientations in predominantly circumferential direction. A part of the hydrides is reoriented in radial direction. Radial hydrides may be larger. The hydride density strongly depends on the zirconium alloy as well as the mechanical and thermal history.

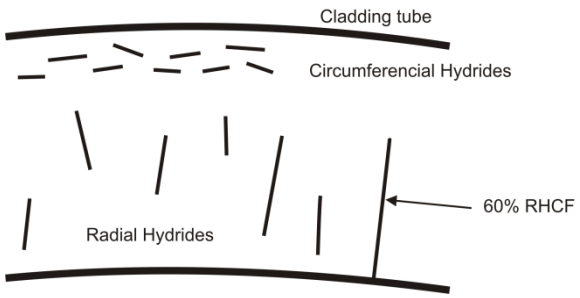

FIG. 1. Typical hydride distribution in a cladding tube.

Further failure mechanisms include creep, delayed hydride cracking (DHC), and stress corrosion cracking (SCC), which have been summarized in Ref. [3]. Creep is expected to occur for long-lasting temperatures above 300°C for loaded cladding material. To assess the risk of failure due to creep, a 1 % strain criterion has been introduced. DHC is a diffusion driven process, at which hydrogen atoms migrate towards existing flaws inside the cladding material. Following precipitation may lead to a growth of the flaws. Because of the decreasing temperature and stresses during long-term storage scenarios, the primary source of failure is expected to be hydride embrittlement in contrast to the other above mentioned phenomena. SCC refers to the process of chemical corrosion at the crack tip. Even at higher stresses than expected, effects of this mechanism seem to be minimal and can be neglected in regard to dry storage.

After long-term interim storage, the mechanical integrity of the cladding tubes should be ensured despite a possible embrittlement while handling, transport, or accidents such as a cask drop. To assess the risk of failure, assessment methods have to be chosen to gain understanding of the behaviour of hydrides in a zirconium matrix. A local damage mechanics approach is proposed by Le Saux et al. [4]. The model uses a modified Gurson-Tvergaard-Needleman model with extensions to incorporate plastic anisotropy and viscoplasticity. Ductile failure was reproduced for temperatures of 25, 350 and 480°C and hydrogen content up to 1200 wppm. Damage nucleation is induced both by hydride cracking as well as the debonding between the matrix and the precipitations. A nucleation rate is deducted from observations of the microstructure. Further parameters are obtained from experiments. Implemented in a finite element (FE) analysis, the model could predict experimental results satisfactorily. Due to the nature of the damage model, the impact of lower hydrogen contents with high degrees of anisotropy were not taken into consideration and could not be modelled.

Assuming hydride platelets can be considered as cracks inside the cladding material, a fracture mechanics approach can be used to address microscopic crack growth as the failure mechanism and consequently investigate the fracture behaviour. Compact Tension (CT) specimens are commonly used for research on fracture behaviour. Using cathodic and gaseous charging processes, studies on hydrogen charged CT specimen show decreasing critical fracture values J_{IC} for increasing hydrogen contents and low temperatures [5]. The macroscopic fracture behaviour was considered being ductile and therefore a transition from ductile to brittle material response could not be noticed. Negative influence on the fracture toughness of zirconium alloys could be confirmed from 25°C up to 300°C and precipitations along grain boundaries have been determined as a reason for the loss in fracture toughness [6]. During a pin loading test performed with hydrogen charged Zircaloy-4 rings, circumferential hydrides reduced the fracture toughness by 20%. Combined radially and circumferentially aligned hydrides could reduce the fracture toughness up to 90% [7]. Even though macroscopic failure appears to be brittle, microscopic crack growth was considered plastic strain driven.

Although common fracture mechanics testing procedures allow the observation of some effects regarding hydride embrittlement, the impact of crack fields remains unanswered. In addition, the result of a CT test with its defined stress state is hardly applicable to the complex structure and geometry of a thin-walled tube.

Therefore, the ring compression test (RCT) was established in order to take into consideration these effects. In this test a cylindrical sample of the cladding material is deformed within movable plates and the loading force vs. displacement curve is obtained. Experimental results from RCTs for different hydride contents and temperatures are used to develop numerical models that help to understand and interpret the micromechanical processes inside the cladding. Based on a predictive simulation, failure criteria for the cladding material can be derived. An experimental procedure was developed for the commercially available alloys ZIRLO™, M5® and Zircaloy-4 (Zry-4) by Billone et al. [8,9]. Both samples from high burn-up fuel rods and as-fabricated, hydrogen charged cladding segments were heated up to 400 °C and pressurized to induce peak hoop stresses from 80 MPa up to 140 MPa to simulate drying and storage. Hydrogen contents ranged from 350 wppm to 650 wppm for ZIRLO™. Afterwards, the ring segments were cooled with cooling rates of essentially 5 K/h. RCTs were performed with as-irradiated as well as hydride-reoriented samples at distinct temperatures from room temperature up to 200°C with a displacement rate of 5 mm/s. The size of radial hydrides was quantified by the radial hydride continuity factor (RHCF) as the ratio of continuous radial hydride length to wall thickness of the cladding, cf. Fig. 1. It could be shown that through-cracks during the tests correlate with the extension of radially aligned hydride formations. Furthermore, the re-precipitation of platelets in radial direction is controlled by the peak hoop stress. The highest testing temperature, at which brittle behaviour occurs, is defined as the ductile-to-brittle transition temperature (DBTT). This temperature depends on the type of material, hydrogen charging, and reorientation stress. The embrittlement criterion is an “offset strain” of 2% [8].

Further RCT procedures were performed by Martin-Rengel et al. [10-11] and Ruiz-Hervías et al. [12]. Ring segments of ZIRLO™ were charged with hydrogen to accomplish contents of up to 2000 wppm. The preparation of heating, re-orientating and cooling was performed in a similar fashion as described above. Two additional failure criteria were proposed based on the strain energy density and the equivalent plastic strain respectively. Furthermore, numerical methods using the finite element (FE) analysis were proposed. A fracture mechanics approach could re-produce load-displacement curves from RCTs using the cohesive crack model and further non-linear optimization could enhance the results [13]. Three point bend (TPB) tests as well as RCTs have been performed by Korinko et al. [14]. Ring samples with reoriented hydrides were used to perform both experiments. It could be shown that the cladding material was less susceptible to brittle failure during the TPB test in comparison with the RCT. Based on the results of 102 RCTs a computational model was developed by Herb et al. [15]. The derived failure criterion considers the ratio of burst stress, i.e. the stress at which the first drop in the load-displacement curve occurs, and the yield stress of the material to predict the ductile-to-brittle transition.

An elastic-plastic fracture mechanics FE simulation model is proposed by Nilsson et al. [16]. A two-dimensional plane strain tube segment was modelled with a suitable constitutive model for Zry-4. Individual straight cracks of different lengths, locations and angles were added to the model. A stress load was applied as a boundary condition and the *J*-integral was calculated. It could be confirmed that circumferentially aligned cracks have no impact on the fracture initiation because *J* values for those cracks were essentially zero. The ligament starts yielding at lower stresses for cracks close to the inner or outer surface. Then, fragmented hydrides were modelled as kinked cracks with angles from -30° to 30° differing from the radial direction. Radial cracks with kinks had the same initiation values for *J* as straight cracks. The likelihood of crack growth is increased, however, for kinked cracks in comparison with straight cracks. Since cladding tubes can contain many hydrides, the distance between the platelets is often smaller than their individual length. Therefore, the complex interaction between cracks was analysed depending on the distance of the crack tips and the angle between neighbouring cracks. For a realistic study, photographs of hydrides were analysed and transferred to an equivalent FE model of a crack field. It was concluded that cladding failure becomes more unstable if several radial hydrides interact. Failure might be triggered by a critical hydride configuration, even if the configuration is very local. Hydride induced failure is strongly influenced by the yielding along the ligaments. The size of the hydrides in radial direction is the most significant parameter for the load necessary to initiate a failure.

In the following, a current research project to investigate spontaneous brittle failure of spent fuel claddings at small deformation under long-term dry interim storage conditions is introduced. Because of the long-term storage, a cladding temperature below the ductile-to-brittle transition temperature is expected. It is important to know which load level for the cladding tubes can be accepted under these conditions without a risk of failure. Cladding tubes made of unirradiated pre-hydrided ZIRLO™ with various configurations of radial hydrides are exemplarily investigated. The experimental set-up is a ring compression test. This test scenario is

simulated with the FE method. At the current project stage a computational model is presented, capable of reproducing the results from RCTs with non-hydrided samples. This model will be enriched with cracks to take account of various hydride configurations. The methodology for the failure analysis based on fracture mechanics will be explained. Aim of the project is the development of safety assessment methods based on a characterisation of the cladding tube including hydrides, applied load, and suitable failure criteria.

2. RING COMPRESSION TEST

The ring compression test is the established experimental method for the investigation and characterisation of the mechanical behaviour of defueled cladding tubes. The test can be carried out in hot cells with irradiated material as well as with unirradiated material under normal laboratory conditions. The effort for hot cell tests is very high, but investigations outside of hot cells require unirradiated mock-up cladding tubes with material behaviour representative for the irradiated material state. Real zircaloy claddings contain circumferential and radial hydrides due to the conditions in water-cooled reactors and subsequent vacuum-drying. Therefore, the influence of these hydrides on the material behaviour must also be emulated.

Realistic distributions of circumferentially or radially oriented hydrides can be created by hydrogen charging of unirradiated zircaloy claddings. Ruiz-Hervías [12] describes a cathodic charging method for a controlled insertion of hydrogen into an unirradiated sample. In an electrochemical reaction the sample acts as the cathode and a platinum wire coiled around the sample as the anode in a KOH aqueous solution. After hydrogen charging, the sample is exposed to an inert gas atmosphere with constant elevated temperature and then slowly cooled down (approx. 1.2 K/min). The hydrides are circumferentially oriented at this stage of the procedure. Based on a subsequent thermo-mechanical treatment with a hoop stress (or reorientation stress resp.), the hydrides can be dissolved and partly reoriented along the radial direction of the cladding. The resulting hydride density and orientation is similar to radial hydrides found in the cladding of irradiated fuel rods after radial-hydride treatment (cf. Fig. 1). Considering zircaloy M5® mostly individual long radial hydrides precipitate. For ZIRLO™ typically a mix of radial and circumferential hydrides is predominant.

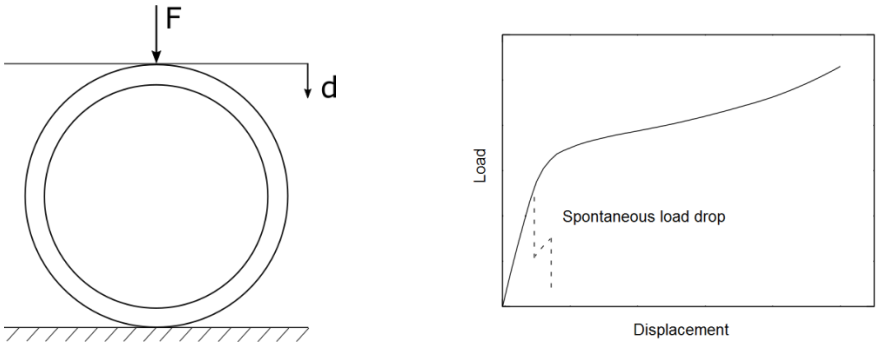

FIG. 2. Ring compression test: Experimental set-up and typical load-displacement curve.

The experimental set-up of the ring compression test is schematically shown in Fig. 2 on the left hand side. Tests are conducted displacement-controlled with displacement *d* by means of two plane and parallel steel plates. The load *F* is measured with a load cell. The upper plate is moving with constant velocity, the lower one is fixed. Maximum tensile hoop stresses occur at the inner surface of the 6 and 12 o'clock positions and at the outer surface of the 3 and 9 o'clock positions. Specimens have the shape of rings with a height of 10 mm to ensure plane strain conditions during the test. The dimensions of the cladding tube may vary. Typical values are an outer diameter of 9.5 mm and a wall thickness of 0.5 mm.

Fig. 2 illustrates on the right hand side the characteristic load-displacement curve of pre-hydrided samples. Cladding samples with circumferentially oriented hydrides typically show ductile behaviour during a RCT. The displacement at maximum load is much larger than the typical gap between tube and fuel. After large deformation a ductile failure is found and the load decreases gradually (not shown in the figure). Contrary, hydrides oriented in radial direction of the cladding can cause brittle failure associated with spontaneous load

drops. The displacements at point of failure can be very small. After the maximum load, failure by crack initiation and subsequent crack propagation is observed. The crack can also stop and reinitiate. In this case the load can decrease in several steps.

3. NUMERICAL SIMULATION

The ring compression test is numerically simulated to understand the complex stress state within a ring segment. At the beginning, tests of samples without hydrides or with hydrides only in circumferential direction were investigated. In these cases the ring segments deform elastically-plastically up to large deformations, i.e. an abrupt load drop due to brittle failure at relatively small deformations is not expected.

The specimen has a length of 10 mm, outer diameter of 9.5 mm and wall thickness of 0.5 mm. Therefore, plane strain conditions may be assumed for the mid-plane of the specimen. Fig. 3 shows the generated two-dimensional finite element mesh with a detail of the mesh at the 12 o'clock position. Finite elements with eight nodes, biquadratic displacement and reduced integration were used. At the bottom, the specimen is supported by a rigid body. At the top, another rigid body pushes with defined displacement onto the specimen. A hard contact with friction (friction coefficient 0.15) was implemented between specimen and rigid bodies. The load is static. Commercial finite element code ABAQUS™ version 6.14 was used for all calculations.

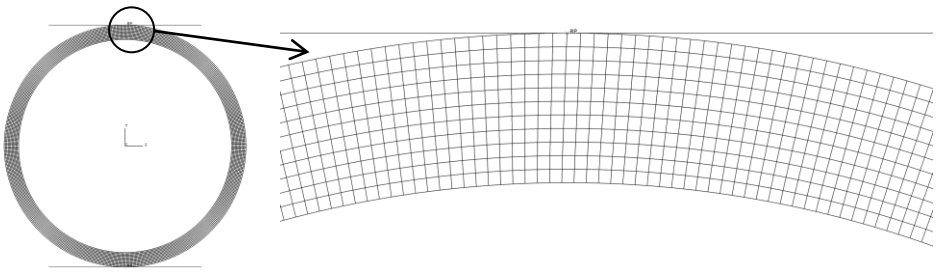

FIG. 3. Finite element model.

The investigations were carried out exemplarily for ZIRLO™ samples. The defined material model is essential for the accuracy of the simulation results. The material behaviour is assumed as elastic-plastic and isotropic with von Mises yield criterion. Even a linear strain hardening with yield stress of 775 MPa and flow stress of 1075 MPa at plastic strain of 0.3 gives acceptable results at room temperature. To improve the results a more accurate flow curve was taken from Gomez [17] based on an inverse method to determine the plastic stress-strain curve from non-conventional tests. Young's modulus of 88 GPa and Poisson's ratio of 0.37 describe the elastic part of the material behaviour [11].

The influence of the fineness of the finite element mesh was investigated. A mesh with only 8 elements in thickness direction contrary to the mesh in Fig. 3 and the same element type produces similar results. An increased number of elements (19 elements) over the wall thickness generates only slightly differing results as well. The use of an element type with biquadratic displacement is recommended however.

The simulated behaviour of ZIRLO™ samples without reoriented hydrides was compared to experimental results. Fig. 4 shows the measured load-displacement curve from Gomez [17] denoted as "UPM Zirlo" (black line). The curve "FE model" (red line) represents the simulation result calculated by means of the finite element model described above. An excellent coincidence is obvious. This example illustrates on the one hand the possible accuracy of the numerical simulation result if a suitable description of the constitutive behaviour is available. Here the stress-strain curve and the load-displacement curve were taken from the same reference [17]. On the other hand, it must be emphasized that a reliable stress-strain curve must be derived with some effort for a given set of experimental parameters. As a consequence, the investigation of samples with reoriented hydrides and failure at small deformations requires additionally the measurement of load-displacement curves without hydride reorientation until large deformations to derive a stress-strain curve as basis for an elastic-plastic material model in the finite element code.

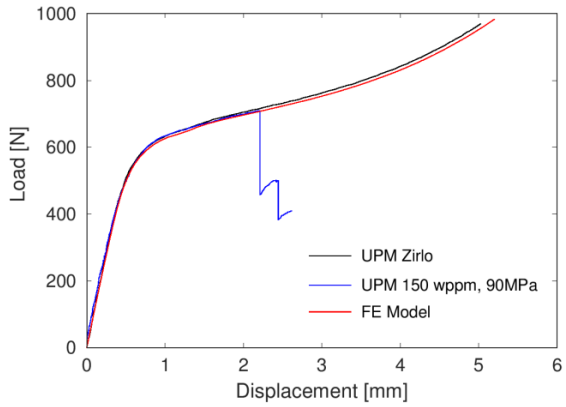

FIG. 4. Load-displacement curve at room temperature.

4. SAFETY ASSESSMENT

In most cases for circumferential hydrides in pre-hydrided samples of ZIRLO™ it is observed that around the maximum load during the RCT a crack is initiated at the surface of the specimen in the 3 or 9 o'clock position which grows in length direction while the load is decreasing gradually, i.e. ductile failure is the prevailing failure process. At a temperature of 20 °C, ductile behaviour was observed in specimens pre-hydrided with 150 wppm hydrogen only for reorientation stresses smaller than 90 MPa (Fig. 4). For higher reorientation stresses, specimens sometimes fail in the initial elastic region or just shortly after passing this region. Around 90 MPa a transition region from ductile to brittle behaviour seems to exist [12]. Despite the existence of reoriented hydrides, the load-displacement curve is practically identical to the curve without reoriented hydrides. The only difference is the abrupt load drop at the moment of brittle failure as illustrated by the curve "UPM 150 wppm, 90 MPa" (blue line) in Fig. 4. Because of the nature of spontaneous brittle failure, in a re-examination of the test another sample could fail at another load. Until failure, however, there is only a small scatter between the individual curves.

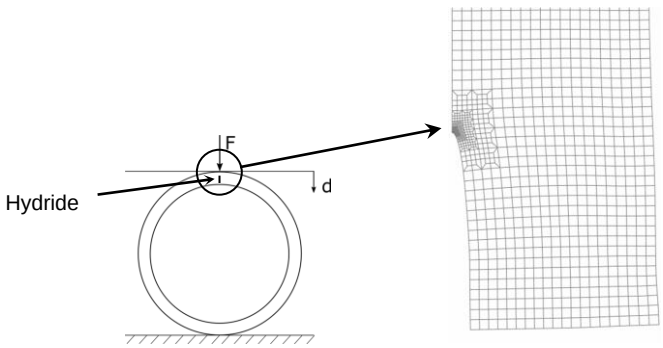

FIG. 5. Fracture mechanical assessment concept.

Hence, the brittle failure can be simulated based on a finite element model as presented above with additionally modelled hydrides in radial direction. Radially oriented hydrides are considered as sharp cracks. Then, the well-known methods of fracture mechanics are applicable. Fig. 5 illustrates the finite element model for a single hydride with a crack tip at its end located at the 12 o'clock position near the inner surface where tension stress prevails. The detail shows the mesh at the crack tip. The crack-tip parameter (*J*-integral) is calculated dependent on the load. When the moment of failure is known from the test, the corresponding *J*-

integral value is a measure for the initiation fracture toughness. Of course, in a real sample there is not only an individual hydride but a field of hydrides or cracks respectively. Consequently the interaction between hydrides must be analysed to isolate the material property from additional interaction effects. In this sense the planned work is an extension to the study of Nilsson [16].

The focus is on the calculation of the initiation fracture toughness at the initial elastic region or just shortly after passing this region in the RCT on the basis of measured load-displacement curves, correlated observed distribution of hydrides and postulated initial macroscopic cracks. Various failure criteria can be found in literature depending on an "offset strain", strain energy density, or equivalent plastic strain [8,9,12]. A fracture mechanical analysis of the sudden failure by brittle fracture may help to understand the underlying processes and to develop appropriate failure criteria based on the initiation fracture toughness of the material. In this sense, the observed reloading indicated by the blue curve in Fig. 4 could be explained by crack jumping.

A fracture mechanics based failure criterion for brittle fracture of fuel rods at small deformation in terms of material's fracture toughness for pre-hydrided zircaloy samples with reoriented hydrides could help to quantify the failure probability dependent on the hydride distribution (size, orientation, and density), the material properties of the cladding tube, temperature, and additional mechanical load from handling, transport or accidents.

5. CONCLUSIONS

A current research project concerning the fracture mechanical analysis of spent fuel claddings during long-term dry interim storage is presented. Spontaneous brittle failure at small deformation < 2 mm during ring compression tests is of special interest with regard to fuel rods filled with pellets under normal or accidental conditions during handling and long-term dry interim storage.

Ring compression tests with specimens containing hydrides only in circumferential direction were numerically simulated so far. To account for brittle failure, radial hydrides are considered as cracks and added to the developed finite element model. Because the hydrides are often located close together, an investigation of hydride interactions within hydride fields is planned. Next, ring compression tests with unirradiated cladding samples with representative hydride distribution including hydride reorientation will be performed as part of this project.

Brittle failure can be excluded by an operating temperature above the ductile-to-brittle transition temperature. After extended long-term dry interim storage the cladding temperature might be so low that a handling of embrittled cladding is necessary. Based on knowledge about the properties of the cladding material including hydride distribution (size, orientation, and density) it should be possible to quantify permissible mechanical loads to prevent brittle failure or to estimate the failure probability.

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