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| Hydride effect on cladding behaviour for spent fuel storage and transport conditions |
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**Abstract.** Morphology and distribution of hydrogen picked-up during in-reactor operation could change due to the evolution of fuel conditions (pressure, temperature, etc) from pool to dry storage. The possible dissolution and re-precipitation of the hydrides in the radial direction could provoke an embrittlement of the cladding.

As the clad behaviour is a key factor during spent fuel dry storage and transport, Spanish organizations CSN, ENRESA and ENUSA have carried out research and development programs to characterize the performance of fuel under these conditions.

The mechanical and fracture behaviour of fresh cladding electrochemically charged at different Hydrogen concentrations has been studied using Ring Compression Tests (RCT). The tests have been performed at different strain rates and temperatures representatives of dry storage and transport conditions.

Additionally a finite element model, based on the experimental RCT load vs. displacement curves has been developed to calculate the fracture energy as a function of the hydrogen concentration.

The results show that samples with homogeneous and circumferential hydrogen distribution, do not present brittle behaviour. Rupture is produced at displacements higher than 3 mm. For radial hydride reoriented samples brittle fracture are obtained for low displacement values at 20°C and 135°C.

# Introduction

The strategy followed in Spain for the spent fuel management set in the Radioactive Waste General Plan, which is currently in its 6th revision (approved in 2006 by the Spanish Government) is the open cycle. The spent fuel is on-site stored in spent fuel pool and in Interim Dry Storage when the pool capacity is over. Additionally, a Centralized Interim Spent Fuel Storage Facility has been approved and it is under design. The installation is based in a vault system, where the fuel assemblies are stored in capsules located in wells and cooled by natural convection. This installation is expected to be operative in 2018, therefore, spent fuel transports will be performed soon in Spain.

The safety functions (subcriticality, confinement, retrievability, heat removal and shielding) should be assured during storage and transport. Spanish Nuclear Authority (Consejo de Seguridad Nuclear-CSN) has published Safety Instructions regarding spent fuel storage and is developing specific regulations for spent fuel transportation, all of them consistent with the US-NRC regulations, 10CFR71 and 10 CFR 72. Cladding is the first barrier for the fissile material, and their properties and behaviour under dry storage and transport conditions (temperature and stress) are crucial to assure the requirements indicated in the standards [1] [2]:

* 10 CFR 72.122(h)(1): “The spent fuel cladding must be protected during storage against degradation that leads to gross ruptures or the fuel must be otherwise confined such that degradation of the fuel during storage will not pose operational safety problems with respect to its removal from storage".
* 10 CFR 71.71 ‘Normal conditions of transport’: "The geometric form of the package contents would not be substantially altered.”

Under normal storage conditions, the major degradation mechanisms of Spent Nuclear Fuel (SNF) cladding that have been hypothesized to lead to failure include creep and hydrogen induced mechanisms [3]. During fuel irradiation hydrogen is absorbed in the cladding and, when hydrogen concentration is high enough, it precipitates as platelets of zirconium hydrides, that produces an embrittlement of the cladding [4]. The temperature and stress evolution during dry storage, may produce a hydride re-precipitation in the radial direction, as a consequence of the temperature reduction under stress. In those conditions, the clad could become brittle.

During transport the fuel rod integrity should be maintained considering the stresses transmitted to the fuel assembly and to the rods, as the bending loads supported by the rod material between grids, the pinch loads transmitted by the grid to the rod at the rod/grid contact point and vibration loads. Additionally, it should be taken into account that the transport can be performed at lower temperatures, with lower cladding ductility as temperature decreases.

Therefore, Spanish organizations, CSN, ENRESA and ENUSA have carried out several test programs to characterize the behavior of spent fuel rod under dry storage and transport conditions. In the past, the programs were focused on creep behavior of PWR and BWR fuel clad material [5] [6]. In this paper the results of a project focused on the effect of the hydrogen content and morphology in the cladding mechanical behavior are presented.

# Objective

The aim of this project is to know the mechanical behaviour of the cladding material in storage and transport conditions. The effect of the hydrides, the mechanical and fracture properties of the cladding as a function of the hydrogen content, hydrogen morphology and testing temperature, and the parameters that could affect to these characteristics, could affect the material behaviour

# Experimental results

The material used for this work is unirradiated PWR cladding (ZIRLOTM alloy [7]) in Stress Relief condition with 9.5 mm outer diameter and 0.57 mm wall thickness. The material had been prehidrurated with different hydrogen content (representative of irradiated material and above) and different hydrides morphology (circumferential and radial hydrides). After that, it has been mechanically tested and analyzed. A FEM analytical model and a failure criterion has been developed.

Test have been performed at high temperatures, representatives of dry storage conditions, and lower temperatures representatives of transport conditions. Additionally, room temperature has also been analyzed, to know the behavior of the fuel after extended periods of storage and for theoretical transport after long time storage.

## Cladding hidruration

Hydrogen was introduced via cathodic charge technique in KOH aqueous solutions. Samples with controlled amounts of hydrogen (150 to 2000 ppm) were obtained by this process. Further details about the charging method and sample preparation are given in [8].

After a thermal treatment, a homogeneous distribution with hydrides oriented along the hoop direction of the samples was obtained (Fig. 1).

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| FIG. 1. Samples with 150 (a), 500(b) and 1200(c) ppm of hydrogen. |

## Hydrides Reorientation

Samples with homogeneous hydride distribution and concentrations of 150, 500 and 1200 ppm of hydrogen were subjected to a thermomechanical treatment to produce the dissolution and reorientation of the hydrides[8]. The population of radial hydrides is a function of the hydrogen concentration and the hoop stress applied during cooling down. Figure 2 shows samples with 500 ppm of hydrogen subjected to different reorientation hoop stresses: (a) 60 MPa, (b) 90 MPa, (c) 120 MPa and (d) 140 MPa.

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| FIG. 2. Samples with 500 ppm of hydrogen subjected to hoop stress values of 60 MPa (a), 90 MPa (b), 120 MPa (c) and 140 MPa |

The hydride reorientation factor Fn (reoriented hydride percentage) has been calculated according to ASTM B811 [9]. For hoop stresses lower than 60 MPa, the reoriented percentage is lower than 3% for all the hydrogen concentrations. A relevant increase is produced for 90 MPa, and this tendency continues for higher hoop stresses. It seems that the threshold hoop stress for hydride reorientation should be around 90 MPa. This value is coherent with other authors results[10] and with the limit imposed by the NRC to avoid radial hydrides in the Interim Staff Guidance ISG-11[11].

Figure 3 shows samples with (a) 150, (b) 500 and (c)1200 ppm of hydrogen. All of them were subjected to a hoop stress of 90 MPa.

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| FIG. 3. Samples with (a) 150, (b) 500 and (c) 1200 ppm of hydrogen and a 90 MPa reorientation hoop stress. |

The analysis of samples with different hydride concentration indicates that the fraction of reoriented hydrides is greater in the sample with 150 ppm of hydrogen, and the radial hydrides are longer than those of 500 and 1200 ppm samples(figs. 3b, 3c).

## Ring Compression Tests

The sample evolution and the Load-Displacement representative curve for RCT are represented in Fig. 4.

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| FIG. 4. Representative RCT evolution |

The RCT performed could be divided into two groups, representative of different irradiated material condition: samples with circumferential hydrides and samples with radial hydrides.

**Samples with circumferential hydrides** tests were carried out at different hydrogen concentrations (0 to 2000 ppm); different temperatures (20, 135 and 300 °C) and displacement rates of 0.5 and 100 mm/min.

The results obtained for all the temperatures indicate displacements higher than 2 mm previous to the point of crack initiation. Besides, while for higher hydrogen concentrations, the displacement where cracks are initiated is lower, for some other material properties, as Yield Strength and Young Modulus, do not show a straight dependence of the hydrogen content (Fig.5).

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| FIG. 5. Load-Displacement curve for different hydrogen concentration, 20ºC and 0.5 mm/min |

For these samples with circumferential hydrides, a 2D finite element model was developed with ABAQUS v 6.7–5 commercial code to simulate RCT results. The results of the numerical modelling versus the experimental results (one example in Fig. 6) show that the whole load vs. displacement curve is correctly predicted, including the downloading after the maximum load.

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| FIG. 6. Experimental vs numerical Load-Displacement curve |

With this model, the mechanical properties and the fracture energy are calculated based on the experimental load vs. displacement curves.

Additionally, to simulate the impact loadings corresponding to cask drop transport accident conditions, some tests have been performed at very high rate of displacement, 3 m/min. For all the temperatures, the obtained load-displacement curves are similar to those obtained at lower displacement rates with higher Yield Strength and Young Modulus, however the rupture of the material is produced at lower displacement values (3 mm vs 4 mm).

**Samples with radial hydrides** tests were performed for 150, 500 and 1200 ppm of hydrogen that have been reoriented with hoop stresses of 60, 90, 120 and 140 MPa. The test temperatures were 20, 135 and 300ºC, and the displacement rate has been 0.5 mm/min.

At the highest temperature, 300ºC, there is no remarkable effect of radial hydrides on material behaviour (Fig. 7)

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| FIG. 7. Load-displacement curve for different reorientation hoop stress (300ºC-500 ppm H) |

At 135ºC, it seems that there is no effect of the radial hydrides for samples with higher hydrogen concentrations (500, 1200 ppm). However for samples with 150 ppm hydrogen, there is an important effect of the radial hydrides, as the rupture of the sample is produced at very low displacement values when the hoop stress used for the hydride reorientation is 140 MPa (Fig. 8). This result is related with the reoriented hydride characteristics indicated before, higher Fn and higher hydride lengths, and is coherent with data published by other authors. [12]

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| FIG. 8. . Load (kN)-displacement (mm) curve for different reorientation hoop stress (135ºC-150 ppm H) |

For the test performed at lower temperature, 20ºC, there is an important effect of the radial hydrides. For reorientation hoop stresses equal or higher than 90 MPa, the sample rupture is produced at displacement values lower than for the sample with no radial hydrides. For higher stresses during reorientation, the rupture is produced in the elastic part of the test (Fig. 9).

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| FIG. 9. Load (kN) -displacement (mm) curve for different reorientation hoop stress (20ºC-150 ppm H) |

## Fractographic analysis

A detailed fractographic analysis to identify the fracture micromechanisms has been performed.

The fracture surfaces were observed by Scanning Electron Microscopy (SEM).

The results are coherent with the RCT data. For 1200 and 2000 ppm samples, there are different behaviours depending on temperature. For 20 and 135 ºC fracture mechanism is ductile to some extent, with crystallographic planes and local orientations which suggest a brittle fracture in some parts of the sample. For 300ºC a ductile behaviour at the microscopic level is observed everywhere.

## Failure criteria

To assess the experimental results, three different failure criteria has been considered: (1) the offset strain criteria proposed by Billone [12] δp/D0 < 2% ; (2) SED at maximum load < 25 N/mm2 and (3) plastic deformation Ɛp < 3% . If any of these conditions is fulfilled, the rupture has been considered brittle.

Applying these criteria for the radial hydrides samples, the following could be concluded:

The behaviour of the samples tested at 135 and 300ºC is ductile, even for the maximum hoop stress used for the reorientation. Only for one case, 150 ppm hydrogen and 140 MPa reorientation hoop stress, it could be considered in the ductile-brittle transition region.

The samples tested at 20ºC are ductile only for reorientation hoop stresses lower than 90 MPa, for any Hydrogen concentration.

# Conclusions

The Ring Compression Test has been used to obtain the fracture properties of hydrided unirradiated ZIRLO cladding.

A finite element model has been developed to simulate the ring compression test. A very good agreement between the simulations and the experimental results has been obtained.

For homogeneous and circumferential hydride distribution, the behaviour of the material is always ductile; even for the highest hydrogen concentration (2000ppm) and the lowest temperature (20ºC).

For samples with radial reoriented hydrides, the hydrogen concentration, the reoriented hydride percentage (related with the hoop stress), and the test temperature are key parameters that affect the material behaviour. At room temperature and 150 ppm of hydrogen, the rupture is produced at low displacements for every reorientation stresses, and there is a better behaviour for 500 and 1200 ppm hydrogen.

# Discusion

The different R&D projects performed during years for the Spanish organizations, CSN, ENRESA & ENUSA, provide interesting data regarding the cladding material behaviour during dry storage and transport. Creep tests performed in the past with irradiated ZIRLO samples result in very good creep behaviour. In the only test that led to failure among the test matrix performed, 17% strain was needed (at constant inner pressure) for cladding rupture.

The results of the project presented here are also very important related to the spent fuel dry storage and transport. It should be marked that for circumferential hydrides, the cladding material behaviour is always ductile, even at room temperatures. Therefore any condition to avoid the radial precipitation of the hydrides during storage will assure a good behaviour of the cladding. For material with radial hydrides, the temperature and the hydrogen content are crucial; at low temperature the material behaviour is brittle. Additionally, for lower hydrogen content, the percentage of radial hydrides is higher and the material is more brittle, this could be related with the radial-hydride-continuity factor (RHCF) [12].

All of these results are coherent with other authors published data for non irradiated material.

Acknowledgements

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