



# Study of the hoop fracture behaviour of nuclear fuel cladding from ring compression tests by means of non-linear optimization techniques



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## ABSTRACT

In this work, the hoop fracture toughness of ZIRLO<sup>®</sup> fuel cladding is calculated as a function of three parameters: hydrogen concentration, temperature and displacement rate. To this end, pre-hydrided samples with nominal hydrogen concentrations of 0 (as-received), 150, 250, 500, 1200 and 2000 ppm were prepared. Hydrogen was precipitated as zirconium hydrides in the shape of platelets oriented along the hoop direction. Ring Compression Tests (RCTs) were conducted at three temperatures (20, 135 and 300 °C) and two displacement rates (0.5 and 100 mm/min). A new method has been proposed in this paper which allows the determination of fracture toughness from ring compression tests. The proposed method combines the experimental results, the cohesive crack model, finite elements simulations, numerical calculations and non-linear optimization techniques. The parameters of the cohesive crack model were calculated by minimizing the difference between the experimental data and the numerical results. An almost perfect fitting of the experimental results is achieved by this method. In addition, an estimation of the error in the calculated fracture toughness is also provided.

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## 1. Introduction

Nuclear fuel cladding is a thin-walled tube which contains the fuel pellets and the fission products. The cladding is made of zirconium alloys due to its neutron flux transparency, remarkable mechanical properties and good corrosion resistance at operation conditions.

In light-water reactors (LWR), the chemical reaction that takes place between the cooling water and the zirconium of the cladding produce hydrogen. A part of this hydrogen is absorbed by the cladding, precipitating as zirconium hydrides when its solubility limit is reached. These hydrides may degrade the fuel cladding, reducing its mechanical and fracture properties. This reduction of cladding properties will depend on the hydride concentration but also strongly depends on the orientation of the hydrides in the cladding. It is well known that the hydrides which adopt a radial orientation in the cladding, may reduce the fracture properties of

the cladding more than the ones that adopt a hoop orientation [1–12].

During the different steps of the fuel cycle, the cladding is subjected to mechanical stresses, mainly in the hoop direction. Given that cladding is the first confinement barrier for the nuclear fuel and fission gases, its structural integrity must be guaranteed along the fuel cycle. Consequently, there is a need to characterize the fracture behaviour of the nuclear fuel cladding to ensure a safe operation even under extreme events and during storage and transport.

Due to its manufacturing process, the fuel cladding properties are anisotropic. It means that its properties will not be the same in the axial and hoop directions [13]. The behaviour of the cracks is different in axial or circumferential direction. Due to its complex geometry (thin-walled tube), it is not possible to determine the fracture properties of the fuel cladding by means of standard methods. The size of the cracks is also extremely small. The associated fracture toughness is then rather plane stress fracture toughness than plane strain fracture toughness.

Grigoriev et al. [14] obtained the fracture toughness, in the axial direction, of irradiated Zircaloy-2 cladding by means of the Pin-

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Loading (PL) tension test. They reported a maximum-load fracture toughness of about  $100 \text{ MPa m}^{0.5}$  at  $300 \text{ }^\circ\text{C}$ .

Raynaud et al. [15] reported values from  $45 \text{ MPa m}^{0.5}$  to  $10\text{--}15 \text{ MPa m}^{0.5}$  as a function of hydrogen content and radial hydride fraction for pre-hydrided Zircaloy-4 sheets at  $25 \text{ }^\circ\text{C}$ . In the same work, these authors reported values  $\geq 50 \text{ MPa m}^{0.5}$  at  $300 \text{ }^\circ\text{C}$  and  $>55 \text{ MPa m}^{0.5}$  at  $350 \text{ }^\circ\text{C}$  for the same material.

In a previous work [6], the authors determined the hoop fracture properties of the fuel cladding from the RCTs. RCT simulates pinch-type loading at grid spacers. Fuel cladding samples with hydrogen contents up to 2000 ppm were employed. Hydrogen was precipitated as  $\delta$ -hydride platelets distributed along the hoop direction of cladding [4]. A crack, which propagates in a stable mode, is generated at the end of this test. The failure process was simulated with the cohesive crack model by using finite elements and the fracture energy was calculated from the model parameters by a trial and error method.

This method was time-consuming and relied on empirical adjustment of the fitting parameters. In the present work, an original process to calculate the fracture energy from ring compression tests is reported. As in our previous works, the cohesive crack model was employed to simulate the damage process. The parameters of the cohesive model have been automatically adjusted to minimize the difference between the numerical results and experimental data, with an excellent agreement between them. The fracture energy along the hoop direction was calculated from the parameters of the cohesive crack model, and the fracture toughness was obtained from the fracture energy. The fracture toughness was calculated as a function of three parameters: hydrogen content, temperature and displacement rate. In addition, a procedure which allows to estimate the error of the proposed method is also reported in this paper. The fracture energy has been obtained by means of an inverse procedure that combines experiments, numerical calculations and optimization algorithms. To evaluate the reliability of the method and compare it with other experimental techniques it is necessary to assess the error introduced. The final result is a toughness value with an error band.

Samples with nominal hydrogen contents of: 0 (as received), 150, 250, 500, 1200 and 2000 ppm of hydrogen were employed in this study. In all the cases, the hydrogen was precipitated as  $\delta$ -hydride platelets distributed along the hoop direction of the cladding. It is important to underline that irradiated ZIRLO<sup>®</sup> cladding shows several features that are not reproduced by the samples prepared for this work: presence of a hydride rim, oxide layer at the outer surface of the cladding and irradiation-induced defects. It is well known that the irradiation may reduce the fracture behaviour of the cladding. On the other hand, the part of the cladding formed by the oxide layer and the hydride rim is a possible source of flaws which may favour the initiation of cracks. The zone below the hydride rim contains much less hydrogen concentration than the average hydrogen content, as a consequence, the crack propagation for this zone may be more difficult. For all of these reasons, the values obtained in this paper cannot be directly accepted as totally valid for High-Burnup ZIRLO<sup>®</sup>.

For each concentration, RCTs were conducted at three different temperatures (20, 135 and  $300 \text{ }^\circ\text{C}$ ) and employing two different displacement rates: 0.5, 100 mm/min.

## 2. Material and experimental programme

The material employed in this work is ZIRLO<sup>®</sup> PWR cladding [16] with 9.5 mm outer diameter and 0.57 mm wall thickness. RCT samples with 10 mm height were cut from the as-received cladding. This dimension was chosen to ensure plane strain conditions during the tests.

Controlled amounts of hydrogen were introduced in the ring samples by cathodic charging in KOH aqueous solutions. The solution temperature was maintained between 299 K and 353 K depending on the intended hydrogen concentration. The ring sample was used as cathode of the electrochemical reaction. A platinum wire coiled around the sample was the anode. The current density and the charging time were varied as a function of the hydrogen content.

After cathodic charging, an homogenization process consisting in keeping the samples 7 h at  $450 \text{ }^\circ\text{C}$  in Ar atmosphere was applied to the hydrided samples (from 150 to 2000 ppm). In order to ensure that the proposed heat treatment does not modify the properties of the samples, in previous works [4], the authors conducted RCTs on as-fabricated samples and on as-fabricated samples subjected to the mentioned homogenization heat treatment (without cathodic charging). Both kind of samples shown the same behaviour.

Hydrogen concentration was measured using the inert gas fusion thermal conductivity detection method. In this way, samples with nominal concentrations of 0 (as received), 150, 250, 500, 1200 and 2000 ppm were prepared. The uncertainty on the measured hydrogen content is estimated to be around 10% based on the authors' experience and previous results. The hydride morphology and distribution was studied by metallography. The hydrogen charging method produced a homogeneous hydride population oriented along the hoop direction of the samples, as it can be observed in Fig. 1. Further details of the hydrogen charging method are given in Ref. [4].

The experimental programme consisted of ring compression tests conducted on samples with the aforementioned concentrations, at three different temperatures ( $20 \text{ }^\circ\text{C}$ ,  $135 \text{ }^\circ\text{C}$  and  $300 \text{ }^\circ\text{C}$ ) and two displacement rates (0.5 mm/min and 100 mm/min).

Mechanical tests were performed with an INSTRON 8801 testing machine. Load was measured with a load cell of 5 kN capacity. Compression load was applied by means of two steel plates (plane and parallel). The relative displacement of the loading plates was measured with a LVDT of  $\pm 5 \text{ mm}$  travel. Tests were carried out by applying a constant displacement rate of 0.5 and 100 mm/min. As the stiffness of the sample is much lower than that of the loading train, the displacement measured with the LVDT is the same as the one measured by the machine actuator. Consequently, the LVDT was not used at high temperature ( $135 \text{ }^\circ\text{C}$  and  $300 \text{ }^\circ\text{C}$ ). Tests at high temperature were performed inside a resistance furnace, and the temperature was measured with a thermocouple located very close to the sample, while at the same time one thermocouple was in contact with each compression plate. In addition, a previous calibration of the RCT sample temperature as a function of the temperature of the compression plates was done. For each hydrogen concentration, three tests were conducted at three temperatures (20, 135 and  $300 \text{ }^\circ\text{C}$ ) and two displacement rates of the plates of 0.5 and 100 mm/min.

The experimental load vs. displacement data are shown in Fig. 2 for all hydrogen concentrations, temperatures and displacement rates studied. Each load-displacement curve is the average corresponding with the three tests performed for each condition described. As can be seen in the figures, the load is initially proportional to the displacement and in a second phase continues to increase with a smaller slope until the maximum load is reached. After that point, the load decreases in a stable way. It has been observed that around the maximum load a crack is initiated at the external surface of the sample and propagates along the axial direction. At the end of the test, the load increases due to the fact that each side of the deformed specimen come into contact with each other (Fig. 3).

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