Fracture toughness of Zircaloy-4 cladding in case of Delayed Hydride Cracking

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Abstract
Spent nuclear fuels are stored after their use in reactors. Dry storage can favor the appearance in the fuel rod cladding of a mechanical-chemical phenomenon referred to as Delayed Hydride Cracking (DHC). DHC is divided into three iterative steps: (i) diffusion of hydrogen in solid solution; (ii) precipitation of this hydrogen into hydrides; (iii) brittle fracture of hydrides. To assess the risk of occurrence of this phenomenon, the fracture toughness is determined by calculating the stress intensity factor below which DHC is not observed ($K_{I_{DHC}}$) based on an experimental procedure and a numerical model.

1. Introduction
Fuel rod cladding tubes are made of zirconium alloys and constitute the first safety barrier of nuclear-pressurized water reactors (PWRs). After their use in the reactor, spent fuel rods are stored. The dry storage method consists in placing the fuel assemblies in containers filled with an inert gas. The cooling lasts several decades with conditions that could favor the appearance of a phenomenon referred to as Delayed Hydride Cracking (DHC). This chemical-mechanical phenomenon is composed of three iterative steps: (i) stress-induced diffusion of hydrogen in solid solution at a pre-existing crack tip; (ii) increase of hydrogen at the crack tip up to the Terminal Solid Solubility in Precipitation (C_{TSSP}) and precipitation into radial hydrides; (iii) hydrides growth. If, for sufficiently large hydrides, the stress intensity factor reaches a critical stress intensity factor referred to as threshold stress intensity factor for DHC $K_{I_{DHC}}$ cracking occurs over the embrittled area. The repetition of those three steps leads to brittle crack propagation until the ductile fracture toughness K_{IC} of the material is reached. This study presents a combined experimental-numerical approach to evaluate the fracture toughness in the case of DHC in Zircaloy-4 fuel rod claddings and to provide a quantitative model for crack extension.

2. Fracture toughness of Zircaloy-4 in case of DHC
Fracture toughness is calculated thanks to an experimental procedure and a finite element method (FEM) model. The G-θ method is used to evaluate the stress intensity factor. The determination of $K_{I_{DHC}}$ is divided into three steps:

a. Precracking of the specimen using DHC: specimens are loaded at a constant displacement and placed in an oven. A thermal cycle is performed to maximize the solute hydrogen content. Hydrogen in solid solution diffuses at the notch tip and precipitates into radial hydrides. The imposed displacement is chosen to be large enough to initiate cracking. The stress intensity factor decreases during the crack propagation down to $K_{I_{DHC}}$: the crack propagation stops, and the specimen is considered precracked (Figure 1a).
b. DHC test on precracked specimens: an imposed mechanical load at a constant temperature is imposed after a thermal cycle to put hydrogen back into solid solution. If nothing happens, the test is stopped after 24 hours (the loaded $K_{I_{test}}$ is under the threshold value $K_{I_{DHC}}$), otherwise $K_{I_{DHC}}$ is reached and a quasi-brittle failure followed by an unstable ductile crack growth is observed (Figure 1b).

c. Stress intensity factor calculation: a FEM model is developed with an elastic mechanical behavior. The G-θ method is used to calculate the Rice's J-integral. A function $K_I = F(P, a/W)$ is determined (where P is the load, a the crack length and W the thickness of the specimen), such that the stress intensity factor is known for any load and crack length.

Thanks to this procedure the threshold stress intensity factor is calculated at 250 °C. In addition, the influence of temperature on the threshold stress intensity factor is evaluated.

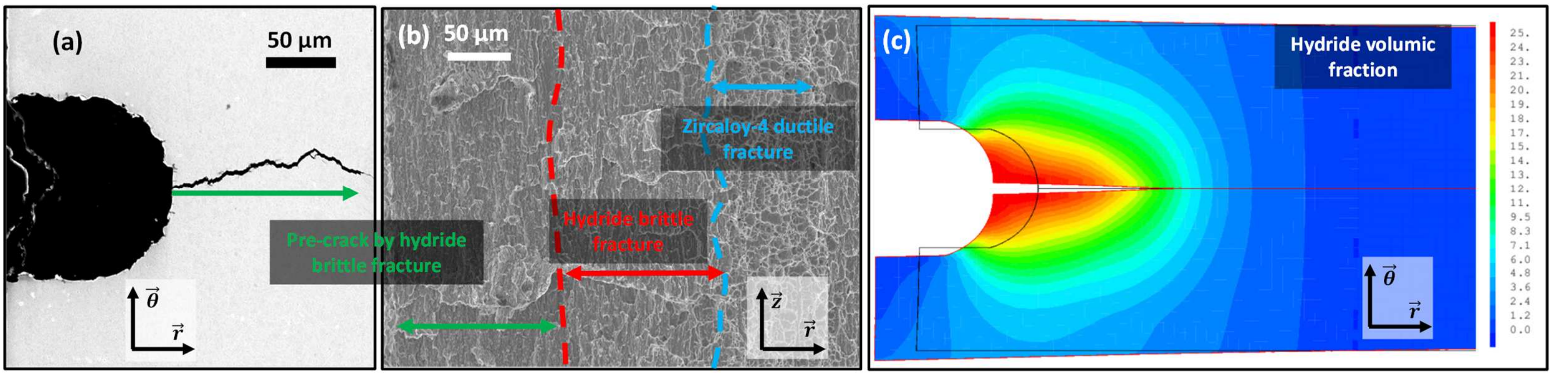

Fig.1 – (a) SEM observation of precracked notched C-ring (b) SEM fractography after DHC fracture (c) FEM model of DHC: crack propagation simulate in the same condition as tests.

3. Numerical model of DHC

A numerical model of DHC is implemented using the Cast3M software to help the understanding of the phenomenon and to predict the specimen behavior in other conditions and after irradiation. The model is multi-physical: the material's mechanical behavior is described using isotropic elasticity and anisotropic plasticity. A stress-dependant diffusion model for hydrogen in solid solution and its precipitation into hydrides is also used. Moreover, the model takes into account some chemical and mechanical phenomena affecting DHC such as:

- Hysteresis: the terminal solid solubility in dissolution (C_{TSSD}) and C_{TSSP} are different (hydrides need a higher amount of energy to nucleate than to dissolve).
- Influence of mechanical fields on the C_{TSSP}: interaction energy.
- Mechanical effects of the presence of hydrogen and/or hydrides: hydride misfit strain and lattice swelling induced by hydrogen in solid solution.

Finally, cohesive zones are used to model crack growth in both quasi-brittle and ductile regimes (Figure 1c), and the cracking criteria from a notch (hydride length and critical stress) are determined.

4. Conclusions

The experimental procedure allows for initiating and controlling delayed hydride cracking in the thickness of claddings. The fracture toughness in case of DHC is determined at several temperatures. A numerical model of DHC is developed: it reflects the experimental observations and permits the prediction of the behavior of specimens under different loading conditions and after irradiation.

Acknowledgments

The authors would like to thank the project Transport & Entreposage of the French *Institut Tripartite CEA-EDF-Framatome* for supporting this study.