

Determination of the threshold conditions for delayed hydride cracking in Zircaloy-4 CANDU fuel cladding – CNL’s contribution to IAEA Coordinated Research Programme

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ABSTRACT (Extended)

Delayed Hydride Cracking (DHC) is one of the significant cracking mechanisms for Zircaloy-4 fuel cladding and is a concern for spent fuel storage. To determine the threshold conditions of the material and develop procedures to produce a consistent set of data, the International Atomic Energy Agency (IAEA) initiated a Coordinated Research Project (CRP) in 2011. Thirteen (13) laboratories all over the world participated in this CRP, including the Canadian Nuclear Laboratories (CNL), representing Canada. The CRP was successfully completed in 2015 with a final report published in 2015 December [1].

Like other advanced structural materials, zirconium alloys are susceptible to hydrogen induced embrittlement. In zirconium fuel claddings, a flaw may extend by DHC due to hydrogen pick up by the material. The hydrogen diffuses up the tensile stress gradient at the flaw tip. If sufficient hydrogen is accumulated to form hydrides and if the stress is sufficiently large, the hydrides will fracture thereby extending the flaw. The process may continue and eventually lead to a through-wall failure. To date, fuel cladding failure due to DHC has been observed in various types of reactors, including pressurized heavy water reactors (PHWRs). This type of axial through-wall failure leads to fission product release and, potentially, loss of fuel material to the coolant. Also, the failure can lead to fuel-temperature increase, due to decrease of the fuel thermal conductivity [2], [3]. For dry storage of spent fuels, DHC can also be a potential degradation mechanism [4].

From the description of the DHC phenomenon, threshold conditions for DHC can be related to:

- Sufficient hydrogen being present to form hydrides at the flaw tip;
- The local tensile stress being large enough to extend the crack thorough the hydrides. A crack will not extend if a threshold in stress intensity factor, called K_{IH} , is not exceeded; and
- A high temperature-limit resulting in the yield stress at the flaw tip becoming too low so that propagating a crack through the hydride is no longer the preferred mechanism to accommodate the macroscopic deformation.

This presentation will report CNL's contribution to the CRP. The focus is on the determination of the K_{IH} and crack growth rate, V_{DHC} , in un-irradiated Zircaloy-4 CANDU fuel cladding in a cold-worked, stress-relieved condition. Some characteristics of the material, including the texture, the microstructure and its mechanical properties will be reported. The cladding contains approximately 130 ppm hydrogen that was charged into the material using a gaseous approach. Two methods were applied to evaluate K_{IH} of the material; both were confirmed to be successful. The methods are:

- Multiple-specimen method: testing several specimens at various values of K_I and finding the value below which no cracking is detected; and
- Up-loading method: gradual increase in the load until cracking starts.

The test specimen and fixture was the Pin Loading Tension configuration recommended by the CRP, and the test temperature ranged from 250 to 296°C. As shown in Figure 1, in the CANDU material the mean value of K_{IH} below 280°C slightly increased with temperature. The value ranged from 7 to 8 MPa√m at 250°C and was 10 MPa√m at 280°C. At higher test temperatures, K_{IH} rose sharply to 16.7 MPa√m while the crack growth rate declined towards zero. For LWR fuel cladding, similar behaviour was observed based on the data generated by the CRP [1]. This behaviour suggests that fuel cladding is immune to DHC above about 300°C. A typical scanning electron micrograph is provided in Figure 2, showing the fracture surface of a DHC specimen tested at 250°C.

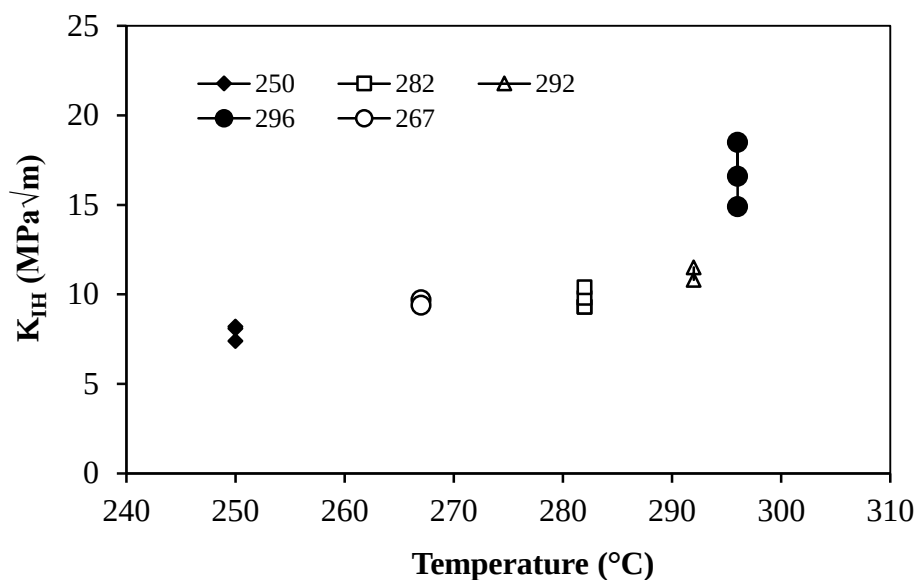


Figure 1: Temperature dependence of K_{IH} between 250°C and 296°C in Zircaloy-4 CANDU cladding with cold worked and stress relieved (CWSR) metallurgical condition.

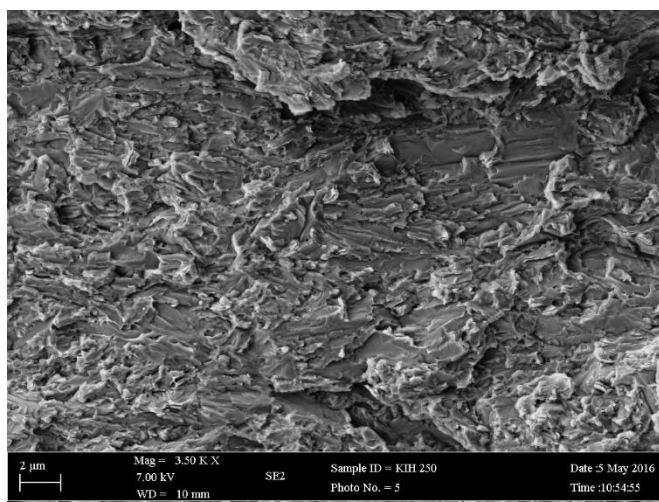


Figure 2: DHC test at 250°C. Note the area of the DHC fracture corresponds to hydride broken, characterized by cleavage fracture.

To date, CNL has established the capability to determine the K_{IH} and V_{DHC} of zirconium fuel cladding materials. This technique can further be applied to determine the K_{IH} and V_{DHC} of:

- 1) CANDU cladding with a re-crystallized or Widmannstätten microstructure, typically seen in heat affect zone of the cladding,
- 2) Advanced fuel cladding materials, e.g., Zr-1%Nb cladding, and
- 3) Irradiated fuel cladding.

References

- [1] International Atomic Energy Agency, “Evaluation of Conditions for Hydrogen Induced Degradation of Zirconium Alloys during Fuel Operation and Storage, IAEA-TECDOC-1781”, IAEA, Vienna, (2015).
- [2] R.A. Verrall, Z. He and J.F. Mouris, “Characterization of Fuel Oxidation in Rod with Clad-holes”, Journal of Nuclear Materials, Vol. 344, 240-245, (2005).
- [3] J.T. White and A.T. Nelson, “Thermal Conductivity of UO_{2+x} and U_4O_9-y ”, *ibid*, Vol. 443, 342-350, (2013).
- [4] C.E. Coleman, et al, “Is Spent Nuclear Fuel Immune from Delayed Hydride Cracking during Dry Storage? – An IAEA Coordinated Research Project”, Zirconium in the Nuclear Industry: 2016 International Symposium, ASTM.

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