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Neutron radiography of hydrogen redistribution in Zircaloy

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During the operation in a nuclear reactor, zirconium cladding tubes take up a part of the hydrogen produced by the oxidation reaction between the reactor coolant water and the hot rod surface. This hydrogen is source of degradation mechanisms affecting cladding safe operation under normal and accidental conditions. Provided the general tendency for hydrogen to diffuse down thermal and concentration gradients, and up stress gradients, hydrogen distribution can often be non-uniform within components, which could arise the risk for the integrity during intermediate dry storage, handling and transportation. Neutron radiography provides a powerful approach to investigate this non-uniform field of hydrogen concentration, given that the absorption cross-section of hydrogen for neutron is about one order of magnitude higher than that of zirconium.
A most typical case of hydrogen redistribution is delayed hydride cracking (DHC) where hydrogen diffuses along the elevated stress to the crack tip area followed by hydrides formation and cracking. In order to look into this hydrogen diffusion process driven by stress, a meso-scale 3-point bending equipment that has been constructed in our group especially for in-situ neutron imaging, was tested in an ex-situ way at the BOA beamline (SINQ spallation neutron source, PSI) using PSI 'Neutron Microscope'instrument [1]. The cold energy spectrum at BOA allows for high hydrogen sensitivity. A notched Zircaloy-4 sheet was pre-charged with ~ 600 wppm of hydrogen followed by annealing for homogeneity. Then the sample underwent a thermo-mechanical cycle of DHC test. Before and after the test, neutron imaging was consecutively performed to determine the variation of the H concentration field due to the stress. The results revealed an area of hydrogen concentration at the notch, with a concentration gradient detectable over 700 μm. A maximum elevation of 160 wppm hydrogen was found at ~200 μm off the notch, where the largest tensile stress is located according to the FEM analysis. The measurement of neutron radiography is in very good agreement with our modeling result yielding an elevation value of 250 wppm of hydrogen concentration.
Another interesting case concerns liner claddings which have been widely used in Swiss LWRs, either an inner liner to protect from PCI in BWRs or an outer liner (DX-D4) to improve corrosion resistance in PWRs. In post-irradiation examination, one always observes high hydrogen contents in liner material, visible as high concentration of hydrides, and a depleted hydrides density in the nearby cladding area. In the previous study, we have assessed qualitatively the effect of different liners on the distribution of hydrides in cladding cooled down at two different cooling rates. However, the quantification of hydrogen concentration in the liner still remains in mystery as well as the gradient at the liner/matrix interface, because the conventional measurement of hot gas extraction provides no spatial information. With this context, the determination of hydrogen concentration across various liner claddings is to be performed in our SINQ beam campaign scheduled in July 6th-8th. We would like to present these fresh results at the IAEA training workshop.
Keywords: hydrogen, diffusion, stress, neutron radiography.

[1] P. Trtik, E. H. Lehmann. Progress in High-resolution Neutron Imaging at the Paul Scherrer Institut - The Neutron Microscope Project. Journal of Physics: Conference Series 2016;746:012004.

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