In-Situ Neutron Diffraction Measurement of Terminal Solid Solubility of Hydrogen in Equilibrium with Gamma and Delta Zr-Hydrides

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In CANDU nuclear power reactors, cold worked Zr-2.5 Nb pressure tubes are used in the reactor core to contain the fuel and heavy water (D_2O) heat transport fluid. The pressure tubes operate at temperatures ranging from ~250-300°C. Over time, the pressure tubes absorb deuterium. When the hydrogen (H and D) concentration exceeds the terminal solid solubility (TSS), the tubes can become susceptible to a crack propagation process called delayed hydride cracking (DHC). The current DHC theory has recently been extended to include the effects of the gamma-hydride to delta-hydride transformation. It suggests that DHC occurs below the gamma/delta transformation temperature because the solubility of hydrogen in the Zirconium matrix in equilibrium with gamma-hydrides is higher than that in equilibrium with delta-hydrides. Since only delta hydrides form at the crack tip (as a result of higher stress levels there) in comparison to the bulk in which both hydrides form, a gradient in hydrogen concentration develops between the bulk and the crack tip. The theory concludes that hydrogen diffuses to the crack tip due to this gradient, causing further precipitation of hydrides and, thus, DHC.

The objective of this work is to measure hydrogen solubility limits in Zr-2.5Nb CANDU pressure tube material in presence of either gamma or delta hydrides. Neutron diffraction provides an opportunity to carry out such measurements, because hydrides generate distinctive diffraction peaks whose integrated intensity is directly proportional to the volume fraction of hydride precipitates in the specimen. Thus, the change in hydride peak intensity as the temperature varies could provide an accurate way of measuring the amount of hydrogen in solution at a given temperature.

Two samples were prepared under special conditions to precipitate 100% gamma hydrides. They were examined at room temperature using neutron diffraction (ND) and they showed only gamma hydride diffraction peaks, proving that the samples were good for further experiments at higher temperatures. One set of ND data were obtained at temperatures ranging from room temperature to 400°C in March 2012. The data was obtained using the E3 spectrometer at NRU reactor and a specialized furnace. The diffraction patterns were analyzed. They showed only the gamma hydride diffraction peak up to about 260°C, transforming gradually to delta hydride above 260°C and only delta hydride peak on cooling the sample. However, the background was too high and the diffraction peaks were not strong enough for accurate determination of the solubility lines. The samples were annealed and quenched again to precipitate 100% gamma hydrides. A second set of diffraction measurements were carried out in September under much improved conditions using the L3 spectrometer at NRU. However, after some measurements, due to power failure and reactor shut down, the measurements had to be postponed to a later date. Since the samples had been heated to above the gamma to delta hydride transition temperature again, they had to be annealed and quenched once more to re generate 100% gamma hydrides. Finally a complete set of measurements, using the L3 spectrometer, were carried out in November 2012. The diffraction patterns are currently being analyzed.