In-Situ Measurement of Hydrogen Sorption Properties of Zircalloy

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In-situ neutron diffraction was used to study the hydrogen sorption in zirconium based alloy zircacloy-4 (~1 wt.% Sn, ~ 0.2 wt.% Fe, ~ 0.1 wt.% Ce, balance Zr) subjected to cold rolling. After cold rolling the sample is textured and we found that the hydrogen absorption and desorption kinetics were enhanced compared to as-cast Zircaloy-4. In particular, desorption temperature was greatly reduced. As the desorption temperature (1023 K) is too high for the apparatus presently installed on C2 only absorption was investigated. Neutron investigation could give some understanding of the mechanism responsible for this temperature reduction. In-situ absorption was performed at 450°C under 15 bar of deuterium using a stainless-steel sample holder. Deuterium was used instead of hydrogen to reduce the incoherent scattering background. Patterns were taken continuously, each scan taking 2 hours to complete (1 hour per detector bank).

In-situ experiments

The as-received sample was first measured. Diffraction patterns were taken from 5 to 117 degrees (2Θ) but for easiness of comparison we presents only the range from 20 to 60 degrees. Figure 1 shows the neutron diffraction patterns for as-cast zircaloy-4. The first pattern was taken under vacuum. The most important peaks are from the SS can. Pure Zr is easily recognizable but there is an important unknown phase present. This unknown phase was almost invisible in X-ray diffraction patterns while it is quite strong in the neutron pattern. Therefore, we could suspect that the elements of this phase are relatively strong neutron scatterers. The investigation on this unknown phase is still going on. The sequence of patterns from 2 hours to 8 hours of exposure time to hydrogen clearly shows the increasing amount of ZrH2 phase and diminution of Zr phase.

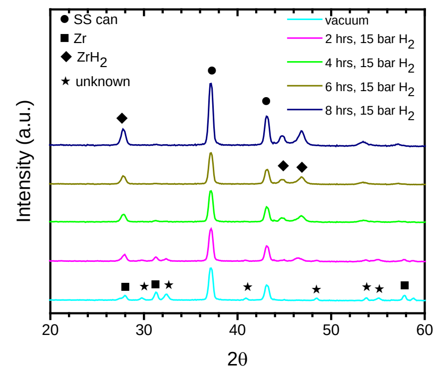

Figure 1 – Neutron diffraction of the ZK60 alloy for different SPD processes and Deuterium contents.

The crystallite size of the hydride phase was determined from Rietveld refinement. Despite the fact that the phase abundance increases with time, the crystallite size is constant (9 ± 1 nm). This mean that the hydride phase abundance increases by addition of new domains instead of growing of the existing domains.

Further analysis on the nature of the unknown phase and on the evolution of the hydride phase during hydrogenation is needed.