ESRF	**Experiment title: The development of stress anisotropy in Zr hydrides during precipitation**	**Experiment number**: MA-2676
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Shifts: 12	**Local contact(s)**: Andrea Bernasconi	*Received at ESRF:*

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Report:

Zr alloys used as fuel cladding material are subject to corrosion in pressurized water reactor. Resulting zirconium hydride precipitation is of particular technological concern during transport and storage of waste assemblies. The goal of this experiment was to investigate the anisotropic strain state in Zr hydrides during temperature cycling by monitoring azimuthal integrated Debye diffraction pattern.

In order to probe all the representative directions of the textured material, three type of sample were machined out of a block of recrystallized Zircaloy-4 with appropriate dimensions for the use of the Electro-Thermo-Mechanical Tester (20*5*.6 mm^3). The plane perpendicular to the X-ray beam was defined as the Rolling Direction & Normal Direction: RDND, Transverse Direction & Normal Direction: TDND and RDTD according to the texture of the pristine cold-rolled material. Four hydrogen contents have been considered corresponding to 0, 24, 48 and 72 hours of electro-chemical charging. A total of 12 type of sample consisting of 4 H contents * 3 directions was prepared for the experiment.

We used the ETMT mounted on the heavy-duty diffractometer in experimental hutch 1 to ensure both heating and loading. The Frelon camera was used as far field detector, with a detector to sample distance of 60cm in order to maximize the resolution of the first Debye rings. Capillary fill of CeO2 with a diameter close to our sample was used to calibrate the fine positioning of the detector. In this configuration, 20 images with acquisition time of 10s each were averaged to obtain a relatively good signal to noise ratio for the most intense hydride diffracted peak. We notice here that with a slit aperture of 50*50μm^2 the obtained resolution was below state of the art literature and could be increase with a higher working distance which imply a bigger surface of detection.

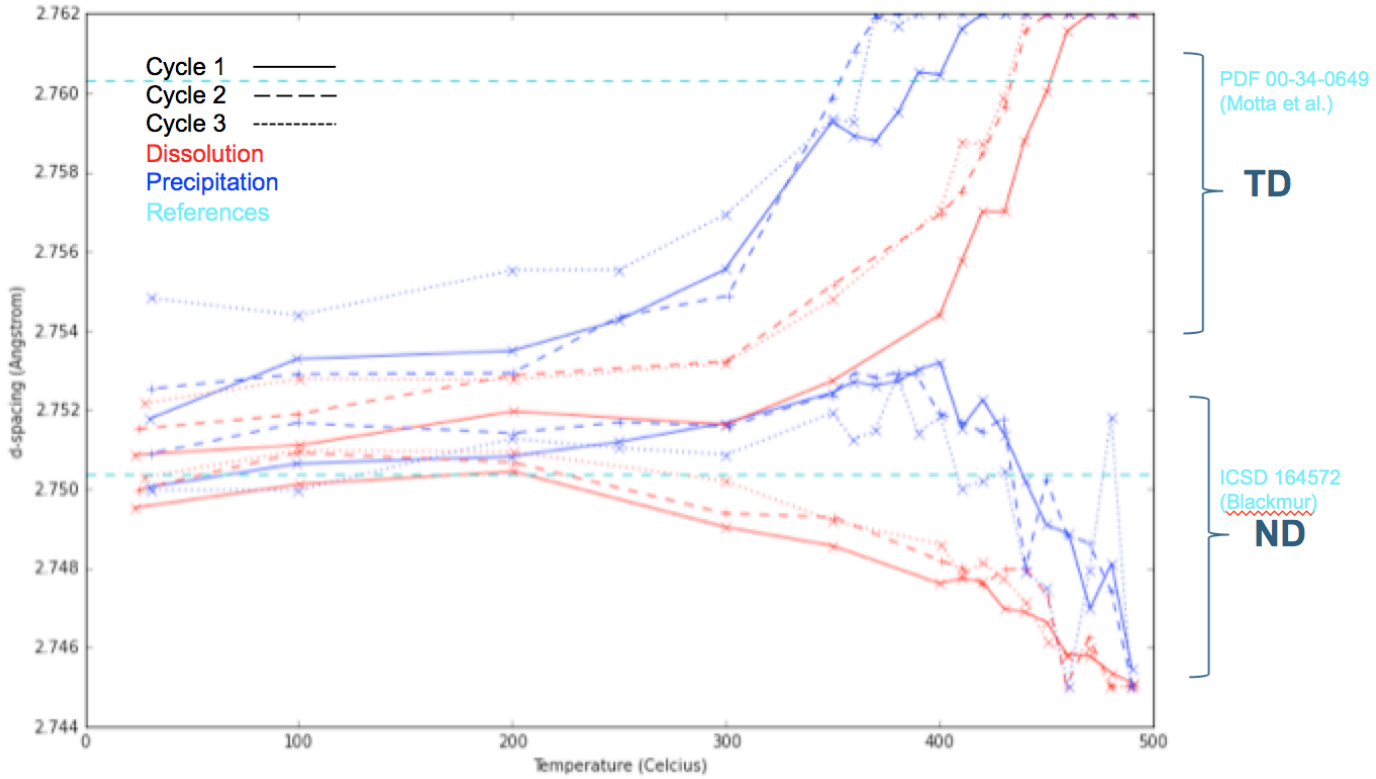

Figure 1: Evolution of the d-spacing for the 111 reflection of γ-ZrH for 3 temperature cycles with dissolution and precipitation plotted in red and blue respectively.

In 90% case, we succeeded in measuring the diffraction signature of Zirconium hydrides during at least 3 cycles of temperature up to 450/580°C depending on the supposed hydrogen content. In 10% of the case, failure of the experiment came from loosening of the spot-welded Pt/PtRd thermocouple followed by melting the sample due to a fast increase of the current. Precipitation under loading was also investigated below the reorientation threshold of the hydrides for the two directions TDND and RDND for a unique H content. However, we were not able to perform a fine monitoring of the dissolution and precipitation for sample with higher H content (electrochemical treatment of 48h and 72h). We observed that the dissolution / precipitation occurs 100°C below the temperature target estimated from the extrapolation of lower hydrogen charging. This drawback might come from saturation during the hydrogen charging and our process has to be qualified for higher H content.

Data treatment was performed with the pyFAI library and we notice here the availability of the author for supporting users. For each acquisition, azimuthal integration was extracted in orthogonal horizontal and vertical directions making possible the study of the anisotropy. The d-spacing evolution of the 111 γ-hydrides peak show a clear different behaviour depending of the probed direction with respect to the texture. As shown in Figure 1 for a H content corresponding to 24h charging, the in-plane (TD) evolution of the 111 d-spacing of γ-ZrH increase with the temperature upon dissolution and conversely upon precipitation with an hysteresis of around 100°C. More surprising, the d-spacing of the normal direction (ND) behave differently: the d-spacing decrease with the temperature when the dissolution occurs. The precipitation shows two steps: first a fast increase of the d-spacing followed by a slow decrease. Further analysis and experiment have to be done in order to disentangled Poison effect of elastic mechanical behaviour from stoichiometry effect.

Justification and comments about the use of beam time:

The small amount of hydrides in the zirconium matrix impose to count several seconds for each 20 necessary images to have a good signal to noise ratio. Each temperature cycle took 1h30 thus 4 to 6 hours were necessary for each sample to perform at least 3 cycles. In conclusion our allocated beamtime of 12 shifts was distributed in 1 shift for solving ETMT/spec communication issues, 1 shift for optimizing the setup and 10 shift for data acquisition which was quite efficient.