doi:10.1017/S1431927622006535
Microsc. Microanal. 28 (Suppl 1), 2022

Cryogenic Focused Ion Beam Sample Preparation for the Analysis of Hydrogen in Zr Alloy APT Experiments

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Zirconium alloys are used as the fuel cladding for nuclear fission reactors. As a result of contact with the coolant water, hydrogen is picked up by the material. Understanding hydrogen pick up and embrittlement at the atomic scale is a viable route to enhancing fuel lifetimes [1]–[3]. APT has been used to image hydrogen, using deuterium tracers, at the atomic scale in Zr alloys and other materials [4], [5].

To enable the analysis of specific microstructural features, such as hydrides, a focused ion beam (FIB) can be used to ‘lift out’ site-specific regions of interest [6]. However, the use of FIB to produce samples is known to cause hydride formation in certain materials, such as Zr and Ti alloys [7]. Performing the final FIB thinning at cryogenic temperatures has been shown by Hanlon et al. to reduce hydride formation in Zr alloy TEM lamellae [8]. However, if we apply this to APT analysis, outstanding questions remain, such as whether cryogenic temperatures are required at the final needle sharpening step only, in order to supress hydrogen pick up. Furthermore, how does this sample preparation approach affect the analysis of deuterium/ deuterides that exist in the material microstructure?

In this study, cryo-FIB has been used to prepare APT needles from base Zr alloys as well from regions containing deuterides. Figure 1 shows that using cryo-FIB in the final stages of tip preparation reduces the level of ^{1}H detected during APT analyses, when compared to samples produced using conventional FIB. Analysis has also been conducted on deuteride-containing samples and the results from this work will be presented (Figure 2). This research demonstrates the potential of limiting the need for cryo-FIB to the sharpening and polishing stage only, in order to reduce hydrogen pick up in Zr and similar materials. For deuterated samples, the results show that we can detect deuterium using APT even in samples exposed to a FIB beam at ambient temperatures for the majority of the standard lift-out and indicate that cryogenic temperatures might only be required in the last step of the specimen preparation process.

https://doi.org/10.1017/S1431927622006535 Published online by Cambridge University Press

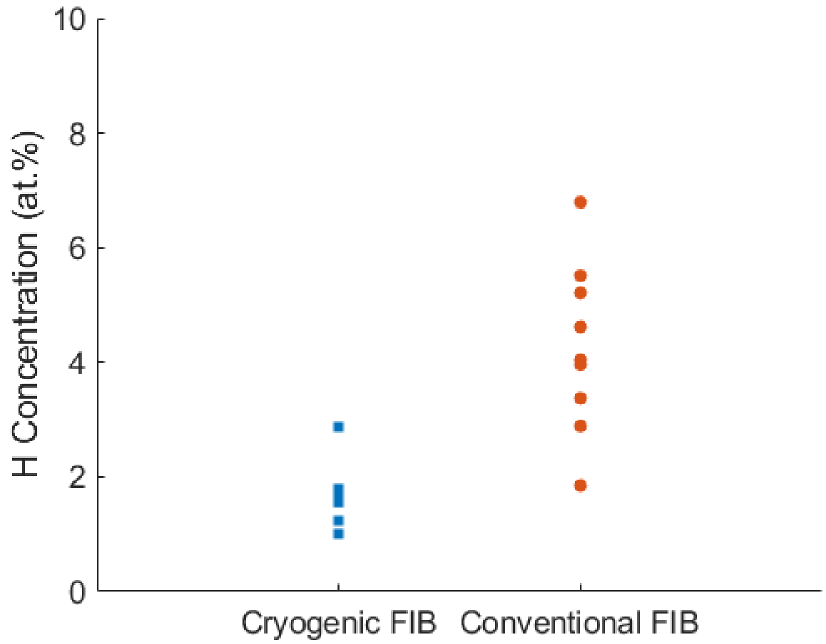

Figure 1. Difference in hydrogen concentration of Zircaloy-4 APT specimens, where the final milling has been completed at cryogenic temperatures or using conventional methods. Each data point is an individual sample.

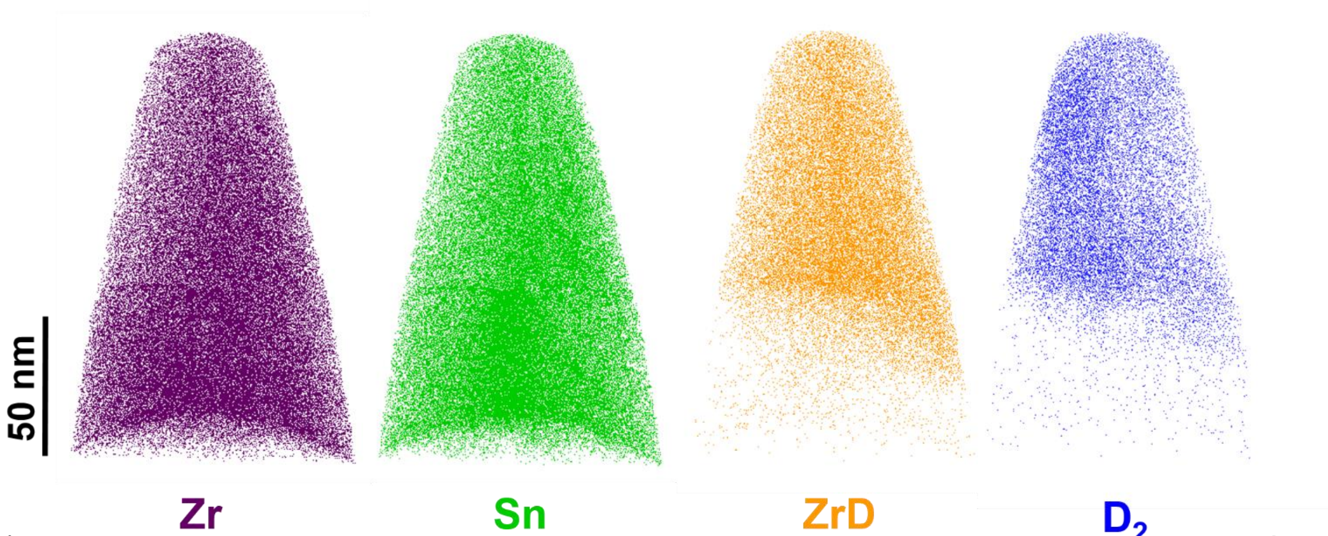

Figure 2. APT reconstruction of a deuterated Zircaloy-4 sample where the majority of the milling occurred at ambient temperatures but the final milling was done at cryogenic temperatures. The data shows a deuterium rich phase at the top of the needle.

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