Tracing hydrogen in APT : Development of new in-situ approaches

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We present new techniques for the analysis of gas species with Atom Probe Tomography (APT), focussing in particular on tracking hydrogen interaction with surfaces. APT is a 3D time-of-flight technique, that provides near atomic-scale spatial resolution, with high quality chemical sensitivity for almost all elements. One exception however, is hydrogen.

In previous works [1][2][3], we have shown that through ex-situ transfer, hydrogen can be incorporated into a target material, and then quantitatively identified in 3D through isotopic labelling with deuterium. By the use of cryogenic methods, this signal-to-noise ratio can be further improved, due to suppression of out-diffusion on experimental timescales. This was shown for both hydride forming materials (i.e. Pd), and for several types of steels using gaseous and liquid charging respectively.

In this work, we show that quantitative tracking of hydrogen can be achieved through the development of new equipment for the controlled injection of deuterium gas. In this configuration, clean metal surfaces are generated within a commercial APT system, which has been coupled via UHV transfer to a "reaction cell" system. The "cell" configuration allows for the gaseous exposure of selected gasses, such as deuterium and heavy-water vapour, at up to atmospheric pressure at controlled temperature. By exposing these UHV clean surfaces to deuterium gas, we show successful transfer and detection in APT.

We simultaneously combine this approach with thermal cycling capabilities, in order to demonstrate that the hydrogen within the material can be manipulated in a controlled manner. This allows for an unambiguous confirmation of hydrogen detection.

We show results for materials such as Pd, and for Zirconium alloys. We additionally show that the 1-hydrogen (as opposed to deuterium) signal is not a valid indicator of hydrogen content within the samples analysed, as this signal primarily arises from chamber effects.

[1] Atom probe tomography observation of hydrogen in high-Mn steel and silver charged via an electrolytic route*, Int. J. of Hydrogen Energy,* 10.1016/j.ijhydene.2014.05.169

[2] Direct observation of individual hydrogen atoms at trapping sites in a ferritic steel, *Science*,10.1126/science.aal2418

[3] Atom Probe Analysis of *Ex Situ* Gas-Charged Stable Hydrides, *Microscopy and Microanalysis*, 10.1017/S1431927616012630

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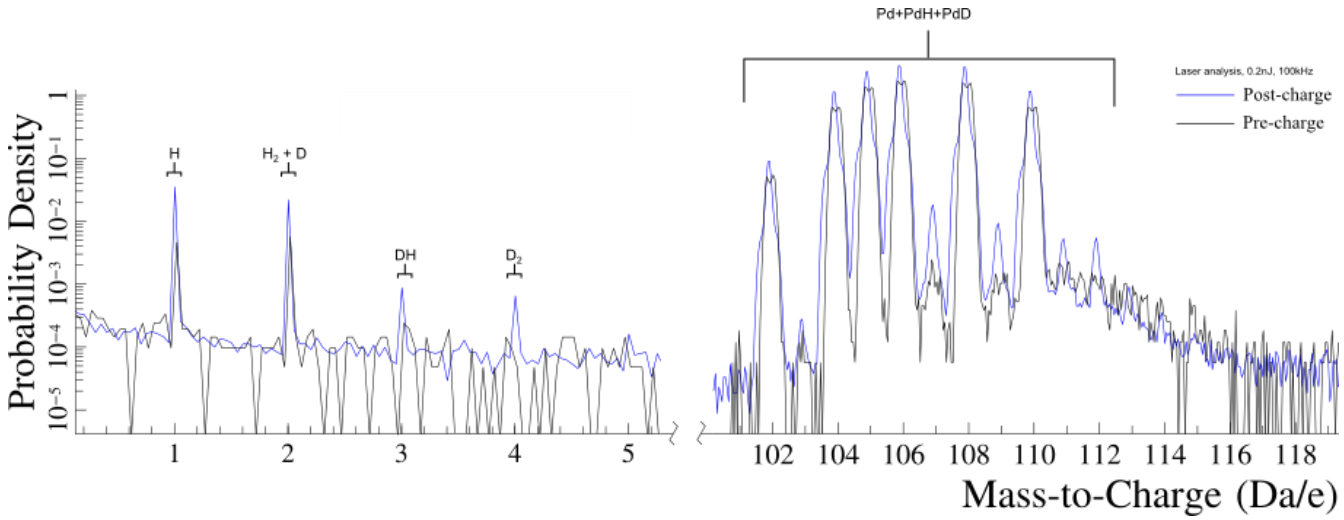

Figure 1: APT mass spectrum showing uptake of Deuterium in a Pd Alloy, with in-situ D charging (1 atm abs).