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# Imaging of hydrogen in metals using an atom probe with ultra-low hydrogen background

Peter Felfer[1], Benedict Ott[1], Martina Heller[1], Martin Weiser[1] and Mehrpad Monajem[1]

[1.] Institute for General Materials Properties, Friedrich-Alexander Universität Erlangen-Nürnberg, Bavaria, Germany
* Corresponding author: peter.felfer@fau.de

Atom probe tomography is a single atom sensitive mass spectrum method with near atomic resolution. Individual atoms, or more specifically ions, are identified by their time of flight. This means, that is principle it is equally sensitive to all elements in the periodic table. In the case of hydrogen however, the field evaporation of spurious hydrogen in from the stainless steel of the measurement chamber was dominating the signal. As a result, hydrogen could only be imaged if the hydrogen in the sample could be distinguished from the contaminant hydrogen, e.g. by using deuterium as an isotopic tracer. Using this method, several studies to locate hydrogen in the microstructure of metals have been carried out [1,2]. The limitation here is that hydrogen forms molecular ions, especially at higher measurement temperatures or when pulsed laser atom probe is used. This is especially critical as the hydrogen embrittlement, which is often the target of the analysis in combination with the high electric fields in voltage pulsed atom probe regularly leads to high rates of specimen failure.

In this talk, we show experiments carried out on a new type of atom probe, which is purpose built for the analysis of hydrogen in materials [3]. To this end, the vacuum system of the instrument (fig. 1) is largely built from titanium, which does not lead to hydrogen ingress into the chamber volume. As a result, the observed level of hydrogen in the data is currently about 2 orders of magnitude lower than in a conventional stainless steel atom probe (fig. 2). The remaining levels of hydrogen are subject to further reduction by way of swapping out the few remaining stainless-steel components, most prominently the cryostat interface. Already the current configuration leads to a reasonably large window of measurement conditions (especially specimen bias voltage), no hydrogen contamination in the atom probe data is observed. This instrument is controlled by a python powered software suite, which is presented in a separate talk. Its source code is open and publicly available through the project git repository [4].

An additional problem when hydrogen that is relevant to fracture processes is to be imaged, is the high mobility of hydrogen in the microstructure. At room temperature, the Einstein diffusion distance of hydrogen in alpha iron is about 1 cm in 1 minute. Especially for small samples, this means a complete loss of any hydrogen that is not contained in deep enough traps. At significantly lower temperature, this can be mitigated. At liquid nitrogen temperatures (77K) for example, the diffusion distance of hydrogen is only about 0.2 mm in one minute. Hydrogen in shallow traps is therefore already likely preserved. To image hydrogen at fracture relevant locations in the microstructure, keeping the samples as cold as possible is therefore desirable.

To this end, we developed a cryo- and vacuum transfer solution that allows for a full cold chain between various sample preparation stations and the titanium atom probe [5]. The most important connection hereby is the connection with the focused ion beam system. In our case, a Zeiss Crossbeam 540 FIB/SEM instrument was connected to our cryo transfer solution via an transfer adapter directly mounted onto the existing airlock of the FIB/SEM. Sample holder of various sizes can thus be moved between the FIB/SEM and the atom probe. Additional sample stations that are connected via the transfer solution include a cryo electropolishing station and a 5-axis UHV specimen coater.

With this set of equipment, it is now possible to analyse hydrogen in materials, wherever voltage pulsing can be used without the need for deuterium as an isotopic tracer. As mentioned earlier, the limitation here is that especially at fracture relevant microstructural features, the data yield is still low. The instrument shown here on the other hand creates the opportunity to also use laser assisted atom probe analysis for the imaging of hydrogen. This was thus far not generally possible due to the formation of molecular ions, which cannot usually be distinguished even from a deuterium tracer. The addition of such a laser to the system is planned for the coming year.

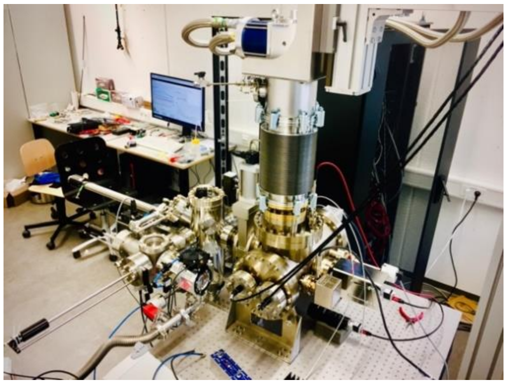

**Figure 1.** Image of the titanium atom probe system.

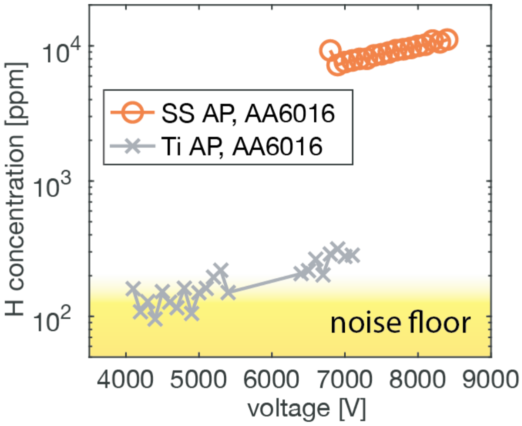

**Figure 2.** Measured hydrogen concentration in an aluminum specimen, compared between the titanium atom probe and a system with a stainless steel chamber (same specimen)

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