

Mysterious Field Evaporation Behavior of Hydrogen in Aluminium Based Material Analyzed with Atom Probe Tomography

Loïc Rousseau^{1,*}, Jean-Baptiste Maillet¹, Leigh Stephenson², Benoit Gervais³, Baptiste Gault^{2,4}, François Vurpillot¹

¹. Groupe Physique des Matériaux, Université de Rouen, Normandie, France

². Max-Planck Institut für Eisenforschung GmbH, D-40237 Düsseldorf, Germany

³. CIMAP, Université de Caen, Normandie, France

⁴. Department of Material, Royal School of Mines, Imperial College, London, England

* Corresponding author: loic.rousseau6@univ-rouen.fr

Those latter years, the ability to detect elemental particles near the atomic scale and irrespectively of their mass has engaged an important interest in the use of the APT technique to characterize Hydrogen in metallic materials. In fact, origins of the characteristic peaks H^+ or H_2^+ species (at 1 and 2 amu) are difficult to precisely define [1]. It is known that gaseous H_2 molecules are inherently present in the UHV chamber during experiments, resulting in H_x^+ detection that have nothing to do with the real distribution of the H atoms in the microstructure.

In this presentation, we get interested in the interactions between Al or H atoms during field evaporation process in Al based specimens. Hydrides peaks are generally located at 28, 29 and 30 amu corresponding to AlH^+ , AlH_2^+ and AlH_3^+ . However, when the specimen is analyzed with a straight flight path voltage pulse APT, these peaks are shifted by a few tenth of amu, resulting in a symmetric bump. The hypothesis stated is that AlH_x molecules present in the material dissociate systematically near the surface, leading to a detection of a neutral particle [2]. This proposed mechanism remained to be demonstrated.

In our experimental analysis, we find that almost all H^+ peaks (around 95%) comes from a correlated event with an Al^+ ion. We apply the Saxey method [3] to plot (Al^+, H^+) and (AlH^+, H^+) events to visualize their energy variation. Then we plot their corresponding mass peaks. As shown on Figure 1, we can easily notice a ‘coma’ waveform shown important dynamic effects [4]. This signature is a clear consequence of the voltage pulse influence on ion velocity variation during their flight, which amplifies the ion energy deficit. As schematically represented, this mechanism mostly happens before the maximum of the voltage pulse and is not related to the classical field evaporation process [5]. It explains the secondary rounded shape peaks at a same level or higher than the primary peak of H^+ or Al^+ .

One of the explanations is to consider contamination of the tip surface by H_2 molecules that are coming from the chamber after each voltage pulse. This create AlH molecules at the surface which have lower field energy barrier than Al atoms [6], so that they get field evaporated in correlated event before the maximum of the next pulse. To validate this hypothesis, we simulate the field evaporation process by considering a decreasing H reserve that is null at the maximum of the pulse. Depending of the proportion of H, the field evaporation law is strongly influenced. Then, we plot the Saxey diagram graduated as function of the probability of events. As shown in Figure 2, our result is in agreement with experimental results. Now, remain objectives are to discriminate this H^+ impurities to correct the Hydrogen concentration in the experimental analysis.

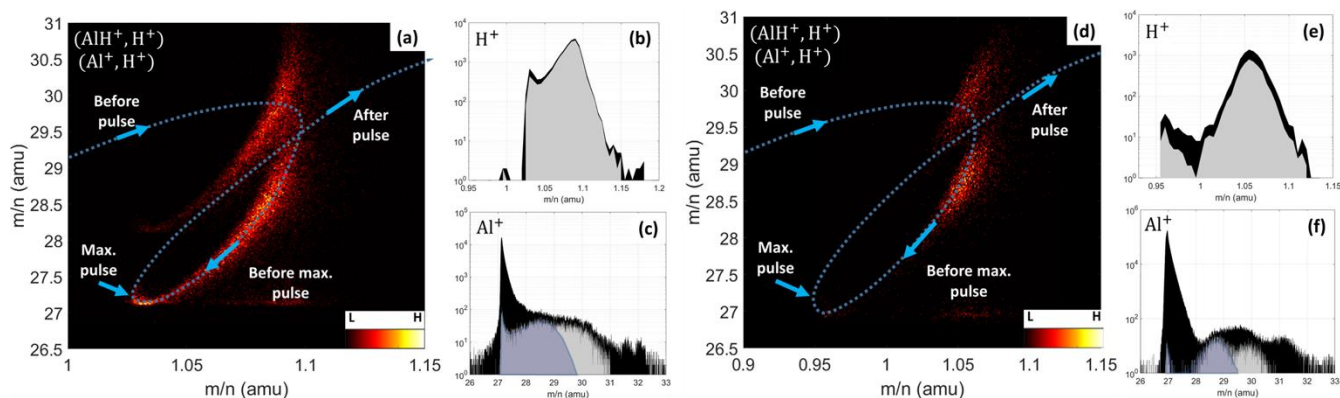


Figure 1. (a, b, c) Experimental results from an AlLi alloy. (d, e, f) Experimental results from a pure Al specimen. (a, d) Saxey diagram of (AlH^+, H^+) and (Al^+, H^+) correlated event. The color map represents the density of events. (dashed blue) A schematic extrapolation is represented. (b, c, e, f) H^+ and Al^+ peaks. (black) Total experimental peak, (grey) Mass peak related to the Saxey diagram, (light blue area) A visual guide is represented to distinguish the Al elements from AlH.

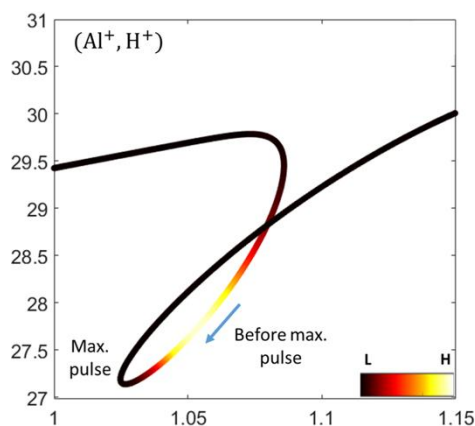


Figure 2. Simulated result of the (Al^+, H^+) correlated events considering an H contamination before the voltage pulse. The color scale represents the density of events.

References:

- [1] Yoo, S.-H. *et al.* Origins of the hydrogen signal in atom probe tomography. *ArXiv210914981 Cond-Mat* (2021).
- [2] De Geuser, F. *Interprétation et traitement des données de sonde atomique tomographique: Applications à la précipitation dans les Al-Mg-Si.* (Université de Rouen, 2005).
- [3] Saxey, D. W. Correlated ion analysis and the interpretation of atom probe mass spectra. *Ultramicroscopy* **111**, 473–479 (2011).
- [4] Rousseau, L. *et al.* Dynamic Effects in Voltage Pulsed Atom Probe. *Microsc. Microanal.* 1–14 (2020) doi:10.1017/S1431927620024587.
- [5] Muller, E. W. Field Desorption. *Phys. Rev.* **102**, 618–624 (1956).
- [6] Müller, E. W., Nakamura, S., Nishikawa, O. & McLane, S. B. Gas- Surface Interactions and Field- Ion Microscopy of Nonrefractory Metals. *J. Appl. Phys.* **36**, 2496–2503 (1965).