

Mechano-chemical coupling effects during hydriding of nanocrystalline metallic thin films

Joris Proost*, Renaud Delmelle, Marc Sinnaeve

Université catholique de Louvain, Division of Materials and Process Engineering
Place Sainte-Barbe 2, 1348 Louvain-la-Neuve, Belgium.

* joris.proost@uclouvain.be

One of the challenges for gaining a more fundamental understanding of the behaviour of hydrogen in nanocrystalline metals is the quantification in real-time of the amount and rate of hydrogen uptake and release. In this respect, we recently developed a new experimental technique for studying the hydriding behaviour of metallic thin films in-situ, based on high resolution curvature measurements [1]. In the thin film geometry, such curvature changes during hydriding arise from the constrained volume expansion upon interaction of hydrogen with the metallic thin film. The use of thin film model systems was also shown to allow more easily to separate the different rate-controlling steps involved during hydriding. In the current paper, we extend this work to address specific mechano-chemical coupling effects during hydriding, and illustrate how these may lead to significant improvements in both hydrogen storage capacity as well as uptake and release rate. In the case of room temperature hydriding of nanocrystalline thin films for instance, a first characteristic regime was observed at the beginning of hydrogen uptake, the kinetics of which are significantly enhanced by the presence of pre-existing tensile internal stresses in the film. Based on such in-situ curvature data, we have been able to quantify the effect of such pre-existing internal stresses on relevant kinetic and thermodynamic hydriding parameters for a number of tailored nanocrystalline thin film microstructures. These include the equilibrium H-concentration, the alpha to beta phase transition, and the hydrogen ad- and absorption velocities. Our findings finally resulted in the construction of a self-consistent kinetic model [2], able to describe the different rate-controllings steps encountered during the complete hydriding cycle of nanocrystalline metallic thin films, as well as their sensitivity to internal stress.

[1] R. Delmelle, G. Bamba, J. Proost, *International Journal of Hydrogen Energy* 38 (2010) 9888

[2] R. Delmelle, J. Proost, *Physical Chemistry Chemical Physics* 13 (2011) 1412