

The Hydrogenation Kinetics of a Magnesium Thin Film: An in-situ Neutron-Reflection and Optical-Transmission Study of a Two-Phase System

Lars J. Bannenberg^a, Herman Schreuders^b, Lambert van Eijck^a, Jouke R. Heringa^a, Nina-Juliane Steinke^c, Robert Dalglish^c, Bernard Dam^b, Fokko M. Mulder^b, Ad A. van Well^a

^aDepartment of Radiation Science & Technology, Delft University of Technology, Delft, Netherlands

^bDepartment of Chemical Engineering, Delft University of Technology, Delft, Netherlands

^cISIS, Rutherford Appleton Laboratory, OX11 0QX, Didcot, Oxfordshire, United Kingdom

Nanostructuring is widely applied in both battery and hydrogen materials to improve the performance of these materials as energy carriers [1]. It changes the diffusion length as well as the thermodynamics of materials. We study the impact of nanostructuring on the (de-)hydrogenation in a model system consisting of a thin film of magnesium sandwiched between two titanium layers and capped with palladium. We verified optically the coexistence of the metallic α -MgD_x and the insulating β -MgD_{2-y} phase [2], and simultaneously studied the kinetics of the (de-)hydrogenation with (off)-specular neutron reflectometry. When the β -MgD_{2-y} domains exceed the neutron's coherence length, we are able to separately determine the volumetric expansion and the deuterium content of both phases. Our results show that there are significant deviations from the thermodynamic solubility limits in bulk magnesium during the phase transformation. This suggests that the kinetics of the phase transformation in nanostructured battery and hydrogen storage systems is enhanced not only as a result of the reduced length scale but also due to the increased solubility in the parent phases [3].

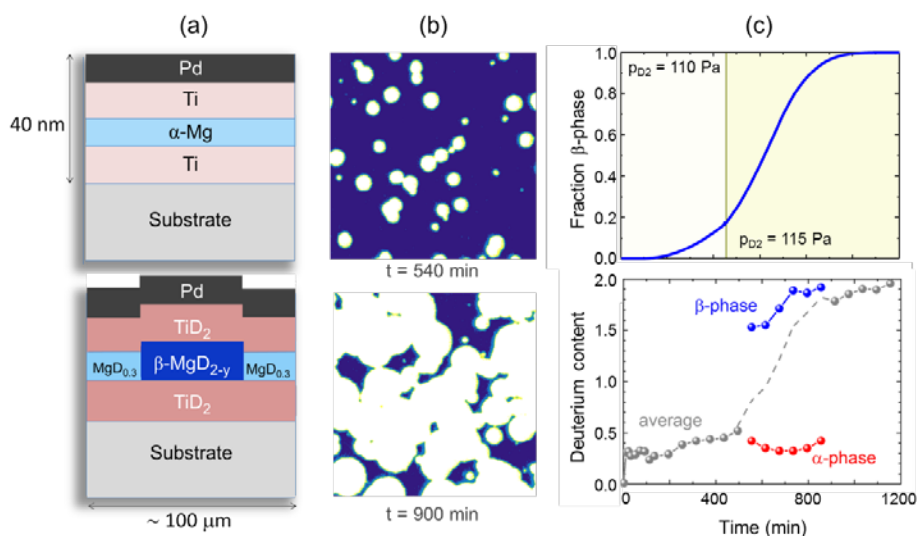


Figure 1 (a) The magnesium-based thin film system under investigation (top) in the virgin state and (bottom) during hydrogenation. (b) Top view optical transmission images at two moments in time. The β -MgD_{2-y} domains show up as bright spots that grow radially outwards. (c, top) Fraction of β -MgD_{2-y} as a function of time as determined from the optical transmission images. (c, bottom) deuterium content of the Mg layer, as determined by neutron reflection.

References

- [1] Arico, A.S., Bruce, P., Scrosati, B., Tarascone, J.M., & Van Schalkwijk, W, *Nanostructured materials for advanced energy conversion and storage devices*, Nature Materials **4** (2005) 366-377
- [2] Mooij, L. & Dam, B. *Nucleation and growth mechanisms of nano magnesium hydride from the hydrogen sorption kinetics*, PCCP **15** (2013) 11501–11510
- [3] Bannenberg, L.J., Schreuders, H., van Eijck, L., Heringa, J.R., Steinke N.J., Dalglish R., Dam, B, Mulder, F.M., van Well, A.A., *Impact of nanostructuring on the phase behavior of insertion materials: The hydrogenation kinetics of a magnesium thin film*, J. Phys. Chem. C **120** (2016) 10185-10191