Neutron Reflectometry Study of Hydrogen Desorption in Destabilized MgAl Alloy Thin Films

H. Fritzsche [1], M. Saoudi [1], J. Haagsma [2], C. Ophus [2], E. Luber [2], C. T. Harrower [2], and D. Mitlin [2]

[1] Canadian Neutron Beam Centre, National Research Council, Chalk River Laboratories, Chalk River, ON, Canada K0J 1J0

[2] Chemical and Materials Engineering, University of Alberta and National Institute for Nanotechnology, National Research Council Canada, Edmonton, AB T6G 2H7

Magnesium-based hydrogen storage is emerging as one of the most viable options for automotive applications since both elemental magnesium and magnesium hydrides are relatively inexpensive and MgH_2 holds 7.6 wt% hydrogen. The critical problem with magnesium-based storage materials is their unacceptably high charging and discharging temperature due to a relatively large negative heat of hydride formation. Mostly, approaches to modify the temperature have consisted of creating nanocrystalline hydrides by using high-energy ball milling and catalysts. Only recently, cosputtering of Mg and Al has been used to destabilize the Mg matrix and create nanocrystalline and amorphous films.

The films were cosputtered onto a Si(100) substrate with a native oxide layer of about 1 nm thickness. The MgAl films had a thickness of about 50 nm and were covered with a 10 nm thick Pd layer in order to facilitate the hydrogen uptake. The films were absorbed with hydrogen in a dedicated high-pressure furnace at 430 K for 20 h at 6.8 MPa.

The neutron reflectometry experiments were performed on the newly commissioned D3 reflectometer. Figure 1 shows the experimental data for a $Mg_{0.7}Al_{0.3}H_y$ film (a) before hydrogen absorption, (b) after hydrogen absorption (y = 1.1) and (c) after complete hydrogen desorption at 448 K (y = 0). The fits, displayed as solid lines, were calculated using the software PARRATT32. The changes in the film structure due to hydrogen uptake can be best visualized by plotting the scattering length density (SLD) profile, i.e. the SLD along the surface normal z of the film. The SLD profiles corresponding to the fits are shown in Figure 1 as insets. By comparing the SLD profiles before and after hydrogen absorption, you can easily conclude an expansion of about 20% for both, the Pd and MgAl layer. The amount of hydrogen absorbed into the MgAl layer can be determined from the decrease in the SLD of the unsorbed film due to the negative scattering length of the hydrogen [1]. From the SLD values of Figure 1b we can calculate a hydrogen content of 110 at.% or 4.1 wt%, respectively, in the $Mg_{0.7}Al_{0.3}$ film. At a temperature of 448 K (Fig. 1c) the hydrogen is completely desorbed.

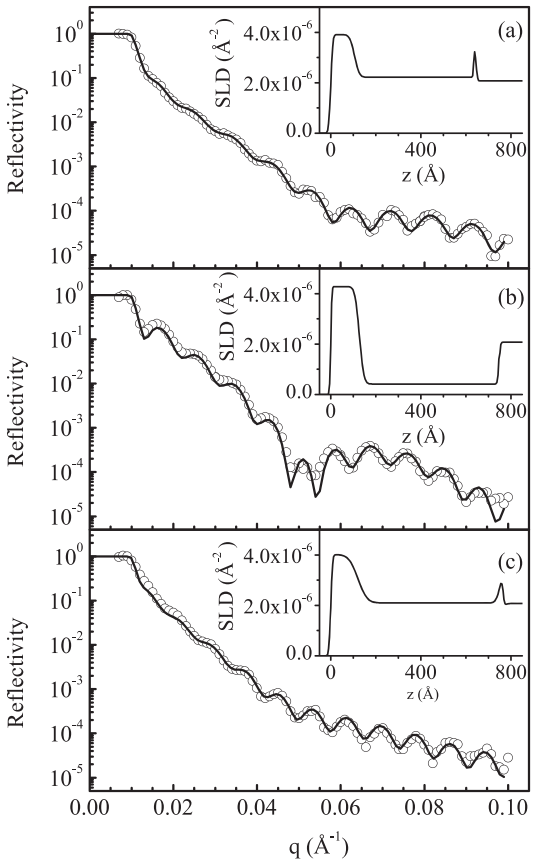

Fig. 1 Reflectivity curve of a Pd-covered $Mg_{0.7}Al_{0.3}H_y$ thin film on a Si substrate (a) before hydrogen absorption (y = 0) measured at 295 K, (b) after hydrogen absorption (y = 1.1) measured at 295 K, and (c) after hydrogen desorption measured at 448 K (y = 0). The open circles represent experimental data, the solid lines represent fits, and the insets show the corresponding SLD profile.

References

[1] H. Fritzsche, M. Saoudi, J. Haagsma, C. Ophus, E. Luber, C. T. Harrower, and D. Mitlin, Appl. Phys. Lett. 92, 121917 (2008)