Interdiffusion of Mg and Fe layers during annealing and deuterium absorption

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As the structural properties (like e.g. interdiffusion and alloy formation) can change the properties of a hydrogen storage material drastically, we applied Neutron Reflectometry (NR) in the present work to study the structural properties of a 55 nm thick Mg film sandwiched between a Ta and Fe layer and capped by a Ta/Pd bilayer during annealing and deuterium absorption, respectively.

The neutron reflectometry data of the as-prepared (black circles) and annealed (red circles) thin film structure 10 nm Ta / 55 nm Mg / 12.5 nm Fe / 5 nm Ta / 5 nm Pd is shown in Fig. 1. The fits (solid lines) are based on the SLD profiles shown in Fig. 2. The different layers of the thin film structure are clearly visible which proves the high quality of the film. The Pd and top Ta layer cannot be distinguished because the SLDs of these materials are very close: 4.0 and 3.8×10^{-6} Å^{-2}, respectively. The film structure changes after annealing for 6 hours at 250 ºC, as can be inferred immediately from the NR curve shown in Fig. 1. The SLD profile for the fit shows that the SLD for the Ta, Pd, and Fe layer changes, i.e. there is noticeable interdiffusion at 250 ºC.

When exposing the sample at RT to 1 bar of D_2 the reflectivity curve changes dramatically as can be seen in Fig. 1 (blue circles). The increase of q_c is due to the uptake of D_2 which has a large positive scattering length. From the SLD profile (blue line in Fig. 2) we can conclude that the whole Mg layer has absorbed deuterium because the SLD increased from 2.1×10^{-6} Å^{-2} to 5.7×10^{-6} Å^{-2}. The quantity of the absorbed D_2 can then be calculated from the SLD and expansion of the Mg layer. We calculate a D/M ratio of 1.9 for our Mg film, i.e. 95% of the Mg reacts to MgD_2.

After evacuating the sample space and raising the temperature to 250 ºC, q_c decreases again back to the value of the as-prepared sample (see Fig. 1 – green circles). This is a clear indication of a full desorption. However, after the desorption at 250 ºC the layer structure has changed completely with an intermixing of the Mg/Fe/Ta/Pd layers so that it was impossible to obtain a reasonable fit. As a conclusion, the desorption at 250 ºC has a more severe effect on the film structure as a simple annealing at the same temperature.

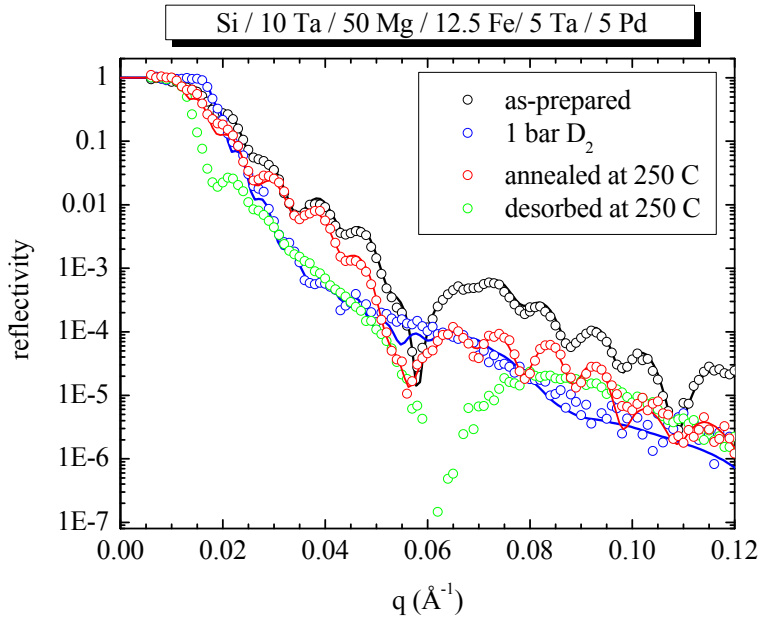

Figure 1 Neutron reflectivity curves for a 50 nm thick Mg film capped with a 12.5 nm Fe, 5 nm Ta and 5 nm Pd layer measured as-prepared (black circles), after annealing of 6 hours at 250º C (red circles), after absorption at 1 bar D_2 (blue circles), and after desorption at 250º C. The solid lines are fits based on the SDL profiles shown in Fig. 2.

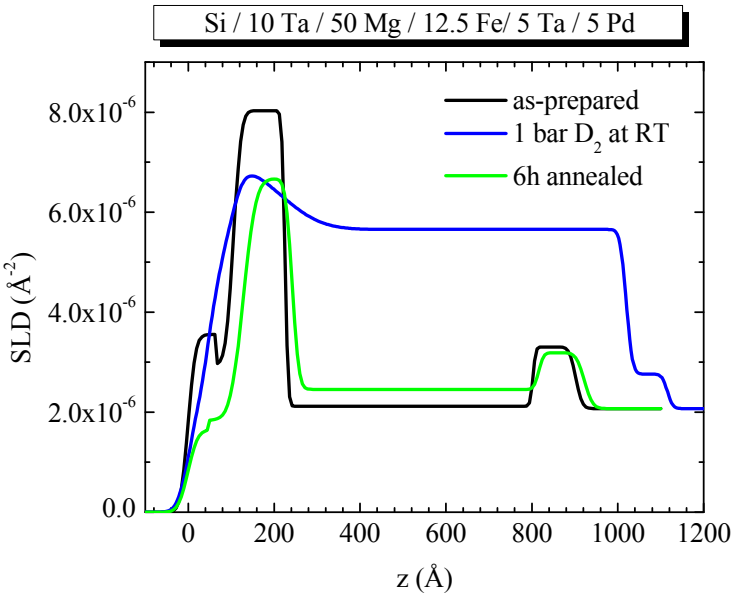

Figure 2 Corresponding SLD profiles to the fits displayed in Fig. 1.