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The role of surface oxides on hydrogen sorption kinetics in titanium thin films

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Abstract:

Titanium is presently discussed as a catalyst to accelerate the hydrogenation kinetics of hydrogen storage materials. It is however known that H absorption in Ti decisively depends on the surface conditions (presence or absence of the natural surface oxide). In this work, we use Ti thin films of controlled thickness (50 to 800 nm) as a convenient tool for quantifying the atomic transport. XRD and TEM investigations allow us to follow the hydrogenation progress inside the film. Hydrogenation of TiO₂/Ti bi-layers is studied at 300 °C, for different durations (10 s to 600 min) and at varying pressures of pure H₂ atmosphere. Under these conditions, the hydrogenation is found to be linear in time. By comparing films with and without TiO₂, as well as by studying the pressure dependence of hydrogenation, it is demonstrated that hydrogen transport across the oxide represents the decisive kinetic barrier rather than the splitting of H₂ molecules at the surface. Hydrogenation appears by a layer-like reaction initiated by heterogeneous nucleation at the backside interface to the substrate. The linear growth constant and the H diffusion coefficient inside the oxide are quantified, as well as a reliable lower bound to the hydrogen diffusion coefficient in Ti is derived. The pressure dependence of hydrogen absorption is quantitatively modelled.

Keywords:

metal hydrides; hydrogenation kinetics; surface oxides; pressure dependency; electron microscopy

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