Excellent hydrogen sorption kinetics of thick Mg-Pd films under mild conditions by tailoring structures

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Detailed experimental procedures, schematic illustration of Mg-Pd films with different structures, relative resistance changes of Mg200-Ti1 sample, XRD and SEM images, relative transmittance changes of Mg500-Pd sample, and activation energies of different samples.

Experimental procedures

Mg-Pd films with different structures were prepared by a custom designed direct current (DC) magnetron sputtering system with a background pressure of around 2×10^{-4} Pa. The glass substrates and Si (110) wafers were ultrasonic cleaned in deionized water, acetone and absolute alcohol step by step, and dried in air before use. Mg layers and Ti layers were deposited onto the substrates using a Mg (99.99%) and a Ti

(99.99%) target respectively. Finally, a 10 nm Pd cap layer was coated on top of the film to protect Mg against oxidation and to promote hydrogen dissociation and recombination. The Argon with purity of 99.99% was maintained at 0.6 Pa with flow rate of 76 sccm. The sputtering power of Mg, Ti and Pd was 50W, 60W and 45W, respectively. Mg-Pd films with different structures were fabricated by dividing the 500nm Mg layer into several parts with 1nm Ti interlayer. Corresponding to different structures, Mg-Pd samples are designated as Mg500-Pd (no Ti interlayer), Mg500-Ti1 (1nm Ti interlayer between 500nm Mg and 10nm Pd), Mg200-Ti1 (500nm Mg is divided as 200nm, 200nm and 100nm by Ti interlayer) and Mg100-Ti1 (five repetitions of a bilayer of 100nm Mg and 1nm Ti) separately for simplicity.

The hydrogenation experiments were conducted at 298K and 353K with a hydrogen pressure of 0.1 MPa. The hydrogen desorption process was carried out in dry air at 353K. The optical transmittance changes of different samples during dehydrogenation were performed over the temperature range 298K-343K for tracing the desorption mechanism. The microstructures of the samples were characterized by power X-ray diffraction (XRD) (Rigaku D/max-200) using monochromated Cu Kα radiation and scanning electron microscopy (SEM) measurements (Hitachi S4800). The resistance changes during hydrogen absorption and desorption were recorded in a gas loading cell equipped with a four-probe resistance measurement, monitored by an Agilent 34401A digital multimeter. The UV-Vis transmission spectra at different temperatures were measured with a Shimadzu UV-3100 spectrometer.

Figures

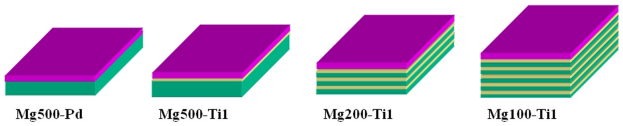

Fig. S1. The schematic illustration of 500nm Mg-Pd films with different structures.

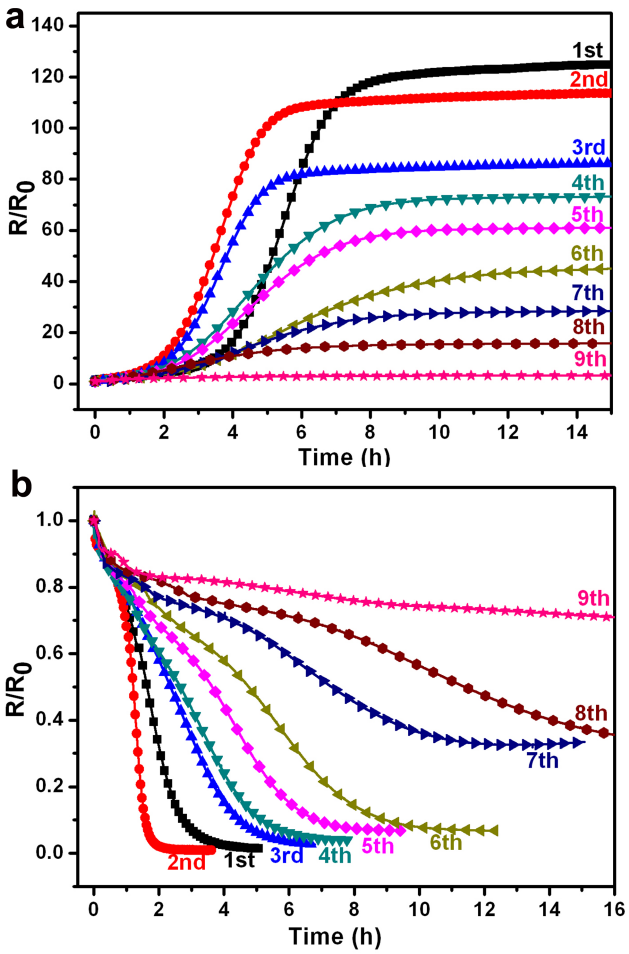

Fig. S2. The relative resistance changes (R/R_0) of Mg200-Ti1 sample during hydrogen sorption cycles. a: hydrogenation cycles in 0.1 MPa H_2 at 353K; b: dehydrogenation cycles in air at 353K.

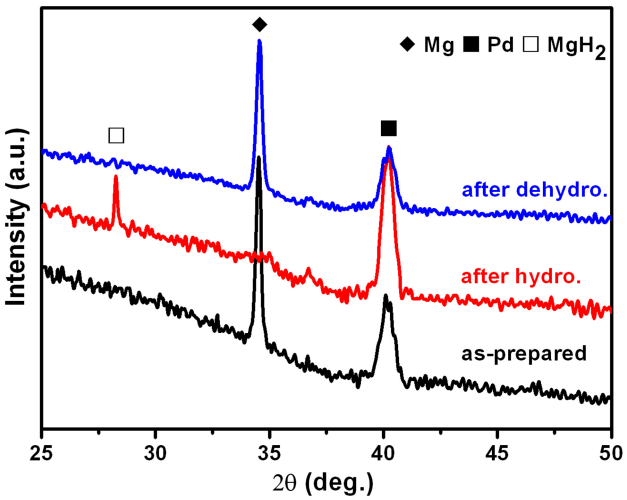

Fig. S3. The XRD patterns of Mg100-Ti1 sample during hydrogen absorption and desorption process.

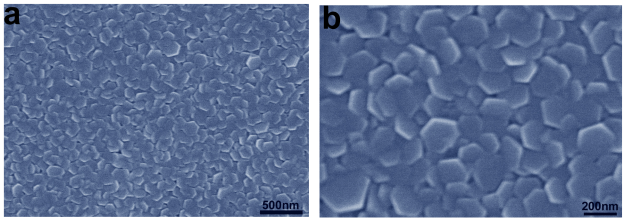

Fig. S4. SEM images of as-prepared Mg100-Ti1 sample.

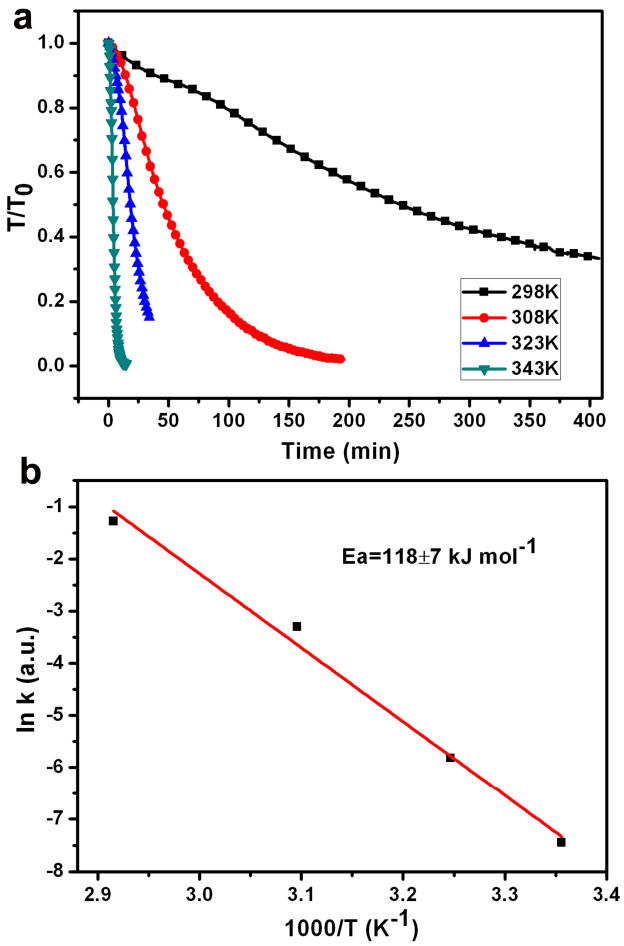

Fig. S5. a: The time dependent relative transmittance changes of Mg500-Pd sample at 500 nm, lnT/T_0, during dehydrogenation in air at different temperatures. b: Arrhenius plot of Mg500-Pd sample for hydrogen desorption in air. The straight line is the linear fit according to data.

Table

Table S1. Activation energies of different Mg-Pd samples

Samples	Activation energy (kJ mol^{-1})
Mg500-Pd	118±7
Mg500-Ti1	107±5
Mg200-Ti1	94±12
Mg100-Ti1	73±3