

Crystal Structure Determination of Ball-milled Mg-Pd-Fe Alloys and Deuterides by Neutron Diffraction

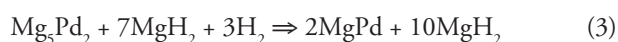
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Metal hydrides are considered valuable candidates as hydrogen storage material for future clean energy systems. However, fundamental knowledge on the hydrogenation mechanism is still needed in order to be able to design new metal hydrides that will meet the industry requirements. Consequently, study of Mg-Pd system could give valuable insight for the synthesis of other more practical systems.

In a previous work we have shown that the hydrogenation of Mg₆Pd is a three step disproportionation reversible reaction [1-4].



Because of the high cost of palladium, this alloy could not be considered as a candidate for practical applications. However, it could serve as a model in the search of new compounds. Recently, we found that by ball milling 0.5 at.% of iron with Mg₆Pd the first hydrogenation is much faster compared to compounds without iron. Neutron powder diffraction was performed to get the crystallographic characteristics of this system and to understand the activation mechanism.

The samples were prepared by ball milling 3 grams of the elemental powders with stoichiometry 6Mg + Pd + 5 at.% Fe in a steel crucible of 55cm³ internal volume with three stainless steel balls for a powder to balls weight ratio of 30. A high energy shaker mill (Spex 8000) was used. After 10 hours of milling in argon atmosphere a fine powder was collected and stored under argon. Before activation, the sample was heated to a temperature of 623 K while being kept under a 0.01 Mpa vacuum. To activate the samples, they were exposed to a hydrogen pressure of 1.33 MPa under a constant temperature of 623 K. After dehydrogenation under 0.01 MPa vacuum at 623 K the alloy was single phase Mg₆Pd as evidence by X-ray and neutron powder diffraction. The fully hydrided sample was synthesized by exposing at 623 K the activated sample to 4 MPa of deuterium gas. Neutron diffraction of the arc melted alloy was performed on the high-resolution neutron powder diffractometer C2 DUALSPEC at Chalk River (wavelength 1.3287 Å).

Figure 1 present the neutron diffraction powder of fully deuterated Mg₆Pd + 5at.%Fe alloy compared to fully deuterated Mg₆Pd pattern reported in a previous study [3]. Both patterns are similar except for the iron Bragg reflections seen in the Mg₆Pd + 5at.%Fe pattern. However, a close inspection shows

that the peaks in the iron catalyzed samples are broader and the relative intensities are different. It may be an indication that iron keep the particle size small thus helping the interdiffusion of the elements during hydrogenation/dehydrogenation process. We are now doing a full Rietveld analysis of these patterns. Complete results will be published in a forthcoming paper.

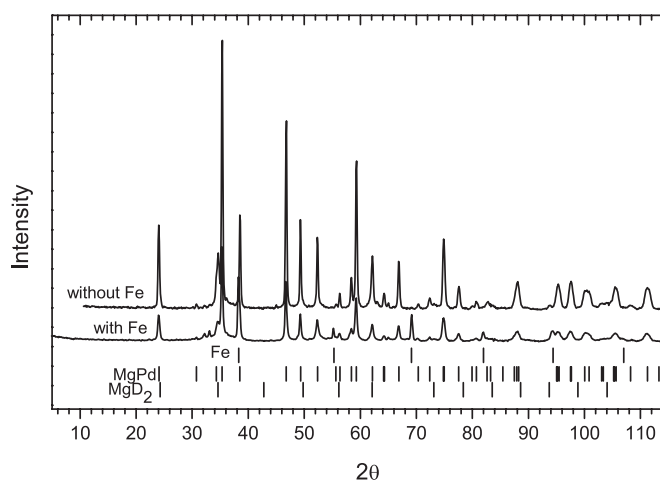


Fig 1. Neutron powder diffraction patterns of fully hydrogenated Mg₆Pd with and without iron. Bragg reflections of each phase are indicated by vertical bars.

References

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