

Composite structure and hydrogen storage properties in Mg-base alloys

M. Zhu*, H. Wang, L.Z. Ouyang, M.Q. Zeng

College of Mechanical Engineering, South China University of Technology, Guangzhou 510640, China

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Abstract

In order to improve the hydrogen absorption/desorption kinetic properties of Mg and Mg–Ni alloys, composite hydrogen storage alloys in the form of powder and film have been synthesized and investigated. For fabricating the composite powder, Mg or Mg–Ni powder was mechanically alloyed with $\text{MmNi}_{3.5}(\text{CoAlMn})_{1.5}$ alloy. For the preparation of the film with a composite structure, evaporation deposition and magnetron sputtering methods have been used to fabricate Mg–Ni film with multi-phase structure and Mg/Mm–Ni and Mg–Ni/Mm–Ni multi-layer film. By controlling the fabrication process, the microstructure feature, such as phase constituent, grain size, interlayer distances, interlayer boundary structure, of the composite can be modified. To reveal the influence of the composite structure on the hydrogen absorption/desorption kinetic properties of Mg and Mg–Ni alloys, hydrogen storage properties of the composite were measured with their microstructure features varied systematically. The present work shows that the hydrogen sorption properties of Mg and Mg–Ni-based alloys can be substantially improved by forming composites having proper microstructure features.

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1. Introduction

Hydrogen storage materials play a key role in the hydrogen economy which has emerged with the increasing demands on environment protection and energy consumption. The most important goal of hydrogen storage materials research is to develop materials with high hydrogen storage capacity, fast hydrogenation/dehydrogenation kinetics and low economical cost [1]. The hydrogen storage capacity of Mg and Mg–Ni-based alloys is rather high, 7.6% and 3.6% in mass, respectively, and they are economical in manufacturing and application. However, their hydrogen absorption and desorption temperature is too high and their hydrogenation/dehydrogenation kinetics is too poor to satisfy practical

applications. Great effort has been made to overcome the drawbacks of Mg and Mg–Ni-based alloys. The main approaches include alloying element addition [2–4], surface modification [5], forming non-equilibrium microstructure [6–9] and forming composite with some catalytic components and other hydrogen storage alloys [10–14].

Early in the 1980s, composite hydrogen storage alloys had been made in the purpose of improving the hydrogen absorption/desorption properties of Mg [15]. However, the property of this type of composite is simply a sum of that of the constituent phases in composite. Further research reveals that interaction between different constituent phases takes place in the interphase boundary region and this interaction can improve the kinetic properties of Mg-based alloys [16]. It is apparent that increasing interphase boundary density is beneficial for the property improvement. On the other hand, in accordance with the understanding of high chemical activity and fast diffusion ability of nanostructured materials,

* Corresponding author. Fax: +86 20 87114484.

E-mail address: memzhu@scut.edu.cn (M. Zhu).

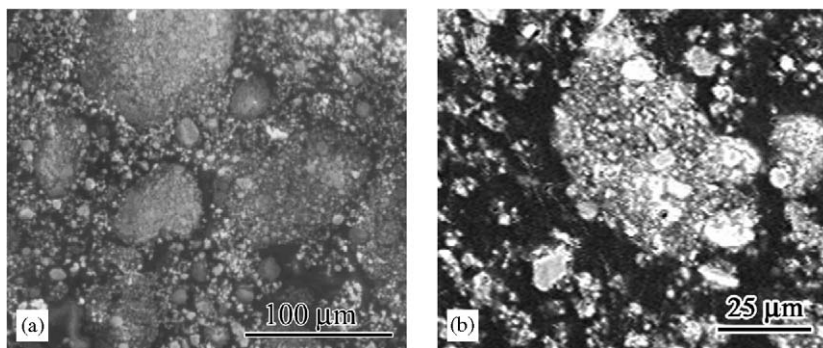


Fig. 1. SEM images showing the morphology of the MmM₅–10 wt% Mg composite milled for 20 h (a) and MmM₅–25 wt% Mg₂Ni composite milled for 10 h (b).

it was found that nanostructures are beneficial to the hydrogenation/dehydrogenation kinetics. Thus, a lot of work has been made to improve the kinetic properties of Mg-based alloys by preparing nanostructured Mg-based composite using various methods. One of the most commonly used methods is mechanical alloying. Mg and Mg–Ni alloys were mechanically alloyed with pure metal, such as Pd [17], V [18] and Ni [19], and other hydrogen storage alloys, such as FeTi [20], LaNi₅ [21] and TiV_{2.1}Ni_{0.3} [22]. Another method, which attracts a lot of attention recently, is preparing multi-layer film by physical vapor deposition methods [23,24]. For instance, by making a Pd/Mg/Pd sandwich structured film [24], the hydrogenation/dehydrogenation temperature of Mg decreased to about 373 K. It should be emphasized that different component combination and microstructure can be obtained in designed way in a multi-layer film. Therefore, it is convenient for investigating the correlation between the microstructure and the properties of composite.

In past several years, the present authors and co-workers investigated the microstructure and properties of Mg/Mm–Ni and Mg–Ni/Mm–Ni composite in order to reveal the influence of the microstructure of composites on the kinetic properties of Mg and Mg–Ni alloys. The present paper reviews our recent results.

2. Grain refining effect in composite prepared by mechanical alloying

Mg and Mg–Ni alloys were ball milled with LaNi₅-type alloy with a composition of MmNi_{3.5}(CoMnAl)_{1.5} (denoted as MmM₅ here below) to form a composite [11,21]. By adjusting the milling parameters, the microstructure of the composite can be modified. Figs. 1 (a) and (b) show the morphology of the Mg/MmM₅ and Mg₂Ni/MmM₅ composite prepared by milling. It shows that the brittle MmM₅ phase was embedded in the relative ductile Mg and Mg₂Ni phase and the two components forms a homogeneous composite after milling. The average grain size of the two con-

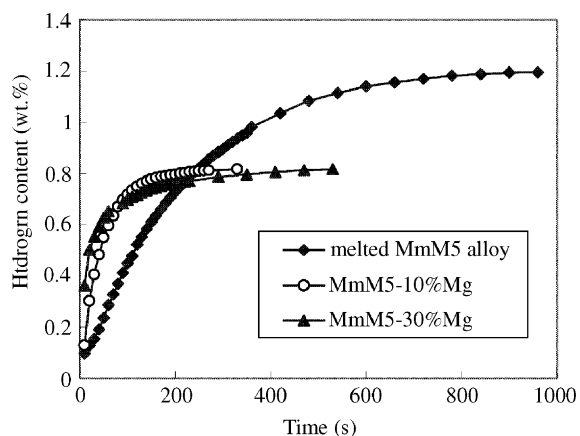


Fig. 2. Hydriding kinetic curves measured under 0.6 MPa at 353 K and their fitting curves for the melted MmM₅ alloy and the nano-phase MmM₅–30% Mg composite. Symbols stand for the experimental data and dash lines are the fitting to the data.

stituent phases reduces with the increasing of milling time. It should also be pointed out that a solid-state reaction takes place between Mg and MmM₅ in the milling process and produces La₂Mg₁₇ and Mg₂Ni phases [25]. With the decreasing of the grain size of the constituent phases, the hydrogenation kinetics of the composite was improved. It is interesting that hydrogenation took place at room temperature for the Mg/MmM₅ composite obtained by milling for 20 h or longer. This is owing to the formation of a nanostructured composite and the fact that most of the Mg reacted with MmM₅ and transformed to La₂Mg₁₇ and Mg₂Ni phases.

Fig. 2 shows the hydrogenation kinetic curves of melted MmM₅ alloy and Mg/MmM₅ composite obtained by 20 h of milling. It shows that the hydrogenation rate of the composite containing Mg is higher than the melted MmM₅ alloy. By fitting the kinetic curves with various rate equations of reactions, it has been found that the kinetic curves

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