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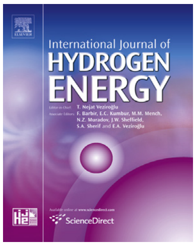

Short Communication

Study on the synthesis and hydrogen storage properties of Mg_2CuH_3

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ABSTRACT

In this study, Mg_2CuH_3 was prepared using the replacement-diffusion method, and its structure was confirmed by X-ray photoelectron spectroscopy. The hydrogen storage properties of Mg_2CuH_3, such as dehydrogenation temperature and hydrogen content, were subsequently characterized. Results indicate that Mg_2CuH_3 is a multiphase material and that the main phase is composed of $MgCu_2$ and MgH_2. The optimum reaction conditions for the synthesis of Mg_2CuH_3 are as follows: 6 MPa hydrogen, hydriding reaction temperature of 350 °C, and reaction time of 5 h. The onset dehydrogenation temperature of Mg_2CuH_3 is about 420 °C, which is lower than that of MgH_2 (440 °C). The dehydrogenation peak temperature of Mg_2CuH_3 is close to that of MgH_2. The hydrogen contents of Mg_2CuH_3 and MgH_2 as measured by simultaneous thermal analysis DSC–TG are about 2.36% and 7.4%, respectively, which are near the results measured by elementary analysis and lower than the theoretical values probably because of the existence of some impurities.

Introduction

Hydrogen is a promising non-polluting power source because of its high-energy content (124 MJ/kg), which is much higher than that of other chemical fuels, such as petroleum (44 MJ/kg) [1]. Safe and efficient hydrogen storage systems must be developed for the practical use of hydrogen energy. Currently, one of the most attractive methods of hydrogen storage is the use of metal hydrogen systems [1–4]. Given their (a) light weight, (b) high storage capacity, and (c) low cost, magnesium-based alloys are among the most attractive materials for hydrogen storage, and their hydrogen storage capacity exceeds that of all known reversible metal hydrides [5,6]. At present, numerous studies on magnesium-based alloys focus on Mg_2Ni as typified by A_2B. However, it has almost no practical application because it has poor hydriding–dehydriding properties at ambient temperature, is thermodynamically too stable, and is kinetically too slow. Many efforts have been recently made to solve these problems. Partial element replacement and the addition of other elements are very effective methods to improve the hydriding–dehydriding properties of the Mg_2Ni series. The addition of Cu catalyzes the hydrogen-absorption reaction and increases the plateau of the P–C–T curves of Mg_2Ni [7,8].

Mg_2Cu as typified by A_2B is another important magnesium-based alloy for hydrogen storage and nominally provides 2.6 wt% of hydrogen. Mg_2CuH_3 has potential application in pyrotechnics and energetic materials [9]. However, reports on the hydrogen storage properties of Mg_2Cu are still very few [10,11].

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In this study, Mg_2CuH_3 was prepared and its hydrogen storage properties, such as dehydrogenation temperature and hydrogen content, were characterized. The optimum reaction conditions for the synthesis of Mg_2CuH_3 were also studied.

Experimental section

The source materials consisting of Mg powder 99%, acetonitrile (CAN, AR grade), $CuCl_2$ (AR grade), hydrogen, nitrogen, and argon were obtained commercially. All reagents were used as received without further purification.

Preparation and structure characterization of magnesium-based hydrogen storage materials

Mg_2CuH_3 was prepared through the replacement-diffusion method (RDM). Mg (with a purity of 99.8% and average particle size of 20 μm) and anhydrous $CuCl_2$ were taken in the correct proportion and reacted in dry CAN in a flask. The flask was shaken for several minutes, and the reaction temperature was kept at 45 °C for 4 h. During this process, $CuCl_2$ was dissolved, and Cu^{2+} was replaced with Mg. The yielded precipitate was separated from the solution by filter and washed with dry acetone. The precipitate was then heated under an argon atmosphere (3 atm) in a stainless steel vessel. The vessel was heated at 550 °C and kept at this temperature for 3 h, during which the component of the precipitate interdiffused in a solid state. Finally, the vessel was cooled at room temperature, and the resultant Mg_2Cu was obtained.

The yielded Mg_2Cu was placed in a stainless steel reactor and was evacuated to about 10^{-2} torr. Then, 6 MPa of hydrogen was introduced to the reactor and heated at 350 °C for 5 h. The reactor was slowly cooled at room temperature, and Mg_2CuH_3 was finally obtained.

Structure and property characterization of magnesium-based hydrogen storage materials

The structural analysis of magnesium-based hydrogen storage materials was performed by X-ray diffraction (XRD) using a Bruker D8 Siemens diffract meter with Cu Kα radiation. The H content and onset dehydrogenation temperatures of the samples were measured with a thermal analyzer TA DSC–TG SDT Q600 apparatus. The H content in the samples was also determined by elementary analysis using Elementar Analysensysteme Vario EL III Elemental Analyzer.

Results and discussions

Structure of Mg_2Cu and Mg_2CuH_3

The X-ray diffraction pattern of Mg_2Cu is provided in Fig. 1, which shows that the XRD Bragg peaks at 20.3°, 22.9°, 37.4°, 40.3° and 46° correspond to the XRD peaks of Mg_2Cu and that Mg_2Cu has a hexagonal crystal structure. No obvious oxide phase and Mg phase trace are observable in the XRD pattern of Mg_2Cu.

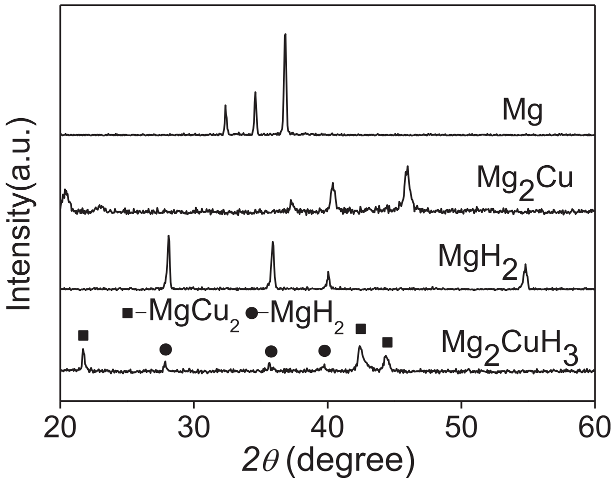

Fig. 1 – XRD patterns of Mg_2Cu and Mg_2CuH_3.

Fig. 1 shows the XRD spectrum of Mg_2CuH_3. In the figure, the XRD Bragg peaks at 21.7°, 42.4°, and 44.4°, which represent further developed versions of the humps correspond to the XRD peaks of $MgCu_2$. The XRD peaks at 27.9°, 35.7°, and 39.7° correspond to the XRD peaks of MgH_2. These findings show that Mg_2CuH_3 is clearly a multiphase material and that the main phases are composed of $MgCu_2$ and MgH_2. Hydrogen is stored in Mg_2CuH_3 in the form of MgH_2, and this property is the same as that of other magnesium-based hydrogen storage materials. $MgCu_2$ and MgH_2 are the products of the hydriding reaction of Mg_2Cu with H_2. The hydriding reaction of Mg_2Cu with H_2 is presented in Ref. [9]:

$$2Mg_2Cu + 3H_2 = MgCu_2 + 3MgH_2.$$

SEM images of Mg_2CuH_3

Fig. 2 shows the SEM images of Mg_2CuH_3. In the figure, the sample particles are packed loosely. The size dispersion of the sample particles is nonuniform, and the average particle size is about 10 μm. The figure also shows some cracks on the surfaces of the particles. After reaction with H_2, the lattice parameters and lattice volume of the metal increase, thereby increasing the metal volume and generating cracks on the outer and inner surfaces of the alloy.

Dehydrogenation temperature of Mg_2CuH_3

Dehydrogenation temperature is an important property of hydrogen storage materials. In this study, the dehydrogenation temperature of Mg_2CuH_3 was measured using a DSC–TG simultaneous thermal analyzer, and the curves of DSC–TG are shown in Fig. 3. The figures show that the DSC curve of Mg_2CuH_3 is similar to that of MgH_2 and that only one thermal decomposition peak that corresponds to dehydrogenation