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The role of SiC on the Desorption Temperature of Mg-based Hydrogen Storage Materials Prepared by Intensive Milling Method

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Abstract Magnesium, theoretically, have the ability to absorb hydrogen in large quantities (~ 7.6wt%). However,the kinetic reaction is very slow, therebyhindering the application of magnesium for hydrogen storage material. In this paper, we reported a series of preliminary studieson magnesiuminserting with silicon carbide (2 wt%) obtain by mechanical millingmethod. The vibratory mill type apparatus was used for 180 hours. As the results, structuralcharacterization by XRD showed that the crystallite size after milling for 180 hours decreased around tensnanometer. It was also found that the desorption temperature for the sample after 180 milling inform us that the material decomposed at 330◦C. It can concluded that Mg catalyzed with 2 wt% of silicon carbide (SiC) can be prepared by vibratory ball milling.

Keywords: MgH_2, catalyst, nanoparticle, hydrogen storage, reactive milling

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1. Introduction

Among the metal hydrides, magnesium has the theoretically highest weight capacity for hydrogen storage (7.6 wt.%), lightweight and a reasonably low cost [1]. However, high working temperature, slow reaction kinetics and difficult activation limit the practical application of Mg-based hydrides. Many efforts have been done to improve the adsorption properties and reaction kinetics such as element substitution (metal or metal oxides) as catalyst in nanometer scale and modification of ball milling technique as well [2-5]. Recently, the reactive ball milling under hydrogen atmosphere was also successfully introduced to prepare hydrogen storage materials [6,7].

In order to find a more suitable co-catalyst material to be used with magnesium, Ranjbar et al [8], introduced a new catalyst based on silicon carbide (SiC) to be inserted in magnesium as host material. It was found that by using SiC as an additive, the grain size of MgH_2 was decreased, and at the same time increased the defect concentration, which subsequently prevent the agglomeration of MgH_2 crystallitesas well. The sorption properties were improved and the kinetics was also very fast. By using this finding as a motive of our study, here we report our work on Mg catalyzed with 2wt% of SiC prepared by vibration ball milling (VBM). The aim of this study was to

synthesize nanocrystalline Mg-SiC by mechanical alloying using intensive mechanical milling method.

2. Materials and Method

In this experiments, Mg (Merck, 0.03 mm, 95%), SiC (99.9%, −400 mesh, Sigma Aldrich) powders wereused. The powders were filled into a home-made hardened steel vial and sealed together with 10 balls (5.6 mm in diameter). The powders were milled in avibratory ball mill (VBM, Kawasaki) at a rotational speed of 900 rpm (ball to powder ratio 10:1) for 180 hours. Every several hours of milling time (30, 40, 60, 80, 100, 160 and 180 hour) small amount of powder were separated into a small container for further characterization. This method was then called as Intensive Mechanical Alloying (IMA). Structural changes occurred on the samples during milling were then characterised by XRD (Philips, PW 3710, Co-Kα radiation). A high resolution scanning electron microscopy (SEM JEOL,JSM-5310LV) was used to observe the morphological changes during the milling process. The onset temperature (T_{onset}) was investigated using DTA (Shimadzu, D-50) with a flow rate of 20 ml/min and heating rate from 20-450◦C.

3. Results and Discussions

Figure 1 shows the evolution of the XRD diffraction pattern for Mg-2wt% SiC as a function of milling time and intensity. At early stage of milling, the starting mixture shows the presence of microcrystalline materials. Then, it can be seen for the next milling time that the diffraction peaks broaden but no changes in the 2θ position. The as-received sample composed mainly Mg. The Mg peaks locate at 2θ = 37,86^o; 40, 56; 43, 66; 56, 8; 68, 06; 75, 28; 82, 5; 84, 24, and 87, 14. SiC peak was not detected due to the amount of SiC was too small (2 wt %). Thus, it is difficult to detect by XRD. The same result was found after 30 h milling.

When the milling time increase up to 40 hour, the XRD patterns still not change significantly. But, at 60 hour the peak broadening was start to appear. At 80 to 100 hour, the Mg peaks were broadening significantly. At 160 hour and 180 hour the peak unity in one peak and composed one phase. It can be seen that the powders already reduced into nanocrystalline. We can conclude that the Mg particles completely decrease after milling in 180 hours. This is due to the high energy vibration ball mill. This result showed that mechanical alloying using VBM is very attractive and promising method to synthesize nanostructure materials for solid hydrogen storageconsidering the applied mechanical deformation between ball and powders. Thus, making use of higher energy during milling promotes the formation of nanostructure of magnesium.

The SEM micrographs of Fig. 2 show secondary electron image of powders intensively milled in the VBM before milling (Fig. 2a), milling 60h (Fig. 2b) and milling 180 hours (Fig. 2c). The surface of the powder for 0 h and 60 h are irregular, as a result of the fracturing during the milling process. The SEM image after 180 hours of milling

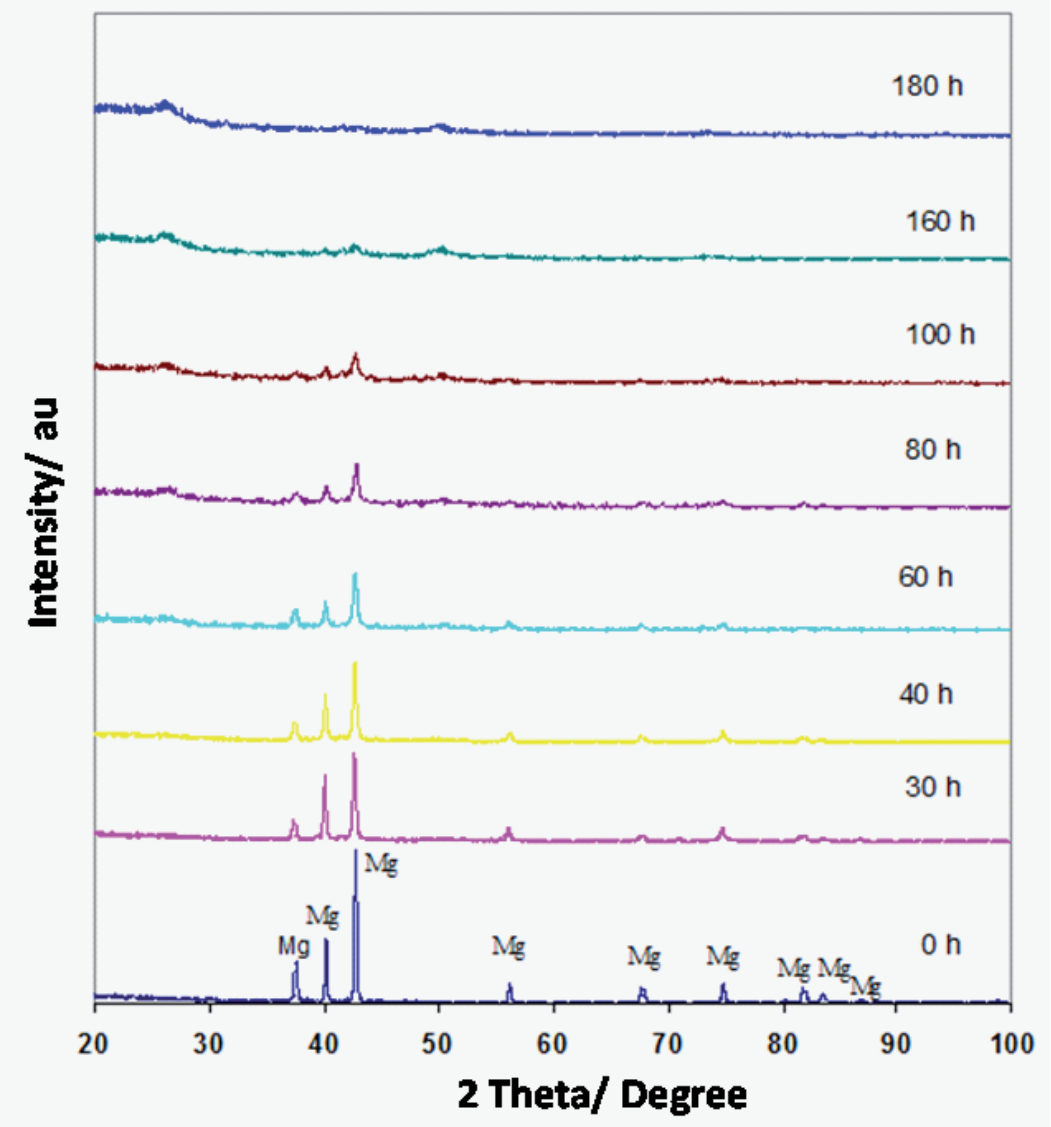

Figure 1: X-ray diffraction patterns of Mg-2wt% SiC produced via VBM.

shown the finer powder. It seem the powder was already in nanocrystal scale. If we compare with XRD, this is consistent with the structural analysis by XRD.

This indicates that the powders is un-uniformly distributed on the metal surface and the grain size, calculated by Scherrer method [9], reaches around tens nanometer after 180h of milling. However, the formation of nanocrystalline material is obtained after long milling (in 180 hours). These results suggest that deformation during milling take longer time due to the ductility of magnesium. Important to note, the silicon carbide in the composites helps to break the magnesium particles into smaller sizes due to itshardness. Ranjbar et al. [8], has clearly demonstrate that the addition of SiC can help to generate smaller MgH_2 nanocrystals and it was found that the MgH_2 peaksin the XRD pattern became very broadened. They indicated that long ballmilling was beneficial for the conversion of Mg powder to MgH_2 andalso helpful for reducing the grain sizes of powders.

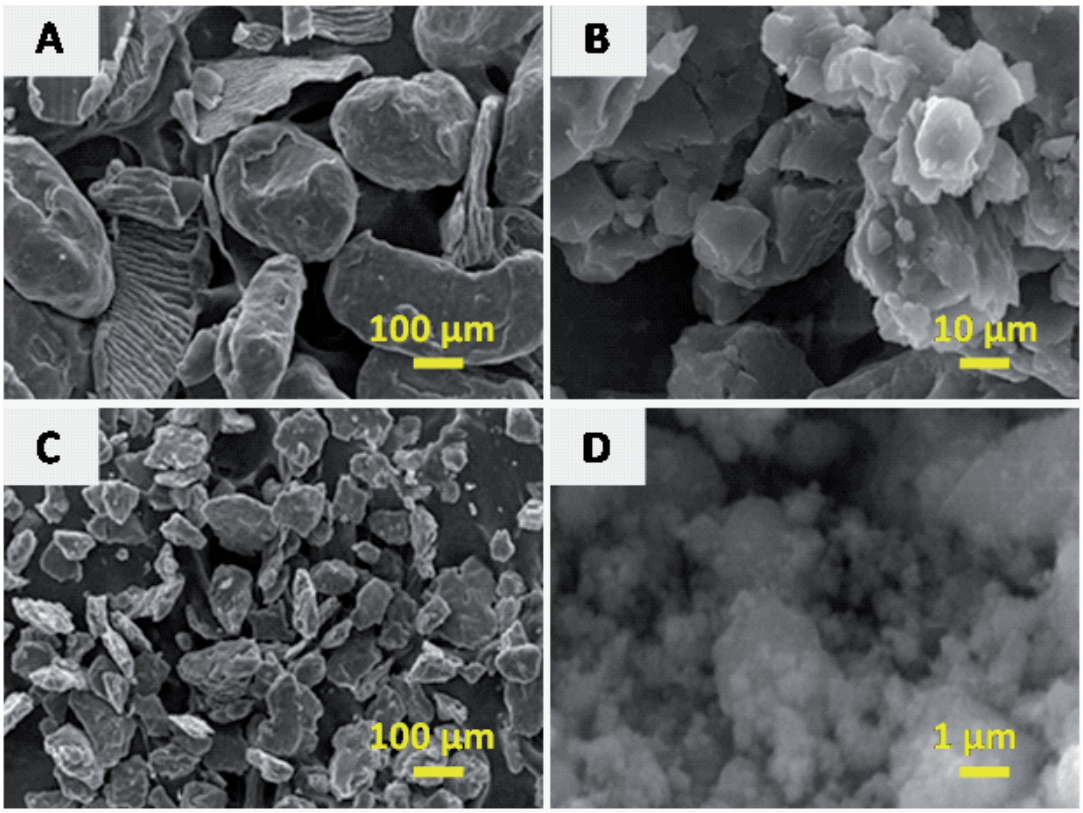

Figure 2: SEM image of Mg-2wt% SiC, (A)before milling, (B) 60, (C)30 and (D) 180 h.

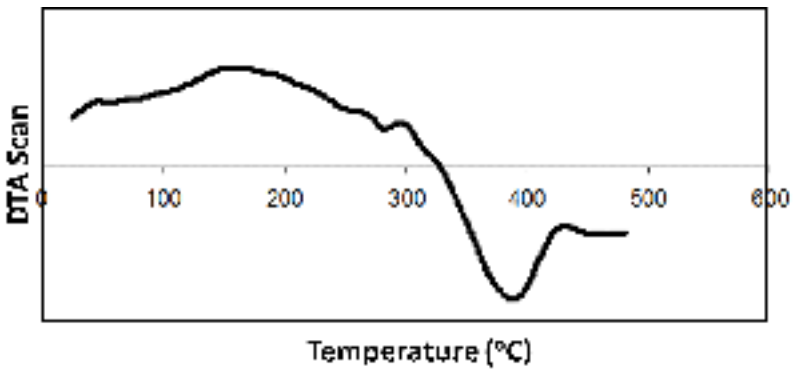

Figure 3: DTA scan of Mg-2wt% SiC after milling 180 h.

Thermal investigation for the sample after 180 milling give information on the state of the material decomposition. Here, it can be clearly seen that the material decomposed at temperature 330◦C (Figure 3). However, this temperature is still high for Mg-based hydrogen storage materials application. It seem that the crystallite size have not a strong influence on the desorption temperature changes.

Kurko et al. [10], explained that this situation occurred due to agglomeration of the material during the milling process. Therefore the reduction of agglomeration is believed will improve the thermal properties of Mg-based hydrogen storage materials.It is already known that the hydrogen storage properties of light metal hydrides is improved with reduced particle agglomeration [11]. Due to the decrease agglomeration

will shorten the diffusion path of hydrogen atoms. Therefore, it is suggested that a significant amount of strain, disorder and defects should be created during milling process, which shorten hydrogen diffusion paths [12].

4. Conclusions

Mg catalyzed with 2 wt% of silicon carbide (SiC) prepared via vibratory ball milling has been successfully done. This process results in high surface area powders with finely dispersed SiC- particles on the surface of Mg. The Mg-SiC material exhibits a microstructure in nanometer scale. Thermal investigation for the powders after 180 millingshown that the material decomposed at temperature 330◦C. Due to these results, nanocrystalline Mg-SiC is suggested a further work for improvement the sorption properties and the kinetics of Mg-based hydrides as hydrogen storage material.

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