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$MG_{17}AL_{12}$ INTERMETALLIC PREPARED BY BULK MECHANICAL ALLOYING

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Abstract

In a last decade, nano-crystallized or amorphous materials are widely investigated. Such materials provide other directions for solving several issues in the field of materials science. Ball milling processes are general tools for preparing the nano-crystallized or amorphous material. The samples, however, show the fine powder-form and the most of those are very sensitive against circumstance around. The bulk-form samples are desirable. The present work proposes the newly developed bulk mechanical alloying (BMA) technique as an alternative method for the ball milling. Successful solid-state reaction and the formation of nano-scale crystalline of $Mg_{17}Al_{17}$ phase will be demonstrated.

Introduction

A ball-milling technique is available as a powerful tool for the preparing of nano-crystalline or amorphous materials. In the case of hydrogen storage materials, for instance, the nano-crystallizing or amorphorization modifies thermodynamic and / or kinetic properties upon hydrogen absorption-desorption processes [1-4]. However, the ball-milling process provides the specimen as fine powder-form, which leads the very sensitive surface for the contaminations from the atmospheres around. The yield of a final sample by ball-milling process is, in addition, significantly low as compared with an amount of starting material.

Bulk Mechanical Alloying (BMA) process has been proposed as an alternative technique for the traditionally applied ball milling method [5, 6]. The loaded materials travel through the different diameters of a die cavity cell [7], and the plastic deformation takes place on the cycle of compaction-withdraw-extrusion-withdraw-ejection as shown in Figure 1. By repeating this cycle, finally, the samples can be received as nano-crystalline or amorphous materials. Previous reports on the synthesis of promising candidates of hydrogen storage media, nano-crystalline Pd [3], Mg_2Ni [4, 8] and $Mg_{17}Al_{12}$ [9], by the BMA demonstrated the possibility of BMA as a candidate on the novel solid-state reaction technique. Interest in BMA as desirable mechanical alloying process has emerged due to the received samples that show bulk-form. During BMA process, the sample contact with only the inner wall of cell and upper / lower punches, and the loss of the sample is efficiently low.

The present work shows more detailed on the material processes of BMA from the synthesis of $Mg_{17}Al_{12}$ intermetallic.

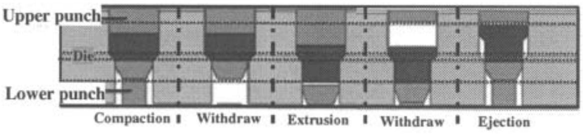

Figure 1. Schematic diagram of typical process for BMA.

Experimental

$Mg_{17}Al_{12}$ intermetallic was prepared by BMA technique. BMA set-up was supplied from KYB Co., Ltd, Japan. Actual set-up of BMA is shown in Figure 2. A stoichiometric mixture of Mg and Al powders with an amount of 5 g was loaded into a die cavity with 20 mm diameter of the die-set as a starting material. The sizes of Mg or Al powder were less than 180 µm or 300 µm, respectively. The purity grade of those was higher than 99.9 %. One compaction-withdraw-extrusion-withdraw-ejection cycle was controlled at the pressing 100 kN for 8 s (see Figure 1). The cycle was repeated for 100, 500, 1000 and 2000 cycles in air with water-cooling system at 298 K.

The formation of $Mg_{17}Al_{12}$ intermetallic phase was characterized by powder X-ray diffraction (XRD). Wavelength dispersive X-ray spectrometry (WDX) and transmission electron microscope (TEM) measurements were made for the confirmation of homogeneous distributions and microstructure of the alloy.

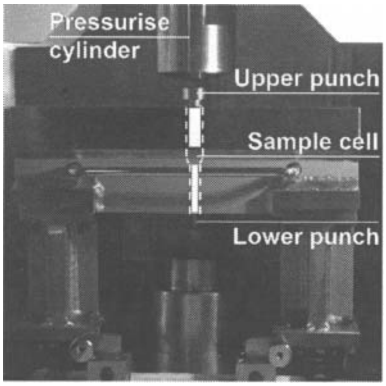

Figure 2. A photograph of BMA set-up.

Results and Discussion

Figure 3 shows a photo image of Mg-Al alloy prepared by BMA process. The sample shows bulk-form, and the sample yielded almost 100 % (5 g) as compared with an amount of starting materials. These are obviously advantages of BMA technique since further handling care for contaminations is not necessary. With increasing repeating cycles, the XRD profile shows broaden peaks, and small contributions from $Mg_{17}Al_{12}$ intermetallic phase appeared at 1000 cycles as shown in Figure 4. Finally, the single phase of the intermetallic $Mg_{17}Al_{12}$ can be seen at 2000 cycles (approximately 4.4 h). In previous work, 2 g of $Mg_{17}Al_{12}$ intermetallics (rate of yield was 80-90 %) was received by a high energy ball-milling for 5 h [10]. BMA can be emphasized as a pronouncedly effective mechanical alloying technique.

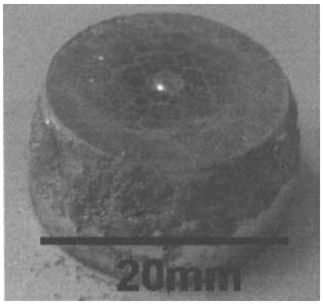

Figure 3. Photo image of Mg-Al alloy after BMA process. The sample shows bulk-form.

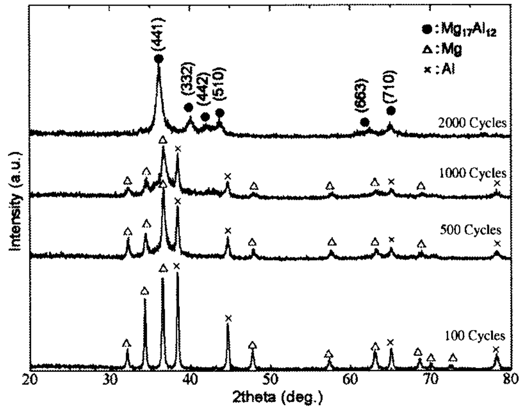

Figure 4. Collected XRD profiles of Mg-Al alloy with cyclic BMA showing (x) Al, (Δ) Mg and (●) $Mg_{17}Al_{12}$. Miller indexes of $Mg_{17}Al_{12}$ are shown together.

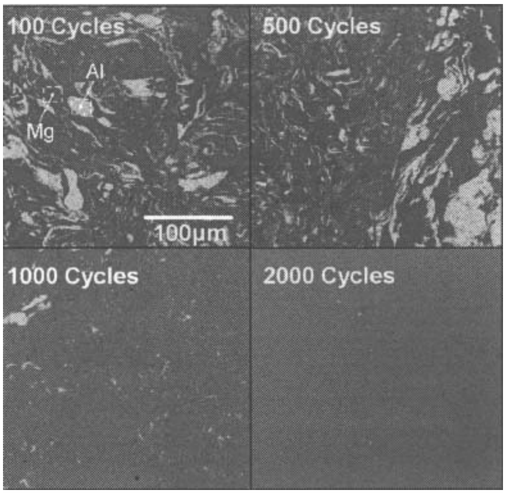

Figure 5. WDX images of Mg-Al alloy with cyclic BMA. The distributions of Mg and Al in alloy can be seen.

The changes in the elemental distributions of Mg / Al during cyclic BMA were monitored by WDX (see Figure 5). The proceeding of the alloying is clearly seen. The increases of cyclic BMA provide homogeneous distribution of Mg and Al, and the formation of intermetallic $Mg_{17}Al_{12}$ phase completely takes place at 2000 cycles. This is well agreed with the data from XRD profiles (see Figure 4). It should be noted as one of advantages in BMA that no significant impurities were found by WDX profile. In ball-milling process, the contamination from vial / balls or atmosphere is a common problem. On the process of BMA, the contamination from cell may be less likely, because the sample contacts with only inner wall of cell during the extrusion-compression cycles.

TEM observation was made for the sample at 2000 cycles as shown in Figure 6. The electron scattering rings are also presented. The crystalline shows around 20-30 nm scales. This concludes that BMA drives nano-crystallines upon mechanical alloying processes.

Conclusion

In this study, nano-crystalline $Mg_{17}Al_{12}$ intermetallic phase was successfully driven by newly developed bulk mechanical alloying (BMA) technique. The alloying time is rather short compared with conventional ball-milling, and the sample yields effectively. For the further possibility as attractive alternatives of ball-milling type technique, the controlling of atmosphere may be required. This is under development for the materials that is very sensitive against oxygen or water, such as Li-based alloy [7].

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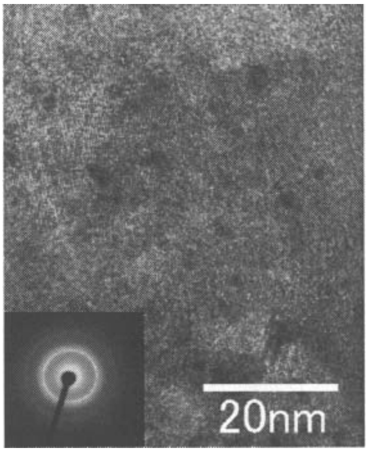

Figure 6. TEM image of $Mg_{17}Al_{12}$ intermetallic microstructure at 2000 BMA cycle. The electron scattering rings are presented together.

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