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Bulk Mg–3Al–Zn alloy with ultrafine grain size produced by powder metallurgy

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ABSTRACT

Ultrafine-grained Mg–3Al–Zn alloys with an average grain size of 180 nm have been made by powder metallurgy. First, the nanocrystalline powders with mean grain size of 45 nm were produced by ball milling under argon atmosphere, and then through vacuum hot pressing at 633 K for 40 min and warm extrusion at 373 K, bulk solid samples were compacted successfully from the mechanically milled powders, and the relative density of the samples was about 98.87% (1.8003 g/cm^3). XRD, SEM and TEM analysis showed that the microstructure of the samples consists of homogeneous equiaxed grains and grain growth has taken place during the consolidation process.

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1. Introduction

Nanocrystalline (NC) and ultrafine-grained (UFG) materials have received considerable attention within the last decade, owing to their improved properties as compared with conventional coarse-grained materials [1–4]. For Mg alloys, NC and UFG microstructures are more attractive because of their high k_y values [5,6]. For example, when the grain size was reduced to about 500 nm, the $MgZn_{3.3}Y_{0.43}$ alloy showed a yield stress of 410 MPa with a 12% elongation [3]. It is significant to mention here that with grain size refinement, ductility has also been found to increase for Mg alloy [6]. The UFG Mg materials (with grain sizes less than 1 μm) have been achieved by a number of different techniques, such as equal channel angular extrusion (ECAE) [7], friction stir processing (FSP) [2], accumulative roll bonding (ARB) [8], high-pressure torsion (HPT) [9] and powder metallurgy (PM) [10]. It has been demonstrated that powder metallurgy process can further refine grain size of Mg alloy, compared to severe plastic deformation (SPD) [2,7–9]. Lu et al. [10–15] have successfully proved that PM was an effective technique to synthesize high strength UFG Mg alloys. But for pure Mg or solute solution hardened Mg alloys (such as AZ31) with a low content of alloying elements, it is difficult to achieve an UFG microstructure due to the rapid growth kinetics of the single-phase grains. For example, Jin et al. [7] cannot fabricate the UFG AZ31 alloys by ECAE. The original coarse grain size could be reduced to less than 16 μm after extrusion and was further only refined to around 1.9 μm after subsequent 8-pass ECAE at 498 K. The UFG AZ31 could not be obtained even after 8-pass ECAE. The present study is an attempt that focuses on the synthesis of UFG Mg–3Al–Zn alloys by powder metallurgy. Moreover, the microstructure characterization of the powders and bulk alloys are investigated.

2. Experimental procedures

The alloy powder with a nominal composition of Mg–3Al–Zn was produced by high energy mechanical milling (HEMM) of a mixture of Mg powder (99%, 325 mesh), Zn powder (99%, 325 mesh) and Al powder (99.5%, 325 mesh) under high purity argon (99%). The milling was carried out with a QM21SP4 planetary ball milling machine. Before the formal experiment, the prepare experiment was carried out to determine the optimum ball milling parameters. The optimum ball milling parameters were following: shaft rotation was 360 rpm, ratio of ball to powder was 40:1 and milling time was 20 h. Prior to ball milling, the vial was sealed in a glove box filled with high purity argon (99.9% pure) to ensure the milling was done under an inert atmosphere. After milling, the powders were canned into an Al container of 35 mm in diameter and about 65 mm in length to avoid oxidation. The cans were sintered for 40 min at 633 K and then pressed in a vacuum furnace under a vacuum better than 1×10^{-2} Pa. The sintered compacts were extruded at 373 K, under an extrusion ratio 6.25 and an extrusion rate 22 mm/s using graphite as lubricant. To prevent the alloy from oxidation, the experimental materials were always kept in a glove box filled with pure argon (99.9%). The microstructure characterizations of the powders and extruded alloys were conducted on XRD, SEM and TEM, due to their small grain size. Density (ρ) measurements were performed in accordance with Archimedes' principle. Four randomly selected extruded rod samples were tested and the average density was calculated. Distilled water was used as the immersion fluid. The samples were weighed using an ESJ200-4 electronic balance, with an accuracy of ±0.0001 g.

3. Results and discussion

3.1. Microstructure and grain size of the powders

Fig. 1 shows the morphology of the nanocrystalline powders used for consolidation. The powder particle after 20 h ball milling

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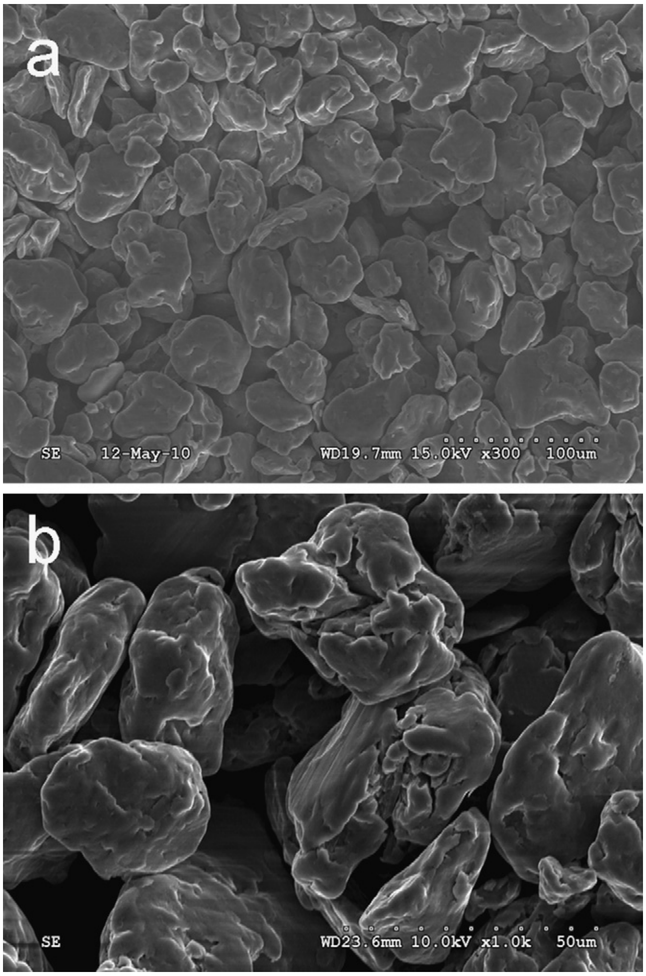

Fig. 1. SEM micrographs of ball milled Mg–3Al–Zn powders obtained after 20 h milling.

is flake-shaped and a mean diameter approximately 30 μm. The appearance of powder is smooth and the size is uniform. However, in spite of the room temperatures, as shown Fig. 1b, the milled particles tend to form agglomerates due to cold welding.

The bright and dark field TEM micrographs and selected area electron diffraction (SAED) pattern, obtained from region with a diameter of 800 nm of the powders milled for 20 h are shown in Fig. 2. As shown in Fig. 2, after 20 h milling, the nanocrystalline powders are mainly composed of equiaxed grains of 30–50 nm surrounded by a few smaller grains (<20 nm). The grains and grain boundaries are very clearly observed. It is also possible to find small inclusions (approximately 2–6 nm) spaced approximately 20–100 nm apart. The inclusions are deduced to be oxide particles originating from the surface films of the initial powder that are broken down during milling. The SADP taken from the as-milled alloy exhibits the rings of diffracted spots, indicating the presence of boundaries with high angle of orientation. These suggest that the dynamic recrystallization occurs during milling. Fig. 2c shows the grain size distribution of the milled specimen, which is summarized from measuring 200 grain diameters in bright field images. It shows that the grain sizes are mostly scattered from less than 20–60 nm, and more than 80% of the grains are refined to less than 50 nm. In this paper, the average grain size of magnesium alloy powders attainable by mechanical milling at room temperature is approximately 45 nm. The grain size value is also coincident with previous report of milled pure Mg [16].

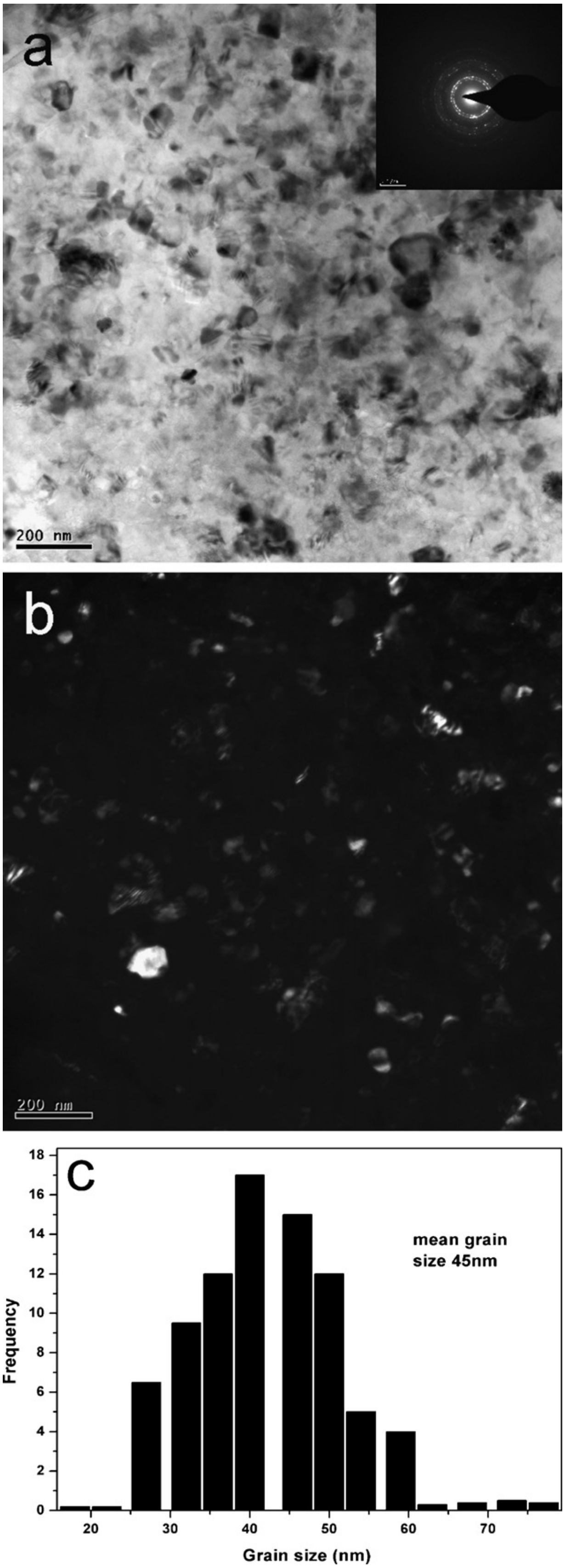

Fig. 2. TEM micrographs of Mg–3Al–Zn powders ball milled for 20 h: (a) bright field image (with SAED pattern as an insert), (b) dark field image, and (c) grain size distribution (mean grain size = 45 nm).

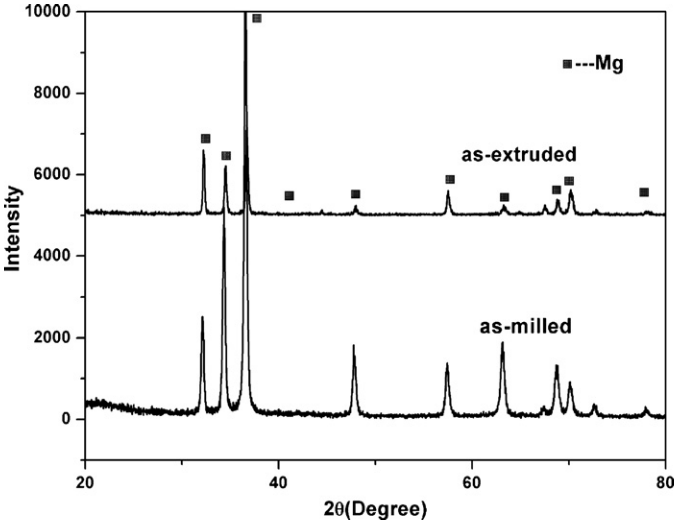

Fig. 3. XRD patterns of both the as-milled and the subsequently as-extruded Mg–3Al–Zn alloy.

Fig. 3 presents the XRD diffraction patterns of the milled powders and the extruded alloys. In both cases, all of the diffraction peaks are readily indexed to various crystal planes of the hexagonal phase Mg. No peaks for other phases can be detected, which indicates that the Al, Zn elements absolutely dissolve into the Mg matrix and form Mg based solid solution. The symmetry of the diffracted peaks before and after consolidation suggests that no precipitation occurs during the consolidation process. The diffracted peaks become slightly narrow after consolidation, which indicates that grain growth takes place. Prior to consolidation, the mean grain size of the milled powder calculated from the line broadening in XRD data using the Scherrer equation is about 42 nm. This average grain size value is similar to that observed directly from the TEM micrograph, which is 45 nm. In addition, the mean crystalline size of extruded sample calculated from XRD is about 100 nm. Comparison of the grain size of the powder and the bulk, we can see that grain size has been doubled during consolidation process. Though the grain growth takes place during consolidation process, the grain size of the bulk still demonstrates that the feasibility of achieved an UFG structures Mg–3Al–Zn by powder metallurgy.

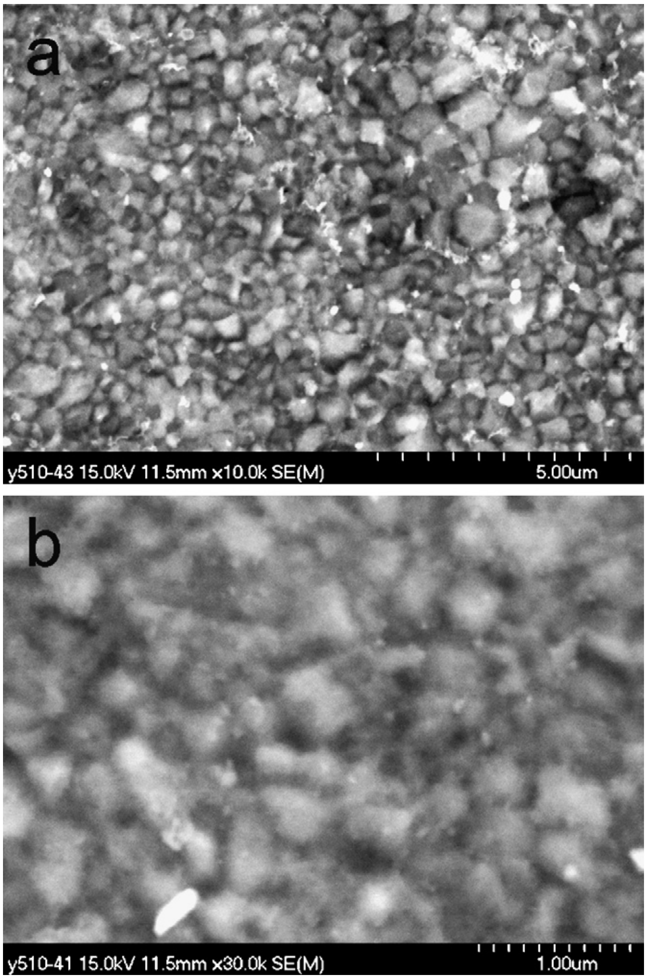

Fig. 4. SEM micrographs of the Mg–3Al–Zn alloy after extrusion at 373 K with 22 mm/s (a) at low magnification showing the uniform UFG structure and (b) at higher magnifications showing ultrafine grain..

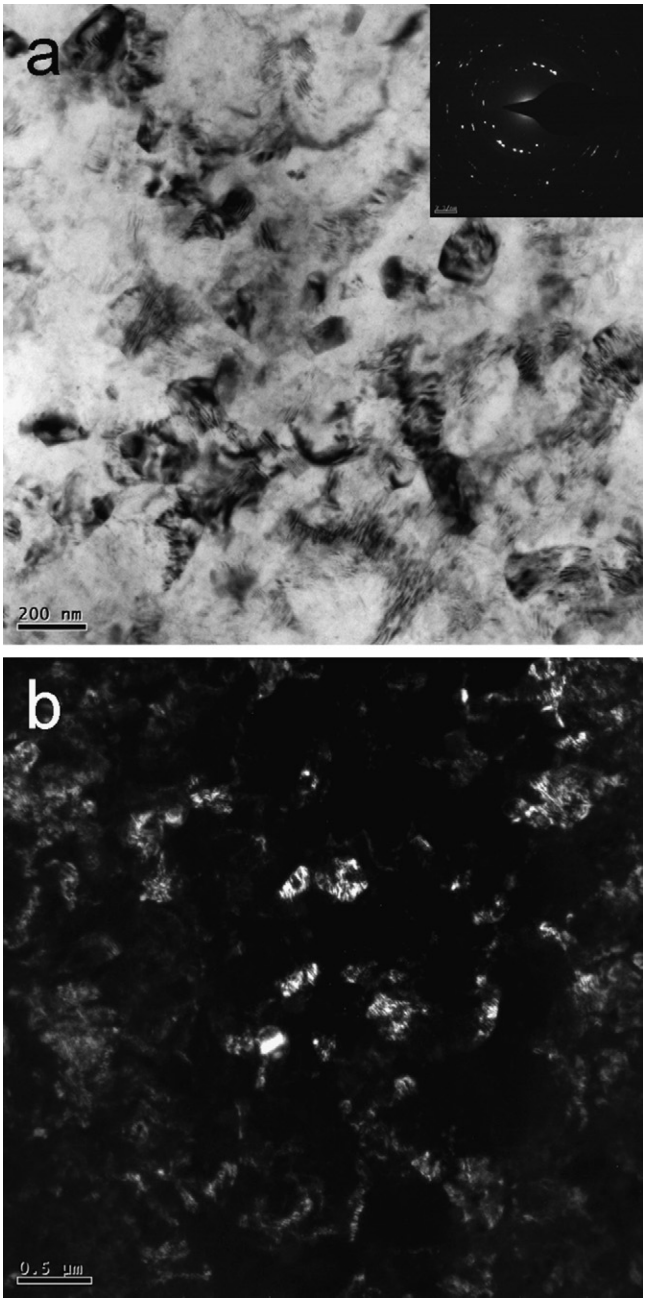

Fig. 5. TEM investigations of the as-extruded Mg–3Al–Zn alloy: (a) bright field image (with SAED pattern as an insert) and (b) dark field image.

After consolidation, the dense structured rods are extracted from the Al tubes. The average density for four tested samples is $1.8003 \pm 0.006\,g/cm^3$. Since the nominal density of an Mg–3Al–Zn alloy is $1.82088\,g/cm^3$, the consolidated samples have theoretical density of $98.87 \pm 0.3\%$. It is also worth mentioning that no macroscopic pores or cracks are observed throughout the samples.

3.2. Structure and grain size of bulk alloy

Fig. 4 shows grain structures of the as-extruded Mg–3Al–Zn alloy. Viewed at low magnification, Fig. 4a shows a well-defined equiaxed and highly homogeneous nature in the observed region. Viewed at higher magnifications, Fig. 4b shows an UFG structure is mainly composed of equiaxed grains of 150–180 nm surrounded by a few smaller grains (<80 nm) and some larger ones (about 300–500 nm). Compared with SPD processes, such as accumulated roll bonding (ARB) [8] or equal channel angular extrusion (ECAE) [7], the UFG microstructure obtained in the present study has clearer grain boundaries and smaller sizes without abnormal local grain growth.

TEM observations are employed to further analyze the microstructure of bulk Mg–3Al–Zn. The TEM images of bulk Mg alloy extruded at 373 K are shown in Fig. 5. The grains are almost equiaxed and the average grain size of the extruded sample is about 180 nm. The grain structures mainly consist of lamellar structure, and the lamellar interiors contain quite high density of dislocations. It is noticeable that there are no voids or pores at the interfaces and the powder particles are well bound. The same result was obtained by Garcés et al. [17]. They have observed AZ92 alloy processed by powder metallurgy and found that the samples extruded at 253 K having the average grain size 600 nm. The grain size of the bulk obtained by TEM is similar to that observed from the SEM micrograph, but these calculated by XRD data using the Scherrer equation is about 100 nm. Thus, the grain size for extruded sample observed directly from the TEM micrographs is about 1.5–2.0 larger than XRD result. This size difference is attributed to the influence of strain on the XRD result.

4. Conclusions

In summary, ultrafine grain structure can be produced in Mg–3Al–Zn alloy by powder metallurgy. The mean grain sizes of the bulk can be refined to about180 nm, which is much smaller than these of the ARB or ECAP Mg–3Al–Zn alloy. The results can be summarized as follows:

(1) Nanocrystalline Mg–3Al–Zn alloy powders have been successively developed by 20 h ball milling under argon atmosphere. The mean grain size of the powder observed from TEM micrographs is about 45 nm, which is similar to XRD result.
(2) The UFG bulk Mg–3Al–Zn alloy has been produced by consolidated nanocrystalline alloy powders using a combine of vacuum hot-pressing (for 40 min at 633 K) and warm extrusion (at 373 K with 22 mm/s). The average grain size of alloy observed by TEM and SEM micrographs is about 180 nm, which is 1.5–2.0 larger than XRD result.
(3) After consolidation, the consolidated samples have theoretical density of $98.87 \pm 0.3\%$.

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