



Formation, microstructure and properties of long-period order structure reinforced Mg-based bulk metallic glass composites

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Abstract

In situ Mg–Cu–Y–Zn bulk metallic glass (BMG) matrix composites, in which Mg solid solution flakes of 0.5–1 μm thickness and 2–10 μm length are dispersed, have been prepared by copper mold casting. The Mg flakes are characterized as a long-period order structure (LOS), i.e. periodic arrays of six close-packed planes distorted from the ideal hexagonal lattice of 6H-type. The formation mechanism of LOS is interpreted as the precipitation of the leading phase of the eutectic reaction above the glass transition temperature. In comparison with monolithic Mg-based BMG alloys, the composites with an LOS exhibit significant improvement in mechanical properties, e.g. a compressive plastic strain of $\sim 18\%$ and ultimate strength of ~ 1.2 GPa, have been measured in $\text{Mg}_{81}\text{Cu}_{9.3}\text{Y}_{4.7}\text{Zn}_5$ alloy. It is suggested that the enhancement of the mechanical properties of the composites can be attributed to the generation of multiple shear bands and the deformation of the LOS.

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1. Introduction

Since the discovery of ternary Mg–Ni–Ce bulk metallic glass (BMG) by Inoue et al. [1], Mg-based BMG alloys with a critical thickness on the millimeter to centimeter scale have been fabricated in the Mg–TM–RE (TM = Cu, Ni; RE = Ce, La, Y, Nd) [2–8], Mg–Cu–Y–M (M = Al, Ag, Zn) [9–11] and Mg–Cu–Ag–Pd–Y alloy systems [12]. Recently, great progress has been made in the glass-forming ability (GFA) of Mg-based BMG alloys. Xi et al. [7,8] found that the addition of rare earth elements (RE = Gd, Pr, Nd, Tb, Y, Dy) can significantly improve the resistance to the deterioration of glass forming ability and manufacturability of Mg-based alloys caused by oxygen in the environments. $\text{Mg}_{65}\text{Cu}_{25}\text{RE}_{10}$ (RE = Gd, Tb, Dy) BMG rods with a diameter of 5 mm can be easily prepared under an atmosphere of 1×10^{-2} Pa. Ma et al. [11] have successfully synthesized a

new quaternary $\text{Mg}_{54}\text{Cu}_{26.5}\text{Ag}_{8.5}\text{Gd}_{11}$ BMG alloy with the maximum sample diameter as large as 25 mm by using the copper mold casting method. In comparison with Mg-based crystalline materials, Mg-based BMG alloys exhibit great improvement in strength property. For example, $\text{Mg}_{65}\text{Cu}_{20}\text{Zn}_5\text{Y}_{10}$ [9] and $\text{Mg}_{65}\text{Cu}_{15}\text{Ag}_5\text{Pd}_5\text{Y}_{10}$ [12] BMG alloy exhibit compressive strengths of ~ 650 and ~ 800 MPa, respectively, which are more than twice that of ordinary crystalline Mg alloys. The excellent GFA and high strength of Mg-based BMG alloys, together with their low density, relative low cost, abundant deposits (Mg is the eighth most abundant element in the earth's crust and the third most plentiful element dissolved in seawater) and easy recycling ability make the Mg-based BMG alloys particularly attractive for engineering applications such as weight reduction and higher fuel efficiency transportation [13].

Like other BMG alloys, Mg-based BMGs have also been found to be brittle due to their inhomogeneous deformation behavior. They always fracture in the elastic regime without observable plastic deformation. It was reported

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that the notched fracture toughness K_C of Mg-based BMG is even as low as $2.0 \text{ MPa } \sqrt{m}$, which approaches that of ideal brittle silicate glasses [7,14,15]. Therefore, the absence of distinct plasticity in Mg-based BMG alloys has become one of the challenging problems for their engineering application.

In the efforts to overcome the brittleness of BMG alloys, the preparation of BMG matrix composites by using in situ methods has been proven to be an effective way. The reinforcing phases in BMG matrix composite are expected to hinder the propagation of shear bands and promote the formation of multiple shear bands, hence retarding the emergence of catastrophic failure in BMG matrix. In this subject, the first discovered reinforcement is the ductile crystalline Ti–Zr–Nb β phase, which is formed in situ in Zr–Ti–Nb–Cu–Ni–Be bulk metallic glass matrix and results in a dramatic increase of plastic strain of $\sim 5\%$ in the composite [16]. To date, in situ BMG matrix composites have been prepared in Zr–Nb–Ti–Cu–Ni–Be [16–18], Zr–Al–Ni–Cu–Nb [19], Zr–Cu–Ni–Al–Ta [20], Cu–Zr–Ti–Ta [21], La–Al–Ni–Cu [22], Pd–Ni–Cu–P [23] and Ti–Cu–Sn–Ta [24] BMG or nanostructured matrix alloys. All of the above composites have exhibited distinct plasticity ranging from 5% to 14.5% at room temperature. For Mg-based BMG alloys, however, ideal reinforcement has not been found until now. Ma et al. [25] synthesized $\text{Mg}_{65}\text{Cu}_{7.5}\text{Ni}_{7.5}\text{Zn}_5\text{Ag}_5\text{Y}_{10}$ BMG matrix in situ composites containing iron as second phase dispersions. The macroscopic plasticity of the composite with a 13 vol.% fraction of iron particles is only about 1%, although the fracture strength is increased to 990 MPa. The plastic strains to failure of Mg-based composites with ex situ TiB_2 particles [26] were found to reach the order of 2–3%. The reason why the Mg-based BMG composites fail to exhibit distinct plasticity may be attributed to their intrinsic brittleness and the unapt selection of the second phase.

A long-period order structure (LOS) has been found in $\text{Mg}_{91}\text{Y}_2\text{Zn}$ and $\text{Mg}_{88}\text{Y}_8\text{Zn}_4$ alloys processed by non-equilibrium synthesis or the cast method [27–33]. It has been proven that these alloys exhibit a maximum tensile yield strength of $\sim 600 \text{ MPa}$ and elongation of $\sim 5\%$. So it is expected that the LOS may improve the plasticity and strength of the BMG alloy if the LOS is uniformly dispersed in the amorphous matrix. To date, however, no work has been reported to introduce the LOS into the glass matrix of Mg-based BMG alloys. In this work, by appropriate composition design and good control of the solidification conditions, we present novel LOS reinforced quaternary Mg–Cu–Zn–Y BMG matrix composites with pronounced plasticity and high strength. Under the consideration of the competitive growth theory of eutectic alloys and the formation of an amorphous phase, the formation mechanism of the LOS is discussed. Meanwhile, the deformation behavior of the composites is also investigated on the basis of the inhomogeneous plastic flow of BMG alloys.

2. Experimental

The nominal composition of the alloys studied in this work is shown in Fig. 1. The starting materials used in preparing the Mg–Cu–Zn–Y alloys were high purity metals: Mg (99.99%), Cu (99.9%), Zn (99.9%) and Y (99.9%). Cu–Y intermediate alloys were first arc melted under a Ti-gettered argon atmosphere. Then Mg–Cu–Y–Zn master ingots of size $\Phi 60 \times 100 \text{ mm}$ were prepared in an electrical resistance furnace under air by using pre-alloyed Cu–Y intermediate alloy, and pure Mg and Zn metals. The plate samples with $\Phi 10 \times 3 \text{ mm}$ particle size were prepared using air-cooling technology. Rod samples of 3 and 5 mm diameter and 70 mm length were fabricated by using a water-cooled copper mold casting method.

The as-cast rods were examined by X-ray diffraction (XRD) in a PHILIPS APD-10 diffractometer ($\text{Cu } K_\alpha$ radiation). A LEO1450 scanning electron microscope (SEM), a 200CX transmission electron microscope (TEM) and a JEM-2010F high-resolution electron microscope (HREM) were employed to analyze the microstructure and fracture morphology of the composites. The volume fraction of the phase formed in situ in the specimen was detected using the QWIN image analysis system. The density of the as-cast samples was determined using the Archimedes' method.

To test the mechanical properties of the composite, the as-cast Mg–Cu–Y–Zn rods were cut to 2 mm in diameter. The resulting compression test samples were 4 mm in height and 2 mm in diameter. For $\text{Mg}_{71}\text{Cu}_{16}\text{Y}_8\text{Zn}_5$ alloy, we employed as-cast rods 3 mm in diameter and 6 mm in height as the compressive samples. Room-temperature compression tests were carried out by using a strain rate of 10^{-4} s^{-1} . In order to measure the deformation displacement accurately, we used extensometer and two bulk tungsten carbides with the size of $20 \times 15 \times 6 \text{ mm}$ as compressive holds. The extensometer was fixed up on the

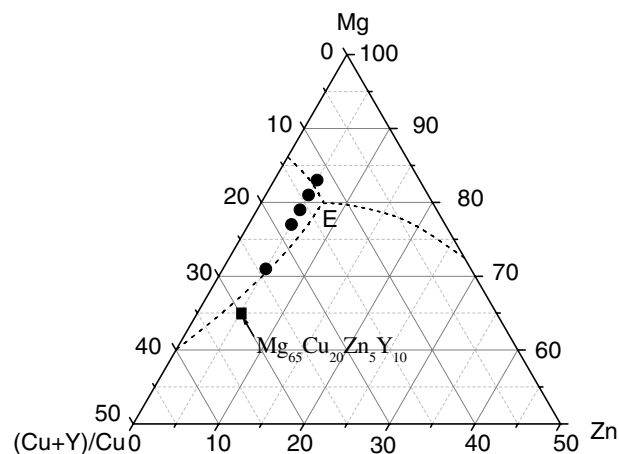


Fig. 1. Pseudo-ternary Mg–(Cu + Y)–Zn diagram showing the alloy compositions studied (represented by solid circles) and the position of the eutectic reactions for ternary Mg–Cu–Zn alloy (represented by short dashed lines).

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