Structural and mechanical characterizations of ductile Fe particles-reinforced Mg-based bulk metallic glass composites

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In this study, the atomic composition of Mg58Cu28.5Gd11Ag2.5 which possess high glass forming ability (GFA) was selected as the matrix alloy for synthesizing the ductile Fe particles-reinforced Mg-based bulk metallic glass composites (BMGCs) by using injection casting method. The results show that the compressive ductility increases with the volume fraction of Fe particles and reaches up to the compressive plastic strain of 8% for the Mg-based BMGC with 20 vol.% Fe. In parallel, multiple-shear bands were revealed on the sample surface which is near the fracture area. This suggests that these Fe particles can branch primary shear band into multiple-shear bands and decrease the stress concentration for further propagation of shear band, and so as to enhance plasticity. Additionally, a slightly decreasing in yield strength with increasing the addition of Fe particles was found presumably due to the formation of brittle Fe2Gd intermetallic phase at the interface between the Fe particle and amorphous matrix. In overall, the ductile Fe dispersoids would provide a good toughening effect in promoting the practical use of amorphous materials.

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1. Introduction

Mg-based bulk metallic glasses (BMGs) were first discovered in the 1990s, are of interest because of their low densities as compared to most other amorphous alloys, such as the Pd--, Zr-, and Fe-based amorphous alloys [1–3]. However, the monolithic Mg-based BMGs have been found to be the most brittle among all BMGs; they tend to break into pieces before yielding [4–7]. To amend the weakness, Zheng et al. [8] predicted the composition of monolithic Mg-based BMGs by using the high μ/B ratio concept; however, the resulting BMGs do not display the manifesting ductility like Pt- and Zr-based BMGs. Therefore, many efforts have devoted to develop Mg-based bulk metallic glass composites (BMGCs) to increase the plasticity of Mg-based BMGs. These include in situ precipitated, ductile metal, and refractory ceramic particles-reinforced Mg-based BMGs, which possess improved compressive strengths and demonstrates the remarkable plastic strains [2,9–13].

More recently, many investigations have proposed that the compressive ductility of BMGs will be improved significantly by dividing the amorphous matrix of BMGs into compartments on micrometerscale [14,15]. The small regions limit the propensity of forming mature shear bands. In fact, each stand-alone compartment can undergo great deformation without failure, as reported in the previous studies [8,16–19]. Following the idea, the authors had successfully explored another method to toughen Mg-based BMGs by adding porous Mo particles into the amorphous matrix [13]. The porous Mo particles act both as ductilizers (as in composites) and as “micro-scale compartment” formers in the particles, and so as to improve the plasticity of the Mg-based BMGs. Accordingly, in this study, the composition of Mg58Cu28.5Gd11Ag2.5 which possess high GFA (γ = 0.421, γ = Tl/(Tg + Tl) [20] and γm = 0.749. γm = (2Tf − Tg)/Tl [21]) is selected as the raw alloy for preparing the BMGC by reinforcing with relatively cheaper ductile Fe particles (which has irregular shape) instead of Mo particles. The effect of particle content, size effect of BMGC rods, and the casting condition on the mechanical properties of the Mg-based BMGCs is discussed in this article.

2. Experimental

The composition of Mg58Cu28.5Gd11Ag2.5 was selected as the raw alloy for preparing the BMGs. High purity Cu and Gd (>99.9%) were pre-alloyed into Cu–Gd alloy ingot by arc melting in a Ti-gettered argon atmosphere. Then, the Cu–Gd alloy was melted together with high purity Mg and Ag pieces to obtain the target composition by induction melting under argon atmosphere. While melting, high purity Fe particles with irregular shape and size of 50–70 μm were added into the matrix alloy under argon atmosphere. Mechanical stirring was exerted to enhance the homogeneous mixing of the particles with the melt. The fraction of Fe particle ranges from 5 vol.% to 20 vol.%. Furthermore, the composite alloy ingot was remelted by
induction melting in a quartz tube and injected into a water-cooled Cu mold by argon pressure to obtain BMG composite rods with sizes of 2–4 mm in diameter. The thermal properties of the monolithic BMG and BMGcs were characterized by TA Instruments DSC 2920 differential scanning calorimeter (DSC) under flowing purified argon with a heating rate of 20 K/min. The structure of the specimen was characterized by Scintag X-400 X-ray diffractometer with monochromatic Cu Kα radiation. The compression tests were performed at a strain rate of $5 \times 10^{-4}$ s$^{-1}$ by a MTS 810 mechanical test system. The compression samples with the height to diameter ratio of 2:1 ($h = 8 \text{mm}/d = 4 \text{mm}$ and $h = 4 \text{mm}/d = 2 \text{mm}$, respectively) were cut to be parallel and carefully polished to insure the flatness from the as-cast BMG and BMG composite rods. The distribution of Fe particles in the amorphous matrix of BMGcs and the morphology of fracture surface were examined by a Hitachi S-4700 field emission scanning electron microscope (FEG-SEM) with EDS capability.

3. Results and discussions

In Fig. 1, the result of DSC revealed that a clear $T_g$ before crystallization for all of the as-cast Mg-based BMGcs with different vol.% of Fe particles. Since the Fe element is immiscible with Mg and Cu elements due to their different crystal structure. Therefore, these Mg-based BMGcs with Fe particles would keep almost the same thermal properties as the based Mg$_{58}$Cu$_{28.5}$Gd$_{11}$Ag$_{2.5}$ BMG, presenting the identical glass transition temperature ($T_g$), crystallization temperature ($T_x$), and supercooled liquid region ($\Delta T_x$). The GFA of all these Mg-based BMGcs with different vol.% Fe dispersoids exhibits nearly the same GFA about $\gamma = 0.421$ and $\gamma_m = 0.749$. In addition, the XRD pattern of the composite with 15 vol.% Fe particles demonstrates that the amorphous phase causes the broadening diffused humps. No apparent crystalline peaks are detected in the pattern except the high intensity crystalline peaks of Fe particles, as shown in Fig. 2. This implies that the addition of Fe particles does not affect the matrix composition of these Mg-based BMGcs and the composition of the matrix is suggested nearly the same as the master alloys and no obvious chemical reaction has been taken place to reduce the glass forming ability. A typical amorphous image of high resolution TEM was revealed in the matrix of the composite, as illustrated in Fig. 3.

The SEM observations of the polished cross-sectional surface of the composite containing different vol.% Fe particles are shown in Fig. 4. The Fe particles, in the range of 40–100 μm in size, not only disperse homogeneously in the metallic glassy matrix, but also exhibit a very good bonding condition with the amorphous matrix as shown in Fig. 4. The final volume fraction of the Fe particles in the composite which is estimated by the image analysis was found very close to the added vol.% of Fe particle as listed in Table 1.

Compression tests indicate an improvement in plastic strain and stress for the Fe particles-reinforced Mg-based BMGs. The 20 vol.% Fe contented Mg-based BMGC exhibits large ductility enhancement with compressive plastic strain up to 7.2% and the yield and ultimate compression stress up to 0.8 and 0.96 GPa, respectively as shown in Fig. 5. Multiple compression tests were conducted for confirming the reproducible trend and the scattering of the stress and strain is less than ±5% or around ±0.5% engineering strain, respectively. Though the performance of Fe contained Mg-based BMGC is not as strong as the porous Mo contained Mg-based BMGC [13], with the plastic strain

| Table 1 |
| Pre-added vol.% and estimated vol.% of Fe particles in the Mg-based BMG composites. |
|----------------|----------------|
| Pre-added vol.% in the melt | Estimated vol.% in the composite |
| Mg-based BMG + 5Fe | 5 | 6.6 |
| Mg-based BMG + 10Fe | 10 | 9.7 |
| Mg-based BMG + 15Fe | 15 | 13.5 |
| Mg-based BMG + 20Fe | 20 | 18.5 |
of 10% and the yield strength of 0.95 GPa, but it is similar to the 8 vol.% solid Nb-reinforced Mg-based BMGC [9], with similar plastic strain and yield strength (~0.8 GPa) due to employing the low-yield ductile metal (i.e., Fe and Nb). However, for considering the cost of materials, the cheaper Fe particle (compare to Nb and Mo particle) is still having the potential to apply on strengthening the Mg-based BMGs.

It is known that the monolithic Mg-based BMG matrix should have a high strength (~0.85 GPa) if premature failure before reaching its elastic limit is avoided [2]. This confirms that the combination of the Fe particles and Mg-based amorphous matrix maintains the high strength from the amorphous matrix, and also brings very good plasticity from the ductile particles. Therefore, the SEM observations on the fracture surface were performed to evaluate the function of the “ductile Fe particles” on the improvement of mechanical performance. The observations on the specimen surface near the fracture area (Fig. 6) indicate that the deformation follows a shear-band mechanism. The fractured specimen with a fracture surface oriented about 43° to the loading axis is shown in Fig. 6(a). In Fig. 6(a), the multiple-shear bands can be observed along the fracture surface and around the Fe particles, indicating that there exists a strong reaction between ductile Fe particle and shear banding from the amorphous matrix.

For composite materials, there are two possible ways to toughen a brittle matrix: (i) by crack bridging from adding a soft second phase with a strong interface, or (ii) by crack deflecting from adding a strong hard phase with a weak interface. Compared with the Mg-based BMGs, Fe particles can be regarded as hard phase because of its higher modulus (211 GPa). It directly shows that the bifurcation of shear bands around the Fe particles has been observed in the present study, as shown in the circle area of Fig. 6(b). On the other hand, Fe particles still can be sheared-off plastically by the shear bands and absorbed large energy of shear bands (as shown in Fig. 7), and so as to enhance plasticity. Nevertheless, a slightly decreasing trend with increasing the addition of Fe particles was suggested due to the formation of brittle intermetallic phase at the interface between the Fe particle and amorphous matrix. A thin layer crystalline phase with a thickness of about 3 nm was found to attach on the surface of Fe particle by TEM examination, as illustrated in Fig. 8. Though this crystalline phase is too thin to be fully identified by the nano-beam diffraction, it can still be presumed by the phase diagram and EDS analysis, as shown in Fig. 8. There is no Fe constituent that was resolved at the amorphous matrix, but a relatively high content of Fe with about 3–1 atomic ratio of Fe:Gd was found at the thin layer crystalline phase. According to the binary phase diagram of Fe and Gd, an eutectic reaction of Fe2Gd/Gd occurs at the composition of 70 at.% Gd and 845°C [22]. Consequently, this layer crystalline phase can be suggested corresponding to the Fe2Gd intermetallic phase.

In Fig. 6, SEM observations show that the deformation occurs mainly through a shear-banding mechanism in the amorphous matrix. It is known that new shear bands are initiated at the sites
of local stress concentration \cite{23}. In this respect, due to the large elastic modulus difference between the particles (\(E = 211\) GPa) and the amorphous matrix (\(E = 40\) GPa), the stress concentration in the particle–matrix interface will induce the initiation of shear bands. Therefore, the Fe particles act in a way analogous to the ductile reinforcements in amorphous alloys described in Refs. \cite{9,24}. On the fracture surface, a vein pattern around the Fe particles is observed in Fig. 9. It means that a substantial increase in temperature in these localized shear bands is achieved. DSC analysis indicates that the melting temperature, \(T_m\), of the amorphous matrix is about 690 K; therefore, the shear banding-induced increases in temperature can adiabatically heat the regions of Fe particles to a high level. With the increase of temperature, two tendencies for the regions of Fe parti-
cles are expected: one is to dilate and another is to soften. However, the dilation process will be restricted by the surrounding Mg-based amorphous matrix, thus a tensile stress will be raised in the matrix surrounding the Fe particles. Applying the Mohr–Coulomb criterion \cite{25}, which is often used to describe stress states in BMGs, the shear bands tend to propagate in such tensile stress fields, especially since
these fields run on average 45° to the loading axis. In this way, these stress fields will cause a shear band, initiated by the concentration in the particle–matrix interface, to be guided in the direction of the neighboring Fe particles. In addition, these ductile Fe particles can be plastically deformed to arrest the propagating of shear bands by reducing the stress at their tips, as shown in Fig. 6(b). The above process makes the deformation distribute more uniformly across the specimens and difficult for a propagating shear band through whole sample. As a result, the mechanical instability is retarded and the ductility improved.

4. Conclusion

In summary, the Mg-based BMGCs with ductile Fe particles developed in the study may constitute a very promising material for structural applications due to their high compressive plastic strains comparable to that of crystalline alloys and high yield strength comparable to metallic glassy matrix. The Fe particles can branch primary shear band into multiple-shear bands and decrease the stress concentration for further propagation of shear band, and so as to enhance plasticity. Nevertheless, a slightly decreasing trend in yield strength with increasing the addition of Fe particles was suggested due to the formation of brittle Fe2Gd intermetallic phase at the interface between the Fe particle and amorphous matrix. In overall, the ductile Fe dispersoids would provide a good toughening effect in promoting the practical use of amorphous materials.

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