Unusual Plasticity of the Particulate-Reinforced Cu-Zr-Based Bulk Metallic Glass Composites

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We report a particulate-reinforced bulk metallic glass composite with high strength and unusual plasticity, which consists of Ta particles homogeneously distributed in Cu_{48}Zr_{48}Al_{8}Ag_{8} glassy matrix. The glassy matrix remains amorphous even after adding up to 15 vol.% of Ta particles. A largest plastic strain of up to 31% was obtained for the 10% Ta-containing composite. Ta particles seed the initiation of multiple shear bands and block the shear band propagation, leading to a net-like homogeneous distribution of the shear bands.

Keywords: bulk metallic glass, particulate reinforced composites, compression test

1. Introduction

Bulk metallic glasses (BMGs) have been developed in a number of alloy systems for the last two decades. They exhibit excellent mechanical properties, such as high strength and high corrosion resistance et al., and then have been considered as future structural materials. However, the plastic deformation of monolithic BMGs is always concentrated into the localized shear bands. As a result, BMGs typically display limited plastic flow in compression (0%–2%). This behavior has seriously limited the applications of BMGs as an engineering material. Since the second phase can act as an obstacle for the propagation of the shear bands and prevent a single shear band from propagating through the entire sample, BMG composites have been synthesized to improve the ductility. The methods for making composites with a glassy matrix and the second reinforced phases include precipitating a dendrite bcc phase from the melt and adding the foreign particles to the melt prior to casting. The in situ dendrite composites have been extensively studied by several research groups. As compared with in situ dendrite composites, the composites containing the foreign particles possess some advantages as the simple synthesis process, adding more kinds of reinforced phases and easy control of the homogeneous distribution of the reinforced phases. Choi-yim et al. firstly synthesized the particulate reinforced Zr_{57}Nb_{3}Al_{10}Cu_{15.4}Ni_{12.6} BMG composites containing the foreign particles, such as W, Ta, WC, SiC etc. However, the plastic strain of Zr_{57}Nb_{3}Al_{10}Cu_{15.4}Ni_{12.6} BMG composites was only about 2%–7%, far less than that of the in situ dendrite composites. Consequently, much effort has been devoted to develop the BMG composites containing foreign particles with high strength and high ductility.

Recently, we developed a new bulk glass-forming alloy with a composition of Cu_{48}Zr_{48}Al_{8}Ag_{8}. The as-cast Cu_{48}Zr_{48}Al_{8}Ag_{8} glassy rod with a diameter of 25 mm was successfully synthesized by copper mold casting. Due to high glass-forming ability, it is possible to prepare the Cu_{48}Zr_{48}Al_{8}Ag_{8} BMG composites. In this paper, we report a composite material consisting of the Cu_{48}Zr_{48}Al_{8}Ag_{8} BMG matrix reinforced by the foreign-added Ta particles. The Cu_{48}Zr_{48}Al_{8}Ag_{8} BMG composites show not only high yield strength, but also an unusual plastic deformation (plastic strain up to 31%) before failure and apparent strain hardening in uniaxial compression.

2. Experimental Procedure

A multicomponent alloy ingot with nominal composition of Cu_{48}Zr_{48}Al_{8}Ag_{8} was prepared by arc melting mixtures of Cu, Zr, Al and Ag with a purity of 99.99%, 99.5%, 99.99% and 99.99%, respectively, in a high purity argon atmosphere. The composite ingot with a composition of (Cu_{0.36}Zr_{0.48}Ag_{0.08}Al_{0.8}Ta_{x}); x = 5, 10, 15 and 20 in atomic percent, which correspond with 5.04, 10.08, 15.11 and 20.14 in volume fraction) was prepared by induction melting the Cu_{48}Zr_{48}Al_{8}Ag_{8} alloy together with Ta powder in a quartz tube under a high purity argon atmosphere. The average size of the Ta particles is about 40 μm. The composite ingots were then remelted in a quartz tube using an induction heating coil and injected through a nozzle into a copper mold with an inner cavity of 2 mm in an argon atmosphere. The structure of the as-cast samples was examined by X-ray diffraction (XRD) and scanning electron microscopy (SEM). Transmission electron microscopy (TEM) investigations were performed using a JEOL 2010 microscope. Room-temperature compressive tests were carried out with an Instron testing machine and the strain rate was 5 × 10^{-4} s^{-1}. The strain gage was used to measure the elastic modulus. The test specimen had a cylindrical form of 2 mm in diameter and 4 mm in height. Fracture surface was examined by SEM.

3. Results and Discussion

Figure 1 shows the XRD patterns of the 2-mm as-cast specimens containing 5–20 at.% Ta, together with the result of pure Ta particles. For the specimens with Ta contents from 5 to 15%, the diffraction patterns show sharp peaks from Ta particles superimposed on a broad scattering feature characteristic of an amorphous phase. No other phases are detected within the sensitivity limits of XRD. This indicates that Cu_{48}Zr_{48}Ag_{8}Al_{8} matrix consists of an amorphous phase. The specimen with 20% Ta shows the diffraction peaks corresponding to the precipitation of a crystalline phase, which can
be indexed as Cu$_{10}$Zr$_{7}$ phase. This result implies that the high content of Ta particles degrade the glass-forming ability of the matrix phase.

The as-cast microstructure of the 10% Ta composite is shown in Fig. 2. The microstructure consists of homogeneously dispersed particles (light phases) embedded in the metallic glass matrix (grey phase). The average chemical composition of the glassy matrix (determined by energy dispersive X-ray analysis) is Cu$_{36.00}$Zr$_{48.74}$Al$_{7.74}$Ag$_{7.98}$Ta$_{1.52}$, which is very close to the Cu$_{36}$Zr$_{48}$Al$_{8}$Ag$_{8}$ alloy. To investigate more closely the interface between the Ta particle and the glassy matrix, TEM work was performed. Figure 3 shows a TEM micrograph of the interfacial region between a Ta phase and the Cu$_{36}$Zr$_{48}$Al$_{8}$Ag$_{8}$ glassy matrix for the 10% Ta-containing composite, together with the selected-area electron diffraction patterns of the interface and glassy matrix.

Figure 4 shows the compressive stress-strain curves of the particulate-reinforced Cu$_{36}$Zr$_{48}$Al$_{8}$Ag$_{8}$ BMG composites containing 0–20% Ta. The monolithic glassy alloy exhibits yield strength of 1885 MPa, and fails immediately after yielding. In contrast, the composites reinforced with 5–15% Ta exhibit apparent work hardening and significantly plastic strain exceeding 20%. For the 10% Ta-containing composite, the yield strength, fracture strength and plastic strain are 1717 MPa, 2600 MPa and 31%, respectively. This is the largest plastic strain for the particulate-reinforced BMG composites. However, the composite containing 20% Ta exhibits a low yield strength of 1520 MPa without any plasticity. In addition, it is seen that the yield strength and young’s modulus decrease with increasing Ta content (as shown in the inset of the Fig. 4).

Figure 5 shows the fracture morphology of the 10% Ta BMG composite subjected to compressive fracture. The compressive fracture takes place along the maximum shear plane. A large number of shear bands are observed over the whole surface of the fracture samples. It is worth noting that most of the shear bands are intersected each other on the surface of the sample. Moreover, the magnified image shows that a number of shear bands have accumulated around the Ta particles (Fig. 5(c)).

Since the second phase added to the glass-forming melt can act as catalytic sites for heterogeneous crystal nucleation and growth, the high glass-forming ability of the melt should be guaranteed to prevent the formation of a continuous crystalline phase layer on the surface of the particles, which weakens the interfacial strength and seriously damages the mechanical properties of the composites.$^{7}$ In our current works, a clean interface is formed between Ta particles and Cu$_{36}$Zr$_{48}$Al$_{8}$Ag$_{8}$ glassy matrix (as shown in Fig. 3), indicating that the Cu$_{36}$Zr$_{48}$Al$_{8}$Ag$_{8}$ alloy has high glass-forming ability enough to resist the growth of a crystalline phase from the Ta/glass interface. Several years ago, Choi-yim et al. firstly synthesized the similar particulate reinforced...
Zr$_{57}$Nb$_3$Al$_{10}$Cu$_{15.4}$Ni$_{12.6}$ bulk glass composites. However, they reported that the composites containing Ta or W particles exhibited a plastic strain of only 2–7%, far less than the plastic strain of 31% for the present Cu$_{36}$Zr$_{48}$Al$_8$Ag$_8$ BMG composite. The significant difference may be due to the formation of a continuous nanocrystalline phase layer on the reinforced particles for the Zr$_{57}$Nb$_3$Al$_{10}$Cu$_{15.4}$Ni$_{12.6}$ BMG composites.

The unusual mechanical properties of the Cu$_{36}$Zr$_{48}$Al$_8$Ag$_8$ BMG composite appear to result from its unique microstructure. As shown in Fig. 2, Ta particles distribute homogeneously in the glassy matrix, which plays an important role in the initiation and propagation of the multiple shear bands. The pure Ta has a large plastic strain of about 40% and a Young’s modulus of 185 MPa at room temperature. Since the Young’s modulus of the glassy matrix is lower than that of Ta particle, high stress concentration occurs at the interface between Ta particles and glassy matrix under compressive loading. As a result, the shear bands initiate at the interface prior to the formation of shear bands on the maximum shear surface of the samples, leading to low yield strength and elastic modulus of the composite as compared with the monolithic BMG (Fig. 4). As the load increases, much more shear bands initiate and accumulate around the particles, resulting in the evident work hardening behavior of the composites. Moreover, the Ta particles can act as an obstacle for the propagation of the shear bands and prevent a single shear band from propagating through the entire sample. It can be imaged that Ta particles, the shear bands accumulated around the particles and the shear bands propagated between the particles might constitute a three-dimensional network structure in the glassy matrix. As the Ta concentration increases, the network structure should be much denser. As a result, much more shear bands initiate, gather or be confined in the network structure. This is may be the main reason for the unusual plasticity of the Cu$_{36}$Zr$_{48}$Al$_8$Ag$_8$ BMG composite containing 10% Ta. However, much larger amount of Ta particles degrade the glass-forming ability of the melt. It was found that a continuous crystalline phase layer was formed at the interface between the Ta particles and glassy matrix for the 15% Ta-containing composite (not shown here), which weakened the bonding between the particles and matrix and then decreased the mechanical properties. For the 20% Ta composite, the crystalline phases precipitated in the glassy matrix, and the matrix has lost the intrinsic properties of the glassy structure. As a result, the 20% Ta-containing composite exhibits significantly reduced mechanical strength without any plasticity.

4. Conclusions

We have successfully synthesized the particulate-reinforced bulk metallic glass composite consisting of Ta particles homogenously distributed in Cu$_{36}$Zr$_{48}$Al$_8$Ag$_8$ glassy matrix. Due to the high glass-forming ability of the Cu$_{36}$Zr$_{48}$Al$_8$Ag$_8$ alloy, the glassy matrix remains amorphous even after adding up to 15 at.% of Ta particles. The optimum concentration of Ta particle for the composite with the highest plasticity is about 10%. The yield strength, fracture strength and plastic strain are 1717 MPa, 2600 MPa and 31%, respectively, for the 10% Ta-containing composite. A large number of shear bands are observed on the whole surface of the fractured samples. Ta particles seed the initiation of multiple shear bands and block the shear band propagation, leading to a net-like homogeneous distribution of the shear bands.

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REFERENCES