Optimized interface and mechanical properties of W fiber/Zr-based bulk metallic glass composites by minor Nb addition

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Abstract

W fiber/Zr-based bulk metallic glass composites were prepared by melt infiltration casting and the interface reaction in composites was studied in detail. It was found that minor Nb addition to the matrix can suppress the interface peritectic reaction and optimize the interface structure in W fiber/Zr-based bulk metallic glass composites. Taking the interface characteristics of composites and glass forming ability of the matrix into account, an optimized alloy Zr47Ti13Cu11Ni10Be16Nb3 was selected as a new metallic glass matrix. The interface in the W fiber/Zr47Ti13Cu11Ni10Be16Nb3 bulk metallic glass composite has excellent interface bonding, and the compressive strength of 70% W fiber/Zr47Ti13Cu11Ni10Be16Nb3 metallic glass composite is 2.6 GPa, 58% higher than the unreinforced matrix. Multiple shear band formation in the matrix, which results from the interaction between W fiber and the matrix, can answer for the high ultimate fraction strength and excellent plastic deformation of the composite.

Keywords: A. Composite; B. Glasses, metallic; B. Mechanical properties at ambient temperature

1. Introduction

Despite their high strength and high elastic strain limit, most monolithic metallic glasses tend to form localized shear band and fail catastrophically upon yielding in tension or compression test [1,2]. Leng’s [3–5] pioneer work motivates the interest of metallic glass matrix composites. They combined metallic glass ribbons with ductile metals’ matrix in layered composites, finding that the constraint imposed by the ductile metal results in the formation of multiple shear bands in the metallic glass ribbons, which promotes the stable propagation of crack and makes the composite exhibit tensile strain. Recently, various bulk metallic glass matrix composites reinforced with fibers and particles have been fabricated with the aim to control the generation and propagation of shear band and improve the mechanical properties of bulk metallic glasses [6–9]. Continuous fibers reinforced metallic glass matrix composites prepared by melt infiltration casting are successful efforts [10,11].

During casting, diffusion and reaction at the interface are inevitably, excessive diffusion and reaction will weaken reinforcements and cause the loss of strength [12]. For the metallic glass matrix composites, these diffusion and reaction are more noticeable because they will alter the composition of the metallic glass matrix and reduce its glass forming ability [13]. In this paper, the interface of tungsten fiber reinforced Zr-based metallic glass matrix composites by melt infiltration casting have been optimized by minor Nb addition, in addition, mechanical properties of W fiber/Zr47Ti13Cu11Ni10Be16Nb3 composite are discussed in detail.

2. Experimental procedure

Zr-based matrix alloys were prepared by arc-melting pure metals with nominal purities of 99.99 wt% in a Ti-gettered argon atmosphere. Tungsten fiber with a diameter of 254 μm
was straightened and cut into 60 mm lengths, then cleaned in an ultrasonic bath of acetone, followed by ethanol. The details of the melt infiltration casting can be found elsewhere [14].

The composite samples were sliced normal to the fiber orientation. The interface structure between tungsten fiber and the matrix was examined by using X-ray diffraction, SEM, and EPMA. The cohesion of the interface is analyzed by using nano-indentation mechanical properties micro-probe (MPM). In this experiment, the depth of indentation is a constant of 150 nm and the distance between two indents is 1.8 μm. Under above indentation conditions, tungsten fiber and interface are in plastic deformation. The rods of 4 mm diameter with an aspect ratio of 1.75 were used in compression tests. The compression tests were performed on an Instron 4200, at a strain rate of 7.5 × 10⁻⁴ mm/s. After the mechanical tests, SEM was used for investigating the fracture surface.

3. Results and discussions

3.1. Formation of the interface reaction in Zr₅₅Al₁₀Ni₅Cu₃₀ matrix composite

Fig. 1 is the SEM backscattering image of the interface in Zr₅₅Al₁₀Ni₅Cu₃₀ matrix composite by infiltration at 1123 K for 10 min, there occurs a reaction layer, at the same time, dendrite phase precipitates in the matrix which is near to W fibers because the cooling rate of water quenching is too slow to reach the critical cooling rate of Zr₅₅Al₁₀Ni₅Cu₃₀ for glass formation. The components of the interface reactant and dendrite phase near W fibers are shown in Tables 1 and 2, the main elements in interface reactant and dendrite phase are W and Zr and Cu and Zr, respectively. Moreover, W to Zr atomic ratio is close to 1.86 and Cu to Zr atomic ratio is 2. Fig. 2 is X-ray diffraction patterns of Zr₅₅Al₁₀Ni₅Cu₃₀ matrix composite, there appear the diffraction peaks of W₂Zr₃ phase and CuZr₂ phase but the diffraction peaks of W fiber. Comparing X-ray diffraction patterns with the components analysis above, we consider the interface reaction production is W₂Zr₃ and the dendrite in the matrix is CuZr₂.

About the interface reaction in the melt infiltration casting, there is a viewpoint of dissolution—precipitation mechanism, the solid reinforcement dissolves into the melt matrix and forms a supersaturation layer near it in the casting, then the interface reactant precipitates there during cooling subsequently. As follows from the W–Zr phase diagram [15], the maximum solubility of W in melt Zr is about 8 at.%. But, Domagała et al. have found that W–Zr alloy with more than 5.3 at.% of W can precipitate lamellar W₂Zr₃ phase quenching form from 1460 °C. Table 3 is the average components of the matrix near W fibers and it only contains 1.68 at.% of W, so the matrix cannot precipitate lamellar W₂Zr₃ phase during cooling from infiltration temperature. In the W–Zr phase diagram, there occurs a peritectic reaction:

\[
\text{Zr}_1 + \text{W} \rightarrow \text{W}_2\text{Zr}_3
\]

taking the morphology of the interface reactant into account, we consider the interface reaction in Zr₅₅Al₁₀Ni₅Cu₃₀ matrix composite a peritectic reaction between Zr in the melt matrix and solid W fibers during infiltrating.

Fig. 3 is the X-ray patterns of Zr₅₅Al₁₀Ni₅Cu₃₀ matrix composite infiltrated at different temperatures for different time, With increasing infiltration temperature and infiltration time, the diffraction peaks of W₂Zr₃ dies down, but the peaks of W₂Zr₃ occurs and are intensified gradually. In the W–Zr phase diagram, there is only a stable peritectic reaction phase W₂Zr₃, this illustrates that W₂Zr₃ phase should be a metastable phase during casting, and it can be transformed into the stable W₂Zr₃ phase with increasing infiltration temperature and infiltration time. Fig. 4 shows variation of the interface reaction with infiltration temperature and infiltration time, the interface peritectic reaction becomes more intensive with increasing infiltration temperature and infiltration time. The interface reaction product breaks off W fiber after peritectic reaction, in this way, the new fiber surface is exposed to the liquid melt.

<table>
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Table 1
Components of the interface reactant

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Table 2
Components of the dendrite phase near W fibers

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**Fig. 1.** SEM backscattering image of the interface in Zr₅₅Al₁₀Ni₅Cu₃₀ matrix composite.
and the interface peritectic reaction can proceed, which will weaken the mechanical properties of W fiber severely, so this peritectic reaction must be controlled effectively.

3.2. Influence of Nb on the interface reaction

Fig. 5 shows SEM backscattering image of the interface in metallic glass matrix composite with Nb. No interfacial reaction is observed at the interface between W fibers and matrix. As Zr$_{55}$Al$_{10}$Ni$_{5}$Cu$_{30}$ matrix composite, some crystalline phases precipitate in the matrix of (Zr$_{55}$Al$_{10}$Ni$_{5}$Cu$_{30}$)$_{0.98}$Nb$_2$ and Zr$_{57}$Al$_{10}$Ni$_{12.6}$Cu$_{15.4}$Nb$_5$ matrix composites. Nevertheless Zr$_{47}$Ti$_{13}$Cu$_{11}$Ni$_{10}$Be$_{16}$Nb$_3$ matrix is completely amorphous. The diffraction peaks of the composite correspond to the pattern of W fiber perfectly (Fig. 6).

In the metallic matrix composites system, it is basically valid that interface reaction can be reduced by alloying the matrix [16]. This method is also effective for metallic glass matrix composites, in our work, a peritectic reaction takes place between Zr$_{55}$Al$_{10}$Ni$_{5}$Cu$_{30}$ matrix and W fibers, but it doesn’t occur for the other three matrix with Nb, it is clear that this peritectic reaction is restrained by adding Nb to the matrix alloys. The diffusion of Zr at the interface and the peritectic reaction rate are required to understand and control the interface peritectic reaction between solid state (W) and liquid state (Zr in the melt matrix alloy). Our previous works have illustrated that the alloy element Nb with a strong affinity for W prefers to segregate at the interface during casting, which can reduce the segregation and diffusion of Zr there (Table 4), this is the main reason why Nb can depress the interface peritectic reaction in the Zr$_{55}$Al$_{10}$Ni$_{5}$Cu$_{30}$ matrix composite [17]. Taking the interface and glass forming ability into account, Zr$_{47}$Ti$_{13}$Cu$_{11}$Ni$_{10}$Be$_{16}$Nb$_3$ alloy was selected as the metallic glass matrix, subsequently, the properties of W fiber/Zr$_{47}$Ti$_{13}$Cu$_{11}$Ni$_{10}$Be$_{16}$Nb$_3$ bulk metallic glass composite will be elaborated on.

3.3. Mechanical properties of Zr$_{47}$Ti$_{13}$Cu$_{11}$Ni$_{10}$Be$_{16}$Nb$_3$ matrix composite

3.3.1. Mechanical properties of the interface

Fig. 7(a) shows the distribution of the indents at the interface measured by using MPM. The magnification of the indent in the matrix is shown at lower left corner of Fig. 7(a). Circular pattern occurs around the indent. This pattern is characterized by using section analysis of atom force microscope (AFM). The surface uplift of the pile-up is shown in Fig. 7(b) and (c). It is found that the circular pattern around the indent is not composed of cracks but overlapping bands in pile-up of displaced materials. Metallic glasses tend to deform in adiabatic localized shear bands, in compression, there is large plastic deformation at the tip of the indent, at the same time, these plastically displaced materials are easy to flow because of higher temperature, so metallic glass is typical example consisted with this phenomenon [18].

Fig. 8 shows the hardness and elastic modulus of different position in Fig. 7(a). It illustrates that both of the hardness and elastic modulus at the interface are higher than that in the matrix. In order to further investigate the interface strength, another extreme indention test was conducted. It is performed by putting the maximal load of Nanoindenter MPM (750 mN) on the matrix near the interface to form cracks. The propagation of the cracks is shown in Fig. 9. It is seen that cracks occur at the points of the indenter besides the overlapping bands in pile-up around the indent, one crack (marked as A) propagates through the interface and continues in W

Table 3

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<tr>
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Fig. 4. Variation of the interface reaction with infiltration temperature and time. (a) 1223 K, 10 min, (b) 1273 K, 10 min, (c) 1273 K, 25 min, (d) 1273 K, 4 min, (e) 1323 K, 25 min.

Fig. 5. SEM backscattering image of the interface in metallic glass matrix composite with Nb. (a) (Zr55Al10Ni5Cu30)0.98Nb2, (b) Zr57Al10Ni12.6Cu15.4Nb5, (c) Zr47Ti13Cu11Ni10Be16Nb3.
fiber, and no expansion of the cracks along the interface is observed, these features show the strong interface bond in W fiber/Zr47Ti13Cu11Ni10Be16Nb3 composite.

3.3.2. Mechanical properties of the composite

Fig. 10 shows compressive stress—strain curves for unreinforced Zr47Ti13Cu11Ni10Be16Nb3 bulk metallic glass and 70% tungsten fiber reinforced composite. The ultimate strength and plastic strain before failure of the composite increase significantly to 2.6 GPa and 13.5%, respectively, and the composite exhibits perfectly-elastic stress—strain behavior, but the compressive strength of matrix alloy is only about 1.6 GPa.

Fig. 11 shows SEM image of the lateral surface of 70%W fiber/Zr47Ti13Cu11Ni10Be16Nb3 composite after compressed to failure. It is observed that the rod sample shows drum appearance (Fig. 11 (a)), and multiple shear bands are formed on the surface of composite, as shown in Fig. 11 (b). Two groups of shear bands with an orientation of ±45° to the axial load are seen in the matrix between tungsten fibers, all the shear bands initiate and terminate near tungsten fibers. The shear bands with two orientations often have the same set, and branch into finer ones surrounding fibers, it can be induced that there are shear band—tungsten fiber interactions in the composite, and fibers act as the constraint to shear band propagation. Stress concentrations build up at the front of propagating shear bands, when the shear band meets fibers, this stress concentrations triggers the formation of another shear band, typically at right angles to the first. New shear band initiates and multiple shear band deformation occurs in the composite. These effects of fibers on shear band initiation and propagation can answer for the high ultimate fracture strength and excellent plastic deformation.

4. Conclusions

(1) An interface peritectic reaction takes place between Zr in the liquid matrix alloys and W fibers at the interface in Zr55Al10Ni5Cu30 matrix composites, the addition of Nb restrains this harmful interface peritectic reaction
Taking the interface characterization of composites and glass forming ability of matrix into account, an optimized alloy Zr47Ti13Cu11Ni10Be16Nb3 was selected as a new metallic glass matrix.

(2) W fiber/Zr47Ti13Cu11Ni10Be16Nb3 composite has excellent interface characteristics and the interface cohesion is very fine.

(3) The compressive strength of 70% W fiber/Zr47Ti13Cu11Ni10Be16Nb3 composite is 2.6 GPa, 58% higher than the unreinforced matrix, at the same time, it exhibits 13% plastic elongation when tested in quasistatic compression. Tungsten fibers interact on the behavior of shear bands in the matrix and multiple shear bands initiate, which results in the increasing of the compressive strength and plastic deformation.

Acknowledgement

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References