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Significant TRIP-effect improvement by manipulating ZrCu-B2 distribution in ZrCuAlCo-based bulk metallic glass composites via inoculating Ta particles

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ABSTRACT

The ZrCu-B2 phase can effectively increase the plasticity for some specified Zr based metallic glasses composite (BMGC) by transformation induced plasticity (TRIP) mechanism. However, large and non-uniformly distributed ZrCu-B2 phases usually precipitate in the Zr-based BMGC samples by conventional copper mold casting. Therefore, the concept of inoculation in conventional solidification process was applied to modify the size and distribution of ZrCu-B2 phase in this study. Ta particles (size of 5−30 μm) with 0−4.0 vol% were added into the Zr48Cu47·5Al4Co0.5 BMG matrix as the inoculant. Ta particles can act as nucleation seeds for precipitating a homogeneously distributed ZrCu-B2 phase in the matrix. Moreover, the ZrCu-B2 precipitate size can be further controlled by different solidification cooling rates. It is clearly to see that the ZrCu-B2 phase embedded in the amorphous matrix for the Zr48Cu47·5Al4Co0.5 cast rods added with 0−0.75 vol% Ta particle cast by a copper mold at −30 °C (243 K, or a cooling rate of 650 K/s). In addition, the ZrCu-B2 phase exhibits a round shape and relatively homogeneous distribution. The optimum processes BMGC can exhibit significantly improved mechanical properties (1890 MPa fracture stress and 14% plastic strain) in comparison with the base BMGC (1560 MPa fracture strength and 7.5% plastic strain).

1. Introduction

Bulk metallic glasses (BMGs) possess attractive characteristics because of their superior properties compared to crystalline counterparts [1,2], such as their high fracture strength, large elastic strain limit (2%), and superior corrosion resistance [3–7]. However, the room-temperature brittleness limits their applications as structural materials. Once a single shear band is initiated in many monolithic BMGs, it will rapidly propagate and then result in catastrophic fracture by the localization of strain-softening nature [8–12]. To improve the low macroscopic plastic deformation at room temperature, BMG composites (BMGCs) with in-situ and/or ex-situ second phases, such as dendrites, particles or fibers [13–20], have been developed to prevent the shear bands from propagation. Recently, it is reported that BMGCs can enhance toughness and work-hardening properties when the second reinforcement consists of a shape memory nature [21–26] and performs a strengthening mechanism with the effect of transformation-induced plasticity (TRIP) [27]. The TRIP effect has been successfully applied to make the $Zr_{48}Cu_{47.5}Al_4Co_{0.5}$ BMGC with a reinforcement of ZrCu phase [28]. However, the fabrication of $Zr_{48}Cu_{47.5}Al_4Co_{0.5}$ BMGC containing with ZrCu phase requires a high cooling rate in the casting process.

Since the ZrCu phase tends to decompose into Zr_7Cu_{10} and Zr_2Cu phases at low temperatures, and it is impossible to precipitate a pure ZrCu-B2 phase from the fully glassy matrix by annealing for the ZrCuAlCo alloy system [29]. In addition, the high cooling rate of fabrication process also creates some issues, e.g., the morphology and volume fraction of the ZrCu phase is strongly dependent on the

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cooling rate. According to the results reported by Wu et al. [24], the sample made by a lower cooling rate (or the sample with a larger diameter) has a larger amount of ZrCu precipitate phase but exhibits irregular shapes and uneven distribution. On contrary, the sample made by higher cooling rate (or sample with a smaller diameter) has a lower amount of ZrCu phase and presents a round shape and uniform distribution.

In order to obtain a composite structure with promising ZrCu phase embedded in the glass matrix (namely, suitable amount, small round-shape particle size, and uniform distribution), the inoculation concept of conventional casting is applied in this study in the rapid solidification process. The function of inoculant is to provide a multitude of heterogeneous nucleation site for precipitating a large amount of small precipitates with a lower energy barrier of nucleation. Nevertheless, it is necessary to consider two aspects. The first is the good coherent of lattice constant between the inoculant and ZrCu phase, and ZrCu phase can easily nucleate on the inoculant surface. The second is the inoculant must be a refractory material and can resist high temperature melting during the arc re-melting process. Accordingly, Ta is selected in this research as the inoculant material because Ta is a refractory metal (with a melting point of 3290 K) and has the very low solubility in the Zr-based BMGs [30]. In parallel, the crystalline structure and lattice constants of Ta and ZrCu phase are quite similar, Ta (BCC, a = 3.302 Å, JCPDs #895158) and ZrCu (ordered body centered cubic BCC or B2), a = 3.256 Å, JCPDs #491483. In addition, the ductile Ta particles are often used to improve the plasticity of Zr-based BMG by in-situ and/or ex-situ dispersion [31–35]. However, this study only focuses on that Ta-inoculant induced ZrCu phase precipitation to get the better morphology. Moreover, the dependence of the microstructure and mechanical properties as a function of cast cooling rate, sample size and fabrication process in the $Zr_{48}Cu_{47.5}Al_4Co_{0.5}$ +Ta BMGC system are all investigated.

2. Experimental procedures

The master-alloy ingots with a nominal composition of $Zr_{48}Cu_{47.5}Al_4Co_{0.5}$, and added with different volume fractions (from 0, 0.25, 0.5, 0.75, 1, 2, 3, to 4 vol%) of Ta particles (5–30 μm in size) were prepared by arc melting. In order to make the composition homogeneous, a two-step melting process was carried out. In-situ composite: raw metals of Zr and Ta, which have the highest melting temperatures in this alloy system were melted together to form a homogeneous solid solution ingot. This binary ingot was remelted with the remaining metals to obtain the target alloy composition in an arc furnace under argon atmosphere. Ex-situ composite: raw metals of Zr, Cu, Al and Co were melted together to form an ingot, which can quite reduce the melting point in this alloy system. This ingot was re-melted with the Ta metals to obtain the target alloy. Due to the melting point of ingot (~1373 K) is different from Ta elements (3290 K). It is difficult to dissolution with each other. At last, the liquid melt of each alloy was suction cast into a Cu mold to form BMGC rods with diameters from 2 to 4 mm.

The thermal properties of the as-cast BMG and BMGC specimens were characterized by the differential scanning calorimeter (Netsch STA449F3, HT-DSC) at a heating rate of 0.33 K/s. The morphology, volume fraction and distribution of the ZrCu phase in the amorphous matrix were examined by the optical microscope (OM, Olympus TH4-100). The evolving phases and microstructures were characterized by X-ray diffraction analysis (Bruker D8A X-ray diffractometer, XRD) with monochromatic Cu-Kα radiation and transmission electron microscopy (Jeol JEM 2100 TEM operated at 200 kV). The hardness of alloy rod was measured by A Vickers microhardness tester (Mitutoyo HM-221) was applied to measure the hardness of the BMGC samples under a load of 0.3 kg. The compressive testing was conducted using a mechanical testing system (MTS) at room temperature and strain rate of $1 \times 10^4\ s^{-1}$. The specimen dimension for the compression test is 2 mm in diameter and 4 mm in height and with parallel two ends, The fracture surface and specimen surface of the specimen for the compression test were examined by scanning electron microscope (FEI Inspect F50, SEM) with energy dispersive spectrometry (EDS).

3. Results and discussion

3.1. Estimate the initial cooling rate in Cu mold

The characteristic of ZrCu-B2 phase has a strong dependence on cooling rate. It is thus very important to clarify the cooling quantity in the solidification process. The infrared thermometer was used to monitor the temperature of liquid melt (T_{melt}), and the cooling temperature between the Cu mold and sample was measured by a thermocouple in the cavity of Cu mold. Meanwhile, the temperature of Cu mold was maintained at a preset temperature by a cooling system to ensure the consistent experiment parameter in each casting. The schematic drawing of casting setup is shown in Fig. 1. Fig. 2 shows the measured time-temperature (t-T) curve during the injection casting. According to the transient heat flow function (Lumped capacitance method - Newtonian cooling), the cooling rate can be expressed by Ref. [36],

$$\frac{dT}{dt} = -\frac{h}{L_c \rho C_p}(T - T_\infty) \quad (1)$$

where dT/dt is the cooling rate (in K/s), h is the heat transfer coefficient (W/m^2 K), L_c is the ratio V/A_s (m), V is the volume (m^3), A_s is the surface area (m^2), ρ is the density (kg/m^3), C_p is the specific heat capacity (J/kgK), T is the casting temperature (K), and T_∞ is the Cu mold temperature (K). It follows that the heat absorption ΔH (in J/kg) can be expressed by

$$\Delta H_s = \frac{T - T_\infty}{\sqrt{\pi}} \frac{C_{p,s}}{\phi e^{\phi^2}\left[\mathrm{erf}(\phi) + \left(\frac{\kappa_s C_{p,s} \rho_s}{\kappa_m C_{p,m} \rho_m}\right)^{1/2}\right]}, \quad (2)$$

where κ is the thermal conductivity (W/mK), ψ is $L_c/[2(t\,\alpha_s)^{1/2}]$, α is the thermal diffusivity (m^2/s), and t is time (s). Meanwhile, ΔH_s, κ_s,

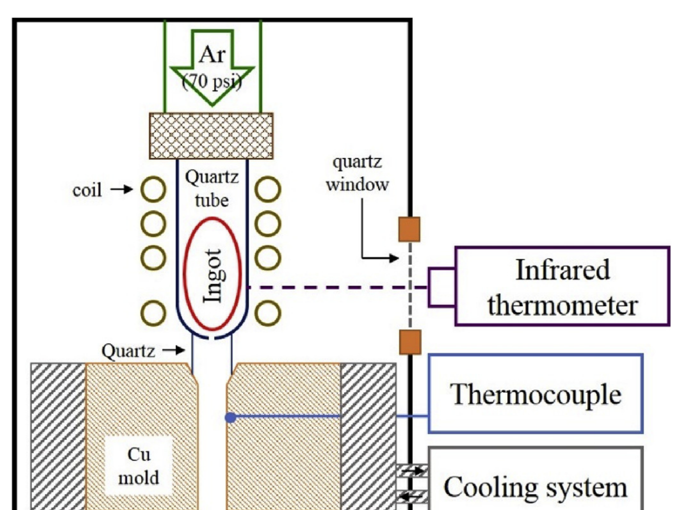

Fig. 1. Schematic drawing of the experimental setup in the injection casting.