

# Designing ductile CuZr-based metallic glass matrix composites

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## ABSTRACT

The CuZr-based metallic glass matrix composites (MGMCs) with different volume fractions of crystalline phases were designed by doping of nickel. Minor addition of nickel element can change the glass-forming ability of the resultant composites. Large compressive plasticity accompanied by strong work-hardening capacity was achieved in the  $\text{Cu}_{47}\text{Zr}_{48}\text{Al}_4\text{Ni}_1$  composite with a volume fraction of crystalline phases of 33%. The excellent compressive properties were mainly attributed to the inhibition for the propagation of shear bands by the ductile crystals and deformation induced martensitic transformation of the B2-CuZr phase. However, no obvious global tensile ductility was obtained, due to the mode I fracture toughness and small plastic-zone size of glass matrix. To uncover the shear-band evolution during deformation, finite element simulation was conducted, revealing that the ductile B2 phase can tune the shear-stress distribution and consequently initiate and retard shear bands, which stimulates the multiplication of shear bands. Accordingly, the spacing of B2 CuZr particles is a vital factor dominating the plasticity of CuZr-based MGMCs, especially upon tension.

## 1. Introduction

Bulk metallic glasses (BMGs) have attracted large attention because of their unique properties, such as high strength, large elastic limit, excellent corrosion and wear resistances, etc [1]. However, they are known for being extremely brittle, failing in a catastrophic fracture at room temperature, due to formation of localized shear bands [2,3]. To enhance macroscopic plasticity, metallic glass matrix composites (MGMCs) have been developed by in situ or ex situ introduction of secondary phases, which can hinder the rapid propagation of shear bands [4]. In recent years, a series of in-situ dendrite-reinforced MGMCs have been developed, such as Ti-based and Zr-based MGMCs [5–7]. Nevertheless, these MGMCs exhibit a macroscopic strain softening with localized necking due to lack of work hardening in the glass matrix at room temperature, especially under tensile conditions, limiting the applications significantly.

Recently, these shortcomings have been successfully addressed in some CuZr-based MGMCs reinforced by ductile B2 CuZr phases [8–15]. It is worth noting that the B2-type CuZr phases are prone to deform plastically compared to other intermetallic compounds with

complex crystalline structure [16–18], resisting fast propagation of highly localized shear bands effectively and promoting the generation of multiple shear bands in the glass matrices. Moreover, as crystalline phases with a shape memory effect, B2-type CuZr phases can undergo a stress-induced martensitic transformation from B2 phase to a monoclinic B19' phase, which imparts an appreciable work-hardening capability and then compensates the softening of the matrix significantly during plastic flows [9,12–14,19]. In this way, the effect of “transformation-induced plasticity” (TRIP) [11,14,20] can be introduced into the ductile MGMCs. Depending on the TRIP effect, the  $\text{Zr}_{48}\text{Cu}_{47.5}\text{Al}_4\text{Co}_{0.5}$  composite, containing 25% volume fractioned spherical B2-CuZr phases, presents strong work-hardening capability and considerable tensile ductility [14]. Subsequently, Wu et al. [21] have demonstrated that a minor 0.5 at% addition of Co in the  $\text{Zr}_{48}\text{Cu}_{47.5}\text{Al}_4\text{Co}_{0.5}$  composites can promote deformation twinning and martensite transformation of B2-CuZr phases by reducing stacking fault energy (SFE). Meanwhile, the SFE of B2 phases is predicted to be strongly reduced by the addition of Ni theoretically. In addition, since the glass-forming ability (GFA) of these BMGs and MGMCs is sensitive to minor additions [9,22,23], an effective adjustment of microstruc-

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tures can be obtained via appropriate trace additions. It is noted that microstructures plays a most effective role in improving the mechanical properties of such alloys [9,13,24]. As such, an improvement of mechanical properties is expected by a minor addition of Ni. However, the effect of Ni addition on the microstructures and mechanical properties of such kinds of MGMCs have been rarely investigated.

In this study, a series of CuZr-based MGMCs with different volume fractions of crystalline phases are systematically designed via minor addition of Ni. Compressive and tensile experiments are carried out to investigate the deformation and fracture mechanisms. Moreover, the correlation between the evolution of shear bands and plastic flow can be revealed vividly, combined with finite element simulation (FES). Additionally, dependences of the mechanical properties on microstructures are explored as well.

## 2. Experimental

Master alloys with normal compositions of  $\text{Zr}_{48}\text{Cu}_{48-x}\text{Al}_4\text{Ni}_x$  ( $x=0, 0.5, 0.8, 1, 1.5$ , and  $5$  at%) were prepared by arc melting the mixture of constituent elements with purity greater than 99.9% (wt%) under a Ti-gettered argon atmosphere. The ingots were melted four times to ensure compositional homogeneity. Plate-shape samples with 1.5 mm in thickness and 10 mm in width were prepared by the copper-mould-casting method. The tensile specimens with gauge dimensions of 10 mm (length)  $\times$  1.5 mm (width)  $\times$  1.5 mm (thickness) and compressive specimens with gauge dimensions of 3 mm (length)  $\times$  1.5 mm (width)  $\times$  1.5 mm (thickness) were prepared by the electric spark method, respectively. Both compressive and tensile tests were conducted at a strain rate of  $5 \times 10^{-4} \text{ s}^{-1}$  at room temperature. Optical microscope (OM) was used to examine microstructures of as-cast samples. The volume fraction of B2-CuZr phase was determined by an Image-Pro Plus 6.0 software based on the OM images. The phases of the plates were identified by X-ray diffraction (XRD), and the thermal properties were analyzed by differential scanning calorimetry (DSC) at a rate of 20 K/min. Scanning electron microscopy (SEM) was employed to observe the microstructures and fractographs. Nanoindentation was performed on Nano-Indenter II tester (MTS Systems, USA) at room temperature to measure the hardness of the glass matrix and B2 crystalline phases of the as-cast and deformed samples. A Berkovich diamond tip was used. The nominal strain rate was  $0.002 \text{ s}^{-1}$  and the maximum indentation depth was 1000 nm. The cross-section of the samples for the nanoindentation tests was carefully polished.

## 3. Results

### 3.1. Microstructural evolution with Ni additions

Fig. 1 shows XRD spectra of as-cast plates of the alloys with different Ni additions. For the ternary  $\text{Cu}_{48}\text{Zr}_{48}\text{Al}_4$  alloy, Bragg diffraction peaks corresponding to the body-centered cubic B2 CuZr phases are superimposed on a broad hump. With addition of 0.5–1% Ni, the peak intensity of the crystalline phase increases gradually, indicating an increasing amount of the B2 phase. Specially, this scenario is obvious appreciably for the 1% Ni-added sample, revealing that 1% Ni addition greatly deteriorates the GFA of  $\text{Cu}_{48-x}\text{Zr}_{48}\text{Al}_4\text{Ni}_x$  alloys. However, the volume fractions of B2 CuZr phases decrease indicated by the declined relative intensity of crystalline peaks for the 1.5% Ni-added sample. With further Ni additions (5%), the crystalline peaks become indistinctive visibly, suggesting that excessive Ni additions could enhance the GFA of  $\text{Cu}_{48-x}\text{Zr}_{48}\text{Al}_4\text{Ni}_1$  alloys.

Fig. 2 presents the DSC curves of these MGMCs. The features of the DSC curves are all similar, namely, an endothermic platform caused by the glass transition and an exothermic heat peak caused for crystallization. The heat release from the crystallization process of these composites decreases gradually with the Ni added from 0% to 1%. This

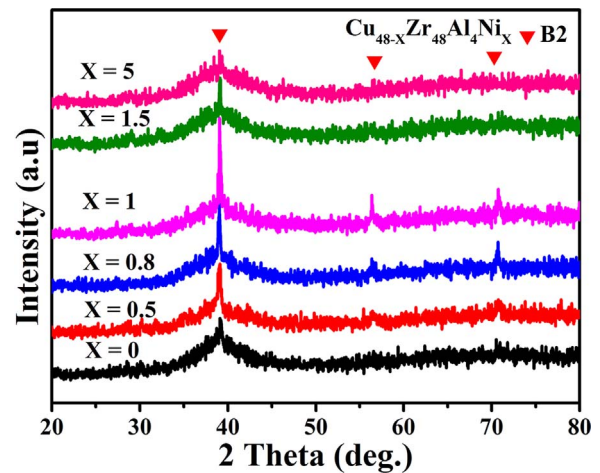


Fig. 1. XRD spectra of the as-cast  $\text{Cu}_{48-x}\text{Zr}_{48}\text{Al}_4\text{Ni}_x$  alloys with different Ni additions.

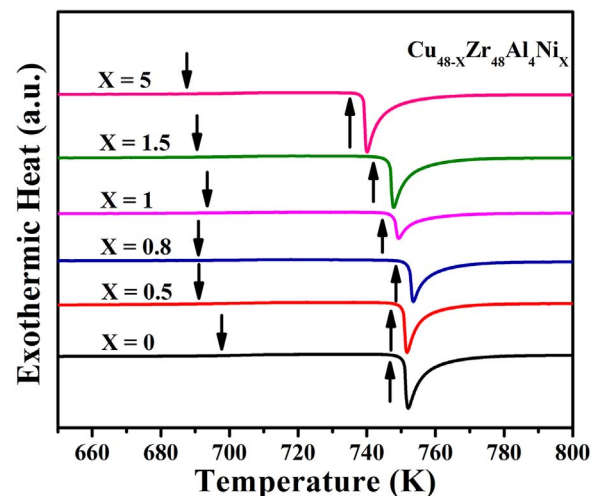


Fig. 2. DSC curves of the as-cast  $\text{Cu}_{48-x}\text{Zr}_{48}\text{Al}_4\text{Ni}_x$  alloys with different Ni additions.

indicates that the volume fraction of the crystalline phase increases. In contrast, with additions of 1.5% and 5% Ni, the exothermic peaks increase, which means that the volume fraction of the amorphous phase increases. The DSC results are consistent with XRD results.

Fig. 3 shows the optical cross-section metallographs of as-cast  $\text{Cu}_{48-x}\text{Zr}_{48}\text{Al}_4\text{Ni}_x$  ( $x=0, 0.5, 1, 1.5$ , and  $5$ ) alloys. For the Ni-free sample, only limited numbers of B2 CuZr particles can be observed, and their sizes are less than  $50 \mu\text{m}$ , as shown in Fig. 3(a). With a 0.5% Ni addition, in addition to some large B2 particles (Fig. 3b), a number of small B2 particles with a diameter of about  $5\text{--}10 \mu\text{m}$  can be found, as presents in the inset of Fig. 4(b). The bimodal sized phenomenon of B2 particles has been reported in Ti-based MGMCs as well [19]. For the 1% Ni-added sample, the crystalline volume fraction further increases, as shown in Fig. 4(c). And a large number of B2 CuZr particles can be found throughout the whole section in Fig. 4(f). With further increase of Ni additions, the volume fraction of B2 crystalline phases decreases, specially, for 5% Ni added sample, only a small amount of B2 phase can be found at the out surface of samples.

Uniaxial compression tests were carried on the as-cast  $\text{Cu}_{48-x}\text{Zr}_{48}\text{Al}_4\text{Ni}_x$  ( $x=0, 0.5, 0.8, 1$ , and  $1.5$ ) samples at room temperature, and the corresponding engineering stress-strain curves for these samples are displayed in Fig. 4. The compressive plasticity of the current composites is enhanced with increasing the B2 CuZr crystalline phase. The ternary  $\text{Cu}_{48}\text{Zr}_{48}\text{Al}_4$  alloy without Ni addition exhibits a distinct yielding at  $\sim 1840 \text{ MPa}$ , and has a fracture with the plasticity of  $\sim 0.5\%$ . With minor additions of 0.5% and 0.8% Ni, the resultant

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