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Effect of stress states on the deformation behavior of Cu-based bulk metallic glass in the supercooled liquid region

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ABSTRACT

The effect of stress states on the deformation behavior of the $Cu_{54}Zr_{22}Ti_{18}Ni_6$ bulk metallic glass (BMG) alloy was studied in the supercooled liquid region. At 723 K, Newtonian plastic flow governed the deformation during the compression test, whereas strain-hardening occurred during the tensile test. At 733 K, a fast failure was observed during tensile test. The diffusion rate of Cu atoms in the BMG alloy plays an important role in the deformation behavior. The fast diffusion of Cu atoms under the tensile stress state caused faster crystallization leading to a fast strain-hardening during the tensile plastic deformation.

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1. Introduction

Bulk metallic glass (BMG) alloys can exhibit characteristic superplastic deformation in the supercooled liquid region (SLR), which is defined by the temperature range between the glass transition temperature (T_g) and crystallization temperature (T_x) [1–4]. Since the atomic configuration of BMG alloys in the SLR varies with time and strain states, microstructural changes including crystallization processes take place in BMG alloys upon annealing or deformation in the SLR [5–9].

During deformation at temperatures in the SLR, crystallization processes in BMG alloys can be varied by the plastic stress states. It has been repeatedly reported that tensile stress accelerated crystallization in Cu [10,11] and Zr [12–14] based BMG alloys. It turned out that BMG alloys was still amorphous in the non-deformed region whereas nano-crystals were observed in the sample gauge, which was attributed to the stress-induced crystallization. In contrast, compressive stress hardly affected the rate of crystallization processes in Zr based BMG alloys [15,16], indicating that the controlling parameter of crystallization was the total annealing time rather than the applied stress. However, detailed studies dealing with the mechanism on the effect of both tensile and compressive stress on plastic deformation behaviors of a Cu based BMG alloy in the SLR have seldom been reported yet.

In this work, the stress–strain curves of the Cu-based BMG alloy in the SLR were determined by uniaxial tensile and compressive tests. Both tests were carried out at the same strain rate and temperature. The present BMG alloy displayed different plastic stress–strain curves under tensile and compressive plastic strain states. Thermal and microstructural analyses were employed to interpret the differences in the plastic flow behavior.

2. Experimental procedure

Bulk metallic glass rods with a diameter and height of 13 and 15 mm were first produced by spark plasma sintering of the atomized metallic glass powder with a composition of $Cu_{54}Ni_6Zr_{22}Ti_{18}$ (at.%) [17,18]. The amorphous nature of the present BMG alloy was confirmed by means of X-ray diffraction and differential scanning calorimetry (DSC) [18–20]. Samples for tensile and compression tests were prepared from BMG rods using an electro-discharge machine. A gauge length of 8 mm and diameter of 8 mm were used for both the tensile and compression test samples.

Uniaxial tensile and compression tests in the SLR were carried out at a constant strain rate of $2 \times 10^{-3}\ s^{-1}$ in a Gleeble tester (System 3500). Samples were heated up to the testing temperature with a heating rate of 40 K/min and loading was applied after 60 s of holding time. After deformation or annealing, all samples were cooled with a constant cooling rate of 200 K/min.

The thermal properties were determined by a DSC (Perkin Elmer DSC7) at a constant heating rate of 40 K/min. Microstructural observations were examined by a high resolution transmission electron microscope (HRTEM, FEI Tecnai F20). Thin foils for the HRTEM were prepared by mechanical grinding and polishing followed by low energy ion milling with LN_2 cold stage. The variations of the viscosity of the BMG alloy in the SLR was determined using a thermal mechanical analyzer (TMA) and Gleeble tester (System 3500) at a constant heating rate of 40 K/min under compressive mode.

3. Results

The SLR of the present Cu based BMG alloy ranges from T_g = 712 K to T_x = 765 K [17]. Tensile and compression tests were carried out at two different temperatures of 723 and 733 K. Fig. 1

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shows the stress–strain curves of the BMG samples determined at a constant strain rate of 2×10^{-3} s^{-1}. Hereafter, the samples deformed by tension and compression are referred to as the TEN ('tensioned') and COM ('compressed') samples, respectively. At 723 K, the flow stress of the COM sample first reached 250 MPa at $\varepsilon = -0.4$ and then remained nearly unchanged until $\varepsilon = -1.39$, which is the typical Newtonian flow commonly observed during the compression of BMG alloys in the SLR [1,21–27]. Homogeneous deformation by Newtonian flow is typically observed at a low strain rate and, under these conditions, homogeneous flow occurs when the creation and annihilation of the free volume are in balance. In contrast, the flow stress of the TEN sample varied significantly with increasing plastic strain. The flow stress increased to 255 MPa at $\varepsilon = +0.12$ and then decreased slightly $\varepsilon = +0.5$. Above the strain, the TEN sample displayed strain hardening with increasing strain. This strain hardening is attributed to the gradual crystallization of amorphous phase, which is commonly observed in a number of Zr- and Cu-based BMG alloys during tensile deformation in the SLR [10–14,26]. At 733 K, inhomogeneous deformation was observed in all samples. The flow stress of the COM sample first reached 30 MPa at $\varepsilon = -0.05$ and then remained to $\varepsilon = -0.2$, which stress level is lower than that of the COM sample tested at 723 K. A low flow stress of the COM sample tested at 733 K is attributed to the decrease in the viscosity with increasing deformation temperature [1,4,21]. Above $\varepsilon = -0.2$, the COM sample observed strain hardening with increasing strain and fractured at $\varepsilon = -0.75$, which was not observed at 723 K. In contrast, the flow stress of the TEN sample increased to the peak stress of 485 MPa at $\varepsilon = +0.25$ and then fractured after necking. This fractured strain is lower than that of the COM sample, which gives rise to the rapid crystallization during the tensile test at 733 K.

Since the atomic configuration of the BMG alloy varies continuously in the SLR, the annealing time plays an important role in microstructural changes during deformation. In the present work, the total exposure time depended on the amount of strain imposed during deformation. Exposure times for holding and deformation to the imposed strain are listed in Table 1. The present BMG alloy displayed different plastic stress–strain curves during compression and tensile tests, as shown in Fig. 1. This result demonstrates the stress state dependence of the plastic deformation behaviors of the BMG alloy.

In order to study the difference in plastic flow behaviors under tensile and compressive stress, the microstructural evolution of the COM and TEN samples was examined using DSC and HRTEM. The samples annealed without deformation were also examined.

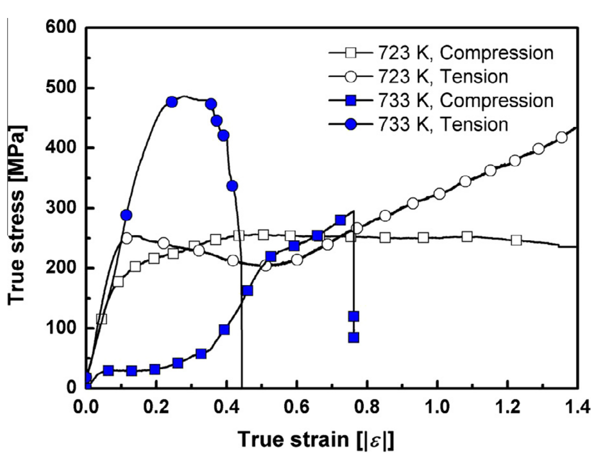

Fig. 1. Stress–strain curves of the BMG sample determined by the compression and tensile tests at a constant strain rate of 2×10^{-3} s^{-1}.

Table 1
Exposure times for holding and deformation to the imposed strain.

Imposed strain	±0.43	±0.69	±1.39
Holding time (s)	60	60	60
Deformation time (s)	215	345	695
Total imposed time (s)	275	405	755

Fig. 2 shows the DSC curves of the (a) annealed, (b) TEN and (c) COM samples exposed for 0, 275, 405 and 755 s at 723 K. The DSC curve of the initial sample displayed two distinct exothermic peaks. Our previous work [19,20] reported that the original amorphous phase first decomposed into Cu-rich and Cu-poor amorphous phases during annealing at 723 K. This compositional decomposition was related to the first exothermic reaction. The formation of various crystalline phases from the decomposed amorphous phases was found to be associated with the second exothermic reaction.

In the annealed sample (Fig. 2a), the integrated area of the first peak decreased gradually with increasing annealing time to 755 s. This suggests that the decomposition of the original BMG alloy into two amorphous phases took place progressively. In the TEN sample deformed to $\varepsilon = +1.39$ (total imposed time = 755 s), the first peak disappeared completely and the integrated area of the second peak decreased, as shown in Fig. 2b. However, in the COM sample deformed to $\varepsilon = -1.39$ (total imposed time = 755 s), the integrated area of the first peak was larger than that of the sample annealed for 755 s without deformation as listed in Table 2. This indicates that compressive stress retards the change in the first peak reaction, whereas tensile stress accelerates it.

HRTEM observations were performed to further examine the structural changes in this BMG alloy during annealing and deformation. Fig. 3 shows HRTEM images and corresponding diffraction patterns of the (a) initial, (b) annealed, (c) TEN and (d) COM samples exposed for 755 s at 723 K. The initial sample showed a fully amorphous structure without lattice fringes (Fig. 3a). In the annealed sample (Fig. 3b), the dark contrast with a size ranging from 5 to 10 nm and two diffraction rings in the corresponding diffraction pattern developed, which were not found in the initial amorphous microstructure. A previous study [19,20] clearly showed that Cu atoms are segregated in the dark contrasts. The diffusion of Cu atoms leads to the decomposition of an original amorphous phase into two amorphous phases, one with Cu-rich and the other with Cu-poor. A typical amorphous microstructure without lattice fringes was observed in the COM sample until it deformed to $\varepsilon = -1.39$, as shown in Fig. 3d. The TEN sample deformed to $\varepsilon = +1.39$ displayed a microstructure with some areas containing nano-crystals of hexagonal $Cu_{51}Zr_{14}$ with a lattice parameter of $a = 1.123$ nm and $c = 0.827$ nm (space group, $P6/m$) as shown in Fig. 3c. Spots from crystalline phases in the diffraction pattern also show the existence of crystalline phases. This result further supports the fact that compressive stress indeed retards the compositional decomposition, whereas tensile stress accelerates the compositional decomposition and the subsequent formation of crystal phases.

The $Cu_{54}Ni_6Zr_{22}Ti_{18}$ BMG alloy at 723 K displayed Newtonian flow behavior until $\varepsilon = -1.39$ during the compression test, whereas it showed strain-hardening caused by rapid crystallization during the tensile test. At 733 K, The fractured strain of the TEN sample was lower than that of the COM sample. The reason for the different stress–strain curves under tensile and compressive stress in the present BMG alloy was discussed.

The present Cu-based BMG alloy is decomposed into Cu-rich and Cu-poor amorphous phases at the early stages of annealing in the SLR. Therefore, the diffusion of Cu atoms governs the