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Elastic moduli and mechanical properties of bulk metallic glasses after quasi-static compression

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ABSTRACT

The effect of quasi-static compressive stress on the elastic moduli and mechanical properties of a $Cu_{46}Zr_{46}Al_8$ bulk metallic glass (BMG) was investigated. When the applied quasi-static stress is below 2 GPa (equivalent to 1.4 times the yield strength of the BMG), the elastic moduli of the deformed BMGs are found to decrease with the applied stress, revealing the softening or dilatation of the bulk metallic glass. The Poisson ratio is relatively stable when the stress is below 1000 MPa, but it decreases significantly afterwards. Both the plasticity and strength of the BMG are found to increase at low applied stress, and achieve a maximum value before decreasing at higher applied stress. The applied stress is shown to enhance the mechanical properties of the BMG and the properties can be controlled by quasi-static compressive stress. The results demonstrate that an applied stress far below the macroscopic yield strength can still result in microscopic yielding and microstructure change in metallic glass systems.

1. Introduction

Bulk metallic glasses (BMGs) with disordered atomic structures have promising potential for various applications due to their unique mechanical properties, including ultrahigh strengths, lower Young's moduli, high elastic strain limits and a large capacity for accumulating elastic energy [1–5]. Metallic glasses generally exhibit two basic modes of deformation: homogeneous flow and inhomogeneous flow. Elastic deformation belongs to homogeneous flow, which leads to recoverable macroscopic structure adjustment [6–10]. At a low stress, the elastic deformation is reversible, instantaneous and fully linear. Structural changes are hard to detect in the deformation process at this stage. However, when a larger elastic stress is applied, the process becomes time-dependent, and in order to achieve equilibrium between the stress and strain in both the loading and unloading directions, a finite time is required. Recent research studies have reported that elastostatic compression below the yield strength σ_y can induce irreversible micro-structural disordering in BMGs, proven by thermodynamic analyses and molecular dynamics simulations [11,12].

Plastic deformation belongs to inhomogeneous flow, which leads to irreversible macroscopic structure changes. When the applied stress is above σ_y, the formation of localized shear bands will lead to very limited inhomogeneous strain to failure for most BMGs. In fact, the deformation of BMG systems is very complicated and different to conventional alloys. In some cases, even with an applied stress over σ_y, BMGs can still undergo a large homogeneous strain without fracture, providing the specimens have low aspect ratio [13]. Because the atomic microstructure is the main factor that influences the value of the elastic moduli, they indicate the best measures to understand the structural changes in metallic glass [14,15]. By observing the changes in elastic moduli, it is convenient to detect the structure evolution in metallic glasses. In the present work, compression tests were undertaken on a $Cu_{46}Zr_{46}Al_8$ BMG with a low aspect ratio of 1.4:1 under different quasi-static stresses (from 0 to 1.4 σ_y,). A pulse echo overlap method was used to obtain the elastic moduli. The changes of elastic moduli and mechanical properties of the BMG after compression were systematically investigated, and the structural evolution induced by quasi-static compression was discussed.

2. Experimental

The $Cu_{46}Zr_{46}Al_8$ glassy alloy was prepared by melting pure Cu, Zr and Al in an arc-melting furnace under a Ti-gettered argon atmosphere. After the alloy was remelted several times, it was cast by suction into a copper mold to obtain a 50-mm-long cylindrical rod, of diameter of 5 mm. Cylindrical specimens with a 1.4:1 aspect ratio were prepared and compressed in a materials testing system (MTS) under different constant quasi-static stresses (within the range of 0–2 GPa) for 2 h. After quasi-static compressive loading, the 5 mm cylindrical samples were cut into 2 mm × 2 mm × 4 mm cuboid shapes as shown in Fig. 1. The mechanical properties of the cuboid samples were characterized by the MTS machine at an initial strain rate of $5 \times 10^{-4}\ s^{-1}$ at room temperature. A pulse echo overlap method was used to measure the acoustic velocities of the BMG after quasi-static compressive loading

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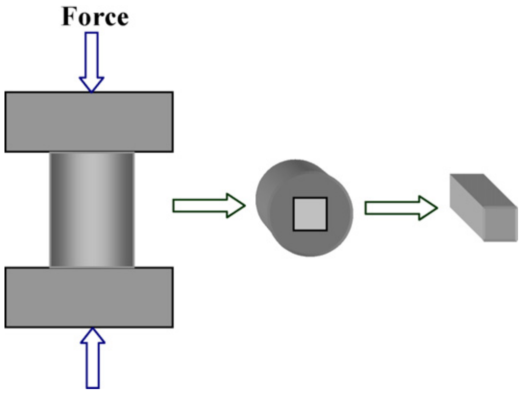

Fig. 1. The schematic plan of quasi-static compressive loading on the BMG samples. After quasi-static compressive loading, the cylindrical samples are cut into cuboid shape with a 2:1 aspect ratio.

at ambient conditions. A MATEC 6600 ultrasonic system with *x*- and *y*-cut quartz transducers was used to measure the travel time of ultrasonic waves through the BMG specimen, at a 10 MHz carry frequency. The measuring sensitivity was of the order of 0.5 ns. Based on the Archimedean technique, the density was measured with an accuracy within 0.1%. Based on the acoustic velocities and the densities, the elastic constants of the BMG including Young's modulus *E*, bulk modulus *B*, shear modulus *G*, and Poisson's ratio ν were determined. The structure of the BMG was characterized by a differential scanning calorimeter (DSC), the Perkin Elmer DSC-7.

3. Results and discussion

Fig. 2 shows the dependences of the ultrasonic longitudinal (V_l) and transverse (V_s) velocities on the applied quasi-static stress. Both V_l and V_s decrease with increasing applied stress. When the applied stress is increased to 2000 MPa, the decrease of V_l (3.1%) is more than that of V_s (2.0%). Based on the measured acoustic data and the densities, the elastic moduli (*E*, *G*, and *B*) were determined and their variations as a function of the applied stress are shown in Fig. 3(a)–(c), respectively. Under an applied quasi-stress of 2000 MPa for 2 h, *E*, *G*, and *B* of the BMG have a reduction of 4.1%, 3.9%, and 6.9%, respectively. The bulk modulus *B* is a measure of resistance to compressibility of a solid and can be defined by the equation: $B = -V(\partial P/\partial V)$, where *P* is pressure, *V* is volume. The reduction of *B* illustrates the softening of the BMG after quasi-static compression [16,17].

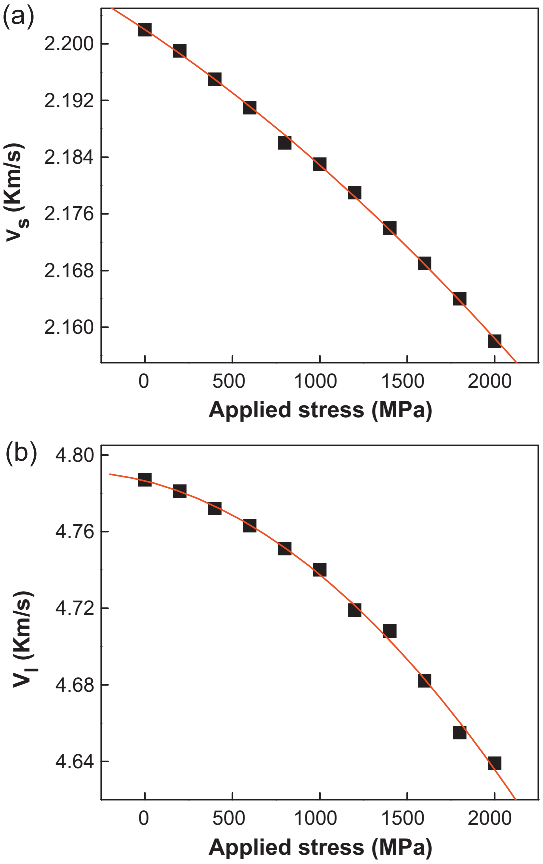

Fig. 2. The dependences of the ultrasonic longitudinal (V_l) and transverse (V_t) velocities on the quasi-static stress for the BMG.

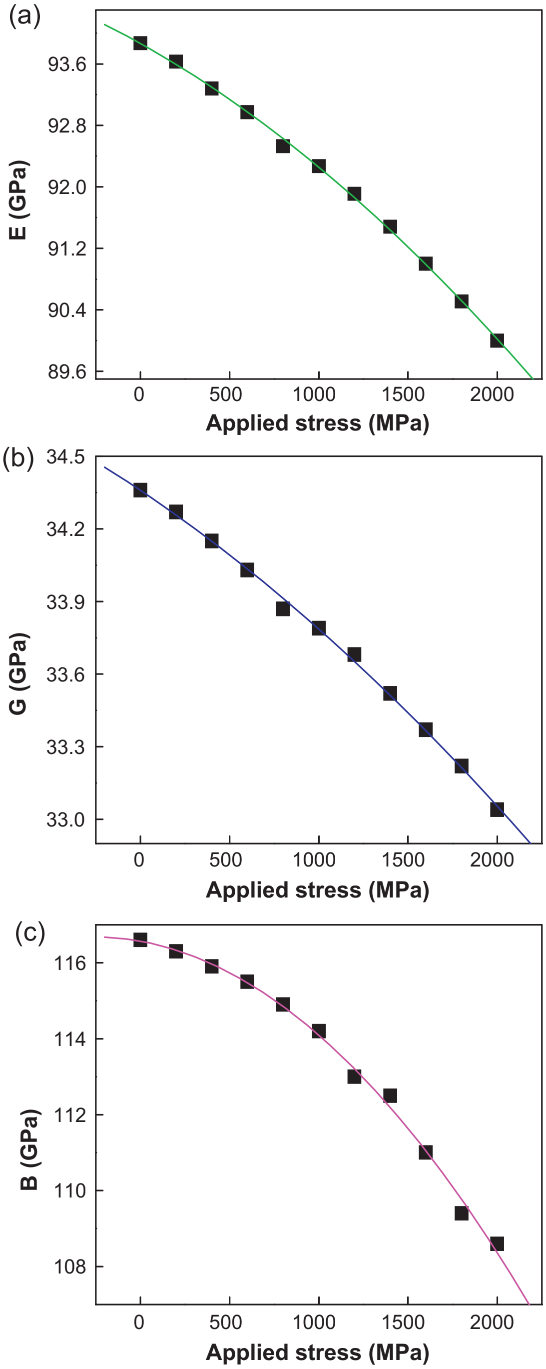

Fig. 3. The variations of Young's modulus *E* (a), shear modulus *G* (b), and bulk modulus *B* (c) of the BMG with quasi-static stress.

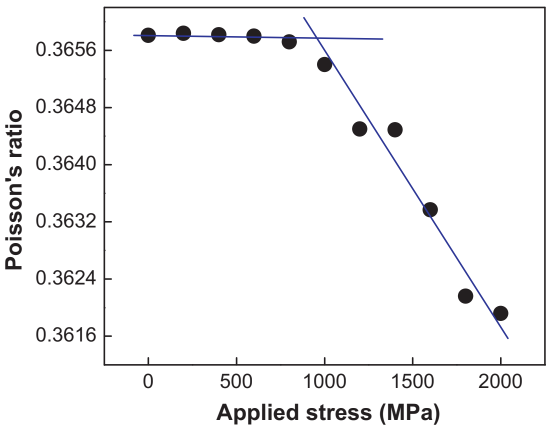

Fig. 4. The variation of Poisson's ratio on quasi-static stress for the BMG.

Fig. 4 shows the dependence of Poisson's ratio on the applied quasi-static stress for the BMG. Poisson's ratio ν is directly related to the bulk and shear modulus ratio and can be calculated as: $\nu = (1/2) - (3/((6B/G) + 2))$, which characterizes the relative value of the compressive and shear deformations and correlates with the atomic configuration in glassy materials. The structural changes of the alloy can be clearly reflected by Poisson's ratio. With an applied stress of less than 1000 MPa, Poisson's ratio of the BMG remains relatively stable, revealing that the structure changes are not obvious. However, when the applied pressure is between 1000 MPa and 2000 MPa, Poisson's ratio of the BMG decreases rapidly, which reflects dramatic structural changes. Homogeneous isotropic elastic materials have their elasticity uniquely determined by any two moduli, hence the dramatic changes of the bulk modulus and Poisson's ratio demonstrate that the microstructure of the BMG has changed significantly.

The mechanical behavior is particularly sensitive to the microstructure change of the glassy alloys. Through a uniaxial compression test, the macroscopic mechanical properties of the quasi-statically deformed BMG were characterized. Fig. 5 shows the compressive stress-strain curves for the specimens with different quasi-static stresses. Interestingly, the plasticity of the BMG changed significantly after quasi-static compression. With the increase of the applied stress, the plastic strain firstly increases and then decreases. Meanwhile, the yield strength of the BMG also has similar change trend. Fig. 6 shows the dependence of the compressive yield strength on the applied stress. The yield strength increases with increasing quasi-static stress and reaches its maximum (an increment of about 32%) around 800 MPa. It then decreases with further increase of the applied stress. This result shows that the compressive plasticity and strength of BMGs can be controlled and enhanced by quasi-static compression treatment.

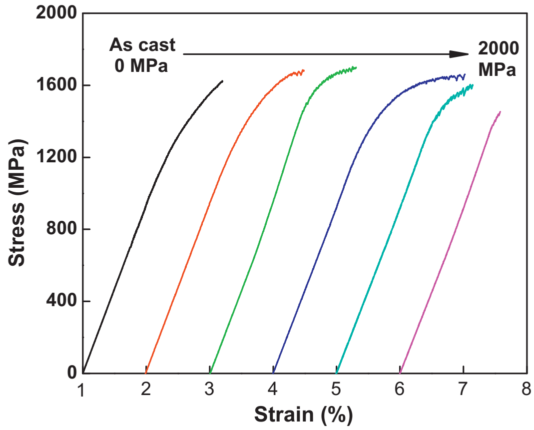

Fig. 5. The uniaxial compressive stress–strain curve for the specimens with different quasi-static stresses.

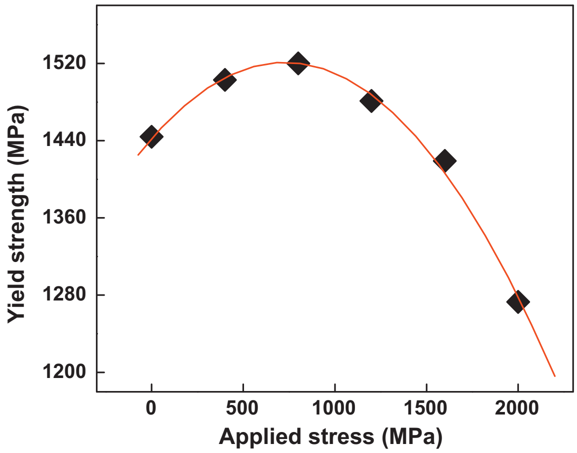

Fig. 6. The dependence of compressive yield strength on the applied quasi-static stress.

Previous studies have shown that the density of metallic glasses will be increased by hydrostatic compression [18]. However, in this work, the densities of the BMG in the as-cast and deformed states (after the applied stress of 2.0 GPa for 2 h) at room temperature were measured to be 7.952 and 7.873 g/cm^3, respectively. The decrease in density due to the applied stress is about 0.14%, which indicates dilatation induced by the deformation. As compared to the hydrostatic pressure process, the decrease in density under quasi-static compression may be attributed to the fact that the deformation is not being totally constrained. This implies that there is volume expansion in the compressive process, similar to natrolite [19]. Most materials diminish in volume when exposed to a uniform, externally applied pressure, and after removal of the applied pressure, the elastic strain is instantaneously recoverable, and the anelastic strain is recovered gradually. The increased volume after quasi-static compression reflects the unique structure of metallic glass materials [20]. There has been much research work to study the structural evolution of metallic glasses under elastic stress using molecular dynamics simulation. Park et al. have found that the Young modulus of Cu–Zr glasses is reduced after elastostatic loading and the fraction of full icosahedra in three binary metallic glasses is significantly reduced with increasing strain [12]. This disordering process is shown to be irreversible even if the applied stress is removed, and the elastostatic loading results in an increase in heat of relaxation and plasticity [12]. Zhang et al. have also reported that there is the destruction of the full icosahedra clusters into distorted ones, and permanent structural change can be introduced in $Cu_{50}Zr_{50}$ metallic glass by an applied tensile stress of 1000 MPa although it is still within the elastic regime and far below its macroscopic yield strength of about 1740 MPa [21]. Recently, the effect of elastostatic loading in a multicomponent BMG has also been experimentally investigated. Ke et al. have reported the increase in volume in a Zr-based multicomponent BMG after elastostatic compression [22].

Fig. 7 shows the DSC traces for the as-cast and quasi-static stress deformed BMGs. Through the analysis of DSC, with the increase of the applied stress, the glass transition temperature T_g has little change, but the crystallization temperature T_x shifts to a higher temperature. Moreover, as compared to the as-cast sample, the width of the supercooled liquid region ΔT_x increases from 77 K to 82 K with the application of the quasi-static stress of 2000 MPa. The quasi-static compression makes the supercooled liquid state of

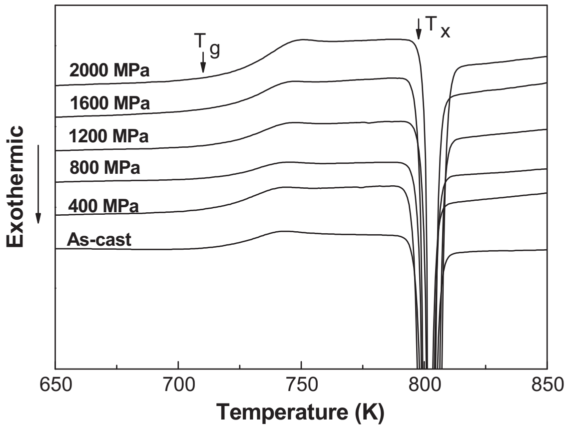

Fig. 7. The DSC traces for the BMG in as-cast and quasi-static stress states.

the alloy more stable. It is revealed by the DSC results that the pretreatment has induced irreversible structural relaxation, and there is reconstruction of the atomic configuration in the BMG. Chen et al. have reported that the thermal stability of a BMG can be improved by a flux treatment [23]. After the treatment, the supercooled liquid region is found to be enlarged, and the improvement is considered to be related to the decrease of heterogeneity after flux treatment. Under elastostatic loading, Park et al. and Zhang et al. have also observed the disordering process of metallic glasses by molecular dynamics simulation [12,21].

Yielding is regarded as a transition point between the elastic strain and plastic strain. It is also the transition point between homogeneous flow and inhomogeneous flows, or reversible and irreversible structure changes for metallic glasses. Previous research work reveals that structure change can take place in metallic glasses at a stress level at 0.8–0.9 of the macroscopic yield strength σ_y [12,22]. In other words, microscopic yielding can occur below the macroscopic yield strength. Since the Poisson's ratio has a sharp decrease at 0.68 σ_y (as shown in Fig. 4) and is very sensitive to structure adjustment, it suggests that the structure of the BMG has significantly changed starting from 0.68 σ_y. The value is lower than described in previous research [12,22], and further proves that microscopic yielding or microstructure change can occur far below the macroscopic yield strength.

It is well known that annealing can reduce the free volume in BMGs, which results in densification of the BMG [24,25]. The reduction of free volume makes the deformation of a BMG more difficult, and thus the macroscopic yield strength is enhanced by annealing treatment. With the increase of quasi-static stress, the yield strength in Fig. 6 shows a slight increase at the beginning and then a rapid decrease. It is interesting to note that the turning point of the curve corresponds to that observed in the Poisson's ratio curve. At low applied quasi-static stress, a slight increase of yield strength implies a small structure relaxation of the BMG. There may be only a reduction of the free volume, like the phenomenon observed in annealing. It is however, the increase in strength and plasticity with a relatively constant Poisson's ratio at low applied stress deviates from a prevailing view that the larger the ν, the more ductile the BMGs becomes [10]. The relation between mechanical behavior and the structure is rather complex especially at low stress. More work has to be done to fully understand the deformation behavior of BMGs. When the applied pressure is increased, microscopic yielding occurs at 0.68 σ_y, shear transformation zones (STZs) will be driven and more free volume will be generated [26,27], which means work softening of the BMG. However, when the applied pressure exceeds σ_y, the continuity and integrality of the BMG will be damaged, which explains the rapid decrease of yield strength under a compressive stress of 2000 MPa.

4. Conclusion

The change of elastic moduli and mechanical properties of a $Cu_{46}Zr_{46}Al_8$ metallic glass BMG after quasi-static compression with the applied stress below 1.4 times of the macroscopic yield strength (σ_y) was investigated. Based on the measured acoustic data, the elastic moduli of the $Cu_{46}Zr_{46}Al_8$ metallic glass BMG was found to decrease with the applied quasi-static stress. When the applied quasi-static stress was below 0.68 times σ_y of the $Cu_{46}Zr_{46}Al_8$ metallic glass, the Poisson ratio remains relatively stable, but it decreases significantly when the applied stress increases from 0.68 to 1.4 σ_y. It demonstrates that microscopic yielding or microstructure change can occur far below the macroscopic yield strength. The mechanical testing further reveals that the compressive plasticity and strength of the BMG after a quasi-static compression change with the applied stress. It becomes feasible to control and enhance the properties of BMGs by applying quasi-static stress. The mechanical properties and elastic moduli of a BMG after quasi-static compression are considered to be related to the changes in free volume, density and structure.

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