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Internal stresses induced by plastic shear deformation of Zr–(Cu,Ag)–Al bulk metallic glasses

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ABSTRACT

Effective internal shear stress σ_i induced by torsional deformation of $Zr_{46}(Cu_{4/5}Ag_{1/5})_{46}Al_8$ and $Zr_{46}Cu_{46}Al_8$ bulk metallic glasses different by the glass-forming ability of the maternal melts has been determined by measurements of stress relaxation upon stepwise unloading. It has been found that the ratio σ_i/σ_0 (σ_0 is the initially applied shear stress) decreases upon increasing the temperature from ≈ 0.8 at $T = 450$K ($T \approx 0.64 \times T_g$) to ≈ 0.08 at $T = 638$K ($T \approx 0.91 \times T_g$) independently of σ_0 and glass composition. The obtained result is in good agreement with earlier data obtained on ribbon metallic glasses. The origin of deformation-induced internal stresses and their connection with deformation mechanisms of metallic glasses has been briefly discussed.

1. Introduction

Plastic deformation of metallic glasses (MGs) continues to be a topical issue from both physical and application viewpoints. Over the past decades, quite a few theoretical models of MGs' plastic flow leading to different constitutive flow laws have been proposed. However, the microscopic flow mechanism remains to be a matter of controversial debates and its adequate theory is still lacking [1,2]. The verification of a flow model includes its comparison with the experiment at different testing modes and stresses. It is almost always assumed that the applied shear stress σ constitutes the driving force for plastic deformation and, therefore, it is this stress, which should be substituted into a constitutive flow law. Meanwhile, it is well known from the physics of crystalline materials that plastic deformation can lead to the formation of large internal shear stress σ_i, which is opposite in the sign to σ [3]. Then, the effective stress $\sigma^* = (\sigma - \sigma_i)$ actually represents the driving force for continuing plastic deformation. Besides that, the knowledge of internal shear stresses occurring during deformation provides information, which is very useful to verify different theoretical approaches. Therefore, the internal stresses induced by plastic deformation of MGs constitute an issue of major importance.

The current information on this issue is limited, to our knowledge, to a few studies [4–6] performed in the nineties of the past century on ribbon Ni–Nb and Co-based glasses produced by conventional single roller melt spinning (production quenching rate $R \sim 10^6$K/s). It was found by means of stress relaxation measurements that room-temperature tensile deformation leads to the formation of the internal stress σ_i, which reaches 93% to 96% of the initially applied stress. An increase of temperature T results in gradual decrease of σ_i, so that it tends to zero upon approaching T to T_g (T_g is the glass transition temperature). We are unaware of any other studies of deformation-induced internal stresses in metallic glasses, including bulk MGs being produced at much smaller $R \approx 10^2$K/s, and even less. In this paper, we present an investigation of internal stresses induced by pure shear of bulk $Zr_{46}Cu_{46}Al_8$ and $Zr_{46}(Cu_{4/5}Ag_{1/5})_{46}Al_8$ glasses. The choice of these MGs is determined by the fact that the corresponding maternal melts, while being only slightly different in the chemical composition, reveal large difference in their glass-forming ability (GFA). It was recently found for the same Zr–Cu–Ag–Al system that the glass produced from the melt with higher GFA always displays significantly bigger amount of structural relaxation upon annealing below T_g [7]. Thus, it is interesting to determine whether the deformation-induced internal stresses in MGs are dependent on the GFA of the maternal melts.

2. Experimental

Bulk $Zr_{46}Cu_{46}Al_8$ and $Zr_{46}(Cu_{4/5}Ag_{1/5})_{46}Al_8$ (at.%) were chosen for the investigation. In the latter glass, 20% of Cu atoms are substituted by bigger Ag atoms. However, the GFAs of the corresponding melts are significantly different: the maximal diameters of fully amorphous rods prepared by melt suction are 5 mm and more than 20 mm for the first and second alloys, respectively [8]. Thus, the GFA of the

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four-component alloy is significantly higher. In the present investigation, MGs were produced by melt jet quenching into a copper mold with a cavity of $2\times5\times60\ mm^3$ at a rate of $\approx$200K/s. The initial state was verified to be purely amorphous by X-ray diffraction in the transmission mode at $\lambda=0.5668$Å using a synchrotron radiation facility at the Kurchatov Institute in Moscow. Differential scanning calorimetry (Mettler-Toledo DSC1 operating at 5 K/min) determined T_g's (as the onset of endothermal heat flow) to be 700 K and 687 K for $Zr_{46}Cu_{46}Al_8$ and $Zr_{46}(Cu_{4/5}Ag_{1/5})_{46}Al_8$, respectively. The corresponding crystallization onset temperatures, T_c's, were equal 755 K and 746 K.

Like in previous experiments [4–6], internal stresses were determined by measurements of stress relaxation upon stepwise unloading [3]. The measurements were performed using a special laboratory-made torsion testing micromachine. The samples were prepared by cutting and grinding the glassy bars (described above) to the final cross-section of $(40-60)\times(40-60)\mu m^2$ as determined by an optical microscope. Gripping of specimens (gauge length of 1–2 mm) was performed mechanically. The applied torque was determined through the torsional deformation of a 100−μm quartz fiber, which was attached in series to the sample, and measured by a laser optical lever using a Hamamatsu S1352 position-sensitive diode. The relative error of the torque measurements was estimated to be $\approx$10%. Computer-controlled measurements were performed in a vacuum of about 0.01 Pa.

3. Results

Fig. 1 gives an example of the performed tests. After heating to $T=550$K, a $Zr_{46}(Cu_{4/5}Ag_{1/5})_{46}Al_8$ sample was loaded by a torque corresponding to an initial surface shear stress $\sigma_0=200$MPa (calculated assuming rectangular sample's cross-section), the total shear strain was fixed, and stress relaxation was measured during 3600 s. Next, the specimen was unloaded by $\Delta\sigma\approx10$MPa, and stress relaxation was again measured during 1000 s. The same procedure was then repeated 11 times. It is seen that the applied stress σ always decreases after first four unloadings. After 5th unloading, σ first increases and next starts to decrease, as shown in inset *c*. The applied stress σ corresponding to the condition $d\sigma/dt=0$ (marked by a vertical arrow) equals to the effective internal shear stress σ_i [3]. After 6th and further unloadings, σ always increases with time, so that the applied stress is less than the internal stress, i.e. $\sigma<\sigma_i$.

If the applied stress does not change with time, then $\sigma=\sigma_i=$ const$\neq f(t)$. The fact that σ first increases and next decreases after 5th unloading (inset *c*) indicates that σ_i is time dependent. This is certainly caused by structural relaxation occurring upon measurements. However, since σ after the first four unloadings always decreases with time but always increases after 6th and further unloadings, one has to conclude that the time change of σ_i is limited by the inequality $74\text{MPa}<\sigma_i<88$ MPa(see insets *b*, *c* and *d*). We performed a preliminary investigation of σ_i time dependence. For this purpose, the first relaxation run at $T=550$K was carried out during i) 2000 s and ii) 6000 s (compared with 3600 s in Fig.1). This allowed changing the amount of structural relaxation prior to unloading. It was found that, within the error, the σ_i-level remains unchanged. Thus, the σ_i time dependence is relatively weak and limited by the uncertainty indicated above.

Overall, stress relaxation experiments were performed at sixteen temperatures in the range $450\text{K}\leq T\leq638\text{K}$ on both glasses tested at σ_0 of 200 MPa, 500 MPa and 1000 MPa (the latter stress was applied only at $450\text{K}\leq T\leq475\text{K}$). Below 450 K, the experiments were impossible because the stress relaxation is smaller than the resolution of the employed technique. At $T\leq550$K, the first relaxation run was performed during 3600 s and followed by the unloading procedure described above (Fig. 1). At higher temperatures, due to rapid stress relaxation, the unloading procedure started after σ reached $\approx0.7\times\sigma_0$ during the first relaxation run. The obtained results are shown in Fig. 2 for glassy $Zr_{46}(Cu_{4/5}Ag_{1/5})_{46}Al_8$ in terms of the temperature dependence of the effective surface shear internal stress σ_i normalized by σ_0. The error bars correspond to the stress ranges where the relaxation changes from purely normal (σ decreases with time) to purely abnormal (σ increases with time). Unloading by smaller $\Delta\sigma$ would result in smaller error bars. The normalized internal stress σ_i/σ_0 gradually decreases with temperature from $\approx$0.8 at $T=450$K ($T\approx0.64\times T_g$) to $\approx$0.08 at $T=638$K ($T\approx0.91\times T_g$). The results obtained for $Zr_{46}Cu_{46}Al_8$ are fully similar and not given in Fig. 2. This Figure shows that the internal stress turns to be

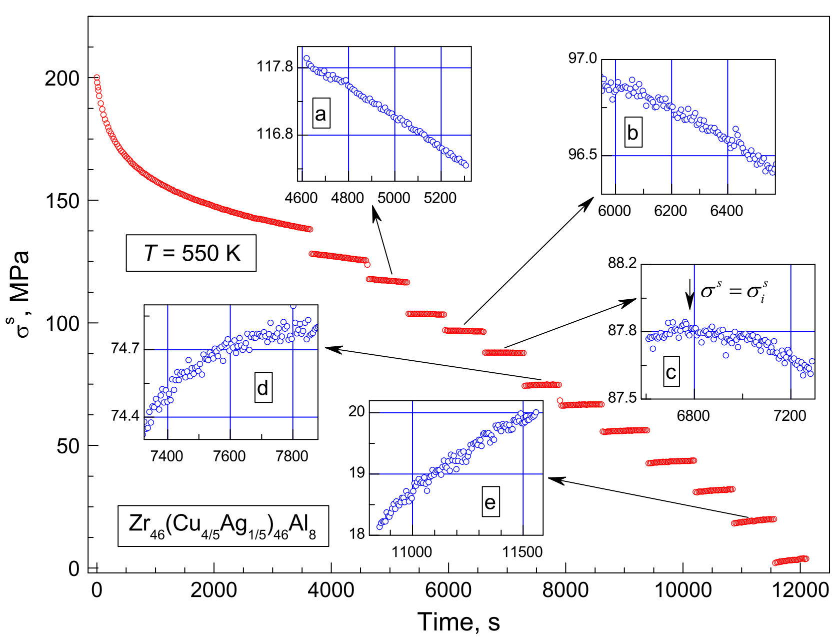

Fig. 1. An example of a stress relaxation test upon stepwise unloading. The insets *a* to *e* give stress relaxation data portions on an enlarged scale. Upon consecutive unloading, the stress relaxation changes from normal (applied shear stress σ decreasing with time) to abnormal (σ increasing with time). The latter case corresponds to the inequality $\sigma<\sigma_i$, where σ_i is the effective internal shear stress, which can be determined from the condition $d\sigma/dt=0$ (see inset *c*).