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Structural rejuvenation in bulk metallic glasses

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Abstract—Using high-energy X-ray diffraction we study structural changes in bulk metallic glasses after uniaxial compressive homogeneous deformation at temperatures slightly below the glass transition. We observe that deformation results in structural disordering corresponding to an increase in the fictive, or effective, temperature. However, the structural disordering saturates after yielding. Examination of the experimental structure and molecular dynamics simulation suggests that local changes in the atomic connectivity network are the main driving force of the structural rejuvenation.

Keywords: Bulk metallic glasses; Structure; Rejuvenation; Creep; X-ray diffraction

1. Introduction

When bulk metallic glasses (BMGs) are subjected to stress at room temperature most of them fail catastrophically through the formation and slip of shear bands without showing much plasticity [1,2]. Only a small number of glass systems, mostly based on noble metals with a high Poisson ratio, exhibit room-temperature plasticity [3–6]. However, even ductile BMGs are sensitive to annealing-induced embrittlement [7–10]. Therefore, effective and simple methods to improve room-temperature plasticity are sought after, and numerous methods have been explored. Generally, they can be categorized into homogeneous [8,11] and inhomogeneous types [12–15]. In the homogeneous method structural rejuvenation occurs throughout the whole volume, whereas the inhomogeneous method relies upon localized plastic deformation resulting in multiple shear bands being introduced into BMGs. The homogeneous method is more attractive since it eliminates the risk of damaging BMG samples by stressing them beyond the yield point. Recently, an effective homogeneous method to recover plasticity by using thermomechanical creep has been found [8]. A structural study of the BMG samples that showed creep-induced plasticity indicated signs of microscopic structural rejuvenation [8]. However, the atomistic mechanism of structural rejuvenation remains elusive. Also, it remains to be determined which part of the creep (anelastic-recoverable or plastic) is contributing to rejuvenation. The purpose of this work is to determine the atomistic mechanism of rejuvenation through an experimental structural study using high-energy X-ray diffraction and computer simulation.

The glassy state is characterized by a fictive, or effective, temperature (T_f) corresponding to the supercooled liquid state into which the glass is frozen [16,17]. Accordingly, the physical implication of structural rejuvenation is an increase in T_f. Numerical simulation based upon the shear transformation zone (STZ) concept [16] suggests that T_f is a function of a strain rate, and so deformation at a sufficiently high strain rate could generate structural rejuvenation. From both theoretical and practical perspectives, it is important to validate these predictions. In this paper, we examine experimentally structural rejuvenation in relation to the strain rate and deformation stage. Because direct measurement of T_f is impractical we carried out structural studies of BMGs deformed at different strain rates assuming that a particular T_f corresponds to a unique structural state of a glass characterized by a pair distribution function (PDF). The fictive temperature can be related to the pressure fluctuations in the glassy structure (e.g. [18]) which can be related to the PDF peak width and thus peak height. It was shown that $\langle p^2 \rangle$ (where p is the atomic level pressure) is related to the height and width of the RDF peak [19,20]. The distribution of $\langle p^2 \rangle$ reflects the structural disorder in glass. For example, if the sample is annealed and undergoes structural relaxation, the distribution becomes narrow, the peak width becomes narrower and the peak height becomes higher. The opposite is true if we introduce disordering. In

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this study we examined the change in the averaged PDF peak height, which linearly tracks the fictive temperature. In principle, the fictive temperature can be directly determined by knowing the PDF of the samples equilibrated at various temperatures. In this study we did not carry out such an elaborate calibration procedure, so that the T_f is only semiquantitatively known from the PDF. Then, combining experimental results with a qualitative simulation study of the structural evolution in a model glass during fixed strain-rate deformation we elucidate the structural micromechanism of rejuvenation. We demonstrate that the atomistic mechanism of structural rejuvenation is due to topological rearrangement in the atomic connectivity network.

2. Experiment

A BMG with the composition $Zr_{55}Cu_{30}Ni_5Al_{10}$ ($T_g = 434$ °C) was prepared by a tilt casting method [21]. Slabs with dimensions of 2.5 mm × 2.5 mm × 5 mm were cut from a single as-cast rod using electric discharge machining (EDM). All samples were annealed at 400 °C for 24 h to acquire the same reference state. Then they were polished to remove contamination on their surfaces. Using a uniaxial mechanical testing system (Instron 5881, Shakopee, MN), displacement-controlled compression tests were conducted at 400 °C, which is the same as the annealing temperature but lower than the glass transition temperature. Samples were first heated and stabilized at 400 °C for 20 min altogether. After the loading test, samples were immediately quenched into iced water. The plastic strain for each sample was measured using a caliper with an accuracy of ±0.005 mm.

A thin slice parallel to the loading axis was cut from each sample by EDM and then finely polished into pieces 0.5 mm thick for X-ray diffraction measurements. The high-energy X-ray study was carried out at the 6-ID and 1-ID beam lines of the Advanced Photon Source, Argonne National Laboratory. The beam energy was 100 keV. A two-dimensional (2-D) stationary detector, placed ~34 cm behind the sample and having 2048 × 2048 pixels with 200 × 200 μm^2 pixel size, was used to collect diffraction patterns. Calibration was performed using a CeO_2 NIST powder standard. FIT2D software was used to correct for beam polarization, dark current and data binning.

Molecular dynamics (MD) studies were performed on a model of amorphous iron [22] with 16,584 atoms. A monoatomic system rather than the alloy used in the experimental study was chosen to eliminate the effect of chemical composition. A realistic MD simulation of an experimentally studied glass is not practical because of the required model size and low strain rates that cannot be reproduced in the simulation. To resolve qualitatively changes in the local topology (thus ignoring chemical ordering) we use a monoatomic model system with strain rates achievable by simulation. Initially, the structure was prepared by an instant quench from a liquid at 1500 K to a glass at 700 K, followed by cooling process at 0.2 K ps^{-1}. After gradual cooling to room temperature, the system was relaxed for 2 ns under the NTP ensemble with pressure set to zero and temperature at 300 K. Initially, the positions of all atoms were displaced with a uniform tensile strain of 2% and a Poisson ratio of 0.33. The deformed structure was relaxed at 300 K with an effective strain rate of 10^6 s^{-1}. During the relaxation process the pressure perpendicular to the tensile direction was kept to be zero. The system stays glassy at this strain rate. The PDF was evaluated for two groups of atoms: atoms that changed their local topology by displacing from the first neighbor shell, and atoms that maintained their original environment.

3. Results and discussion

3.1. Structural characterization

Samples quenched to room temperature following fixed strain-rate deformation show evidence of structural anisotropy in the plane parallel to the loading axis while they remain isotropic in the plane normal to the loading direction. This is evidenced in the 2-D diffraction patterns. For each plane, we measured two orientations rotated by 90° about the axis parallel to the X-ray beam. The diffraction rings for the plane parallel to the stress are distorted and are elliptical in shape. Therefore, the difference in the intensities of these two images shows characteristic a 2-fold pattern as displayed in Fig. 1a. On the other hand, for the plane normal to the stress, the diffraction pattern is isotropic and the difference between 0° and 90° shows negligible intensity fluctuations, as shown in Fig. 1b. Since the structure of the BMG after deformation is no longer isotropic a spherical harmonics expansion [23–25] is employed to separate the isotropic and anisotropic components of the structure function $S(Q)$ and the PDF $g(r)$:

$$S(\vec{Q}) = \sum_{l,m} S_l^m(Q) Y_l^m\left(\frac{\vec{Q}}{Q}\right), \quad g(\vec{r}) = \sum_{l,m} g_l^m(r) Y_l^m\left(\frac{\vec{r}}{r}\right). \qquad (1)$$

Because of the uniaxial symmetry, anisotropic terms are limited to the ($l = 2$, $m = 0$) elliptical component. Next, by Bessel transformations the isotropic part of the pair distribution function PDF, $g_0^0(r)$, and anisotropic component, $g_2^0(r)$, are obtained [23,25]. The isotropic part of the PDF describes the average structure of the glass and is intimately related to the fictive temperature. The $g_2^0(r)$ component characterizes the anisotropy introduced by the high-temperature deformation. During the high-temperature deformation the applied stress induces local structural rearrangements (relaxation). After quenching and removing external stress, these local rearrangements produce local stress fields that add up to a non-zero stress, which is balanced by a long-range stress due to macroscopic deformation of the sample. This macroscopic deformation is the signature of anelastic deformation and represents a memory state of the high-temperature creep deformation under constant stress. The structural anisotropy due to this effect has been observed by previous X-ray diffraction studies [23–28].

The strain anisotropy due to affine (uniform) deformation gives rise to the elliptical ($l = 2$, $m = 0$) component of the anisotropic PDF:

$$g^0_{2,\mathrm{affine}}(r) = \varepsilon \frac{2(1+\nu)}{3\sqrt{5}} r \frac{d}{dr} g_0^0(r). \qquad (2)$$

Here, ε is a uniaxial strain and ν is the Poisson's ratio. The $g_2^0(r)$ for a sample that underwent anelastic deformation is similar to the affine PDF beyond ~9 Å. However, at a shorter range it deviates significantly from Eq. (2) [21–26].