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Estimation of the shear transformation zone size in a bulk metallic glass through statistical analysis of the first pop-in stresses during spherical nanoindentation

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The size of the shear transformation zone (STZ) that initiates the elastic to plastic transition in a Zr-based bulk metallic glass was estimated by conducting a statistical analysis of the first pop-in event during spherical nanoindentation. A series of experiments led us to a successful description of the distribution of shear strength for the transition and its dependence on the loading rate. From the activation volume determined by statistical analysis the STZ size was estimated based on a cooperative shearing model.

Keywords: Bulk amorphous materials; Nanoindentation; Yield phenomena; Shear transformation zone

It is widely accepted that the fundamental carriers of plasticity in amorphous alloys are the shear transformation zones (STZs), which are atomic clusters that undergo inelastic shear straining under the influence of an applied stress [1–3]. STZs in metallic glasses are the analogs of mobile dislocations in crystalline solids. While dislocations can be imaged using a variety of techniques, direct observation of STZs is nigh impossible because of their transient nature. While it is generally understood that the activation of STZs occurs preferentially in those regions of the material where the atomic packing efficiency is relatively smaller (higher free volume content), the volume (or size) of an STZ is still an actively debated point. This is particularly so because of the difficulty associated with direct experimental assessment of it. This led to the study of STZ size either through molecular dynamics (MD) simulations or indirect experiments. For example, Pan et al. [4,5] utilized the rate dependence of hardness to estimate the volume of STZs. For this they modified the cooperative shear model (CSM) of Johnson and Samwer [6], which was developed on the basis of the potential-energy landscape. However, hardness is an indicator of the resistance to plastic flow rather than its initiation (yielding). Since plastic flow in metallic glasses occurs through localization (shear bands), negative rate sensitivity ensues. In such a scenario one cannot simply obtain the underlying kinetics of STZs.

One of the most popular methods to study small-scale yielding of materials is nanoindentation. During nanoindentation with a spherical or round indenter the load–displacement (P–h) curves often exhibit either a sudden burst of displacement when the test is performed under load control or a sharp load drop if the test is conducted under displacement control. This phenomenon is often referred to as “pop-in”. Since the pioneering work by Page et al. [7] the study of pop-ins, especially the first pop-in, has gathered wide interest because it indicates to elastic-to-plastic transition in crystalline and amorphous materials [8–14]. Recently Wang et al. [15] attempted to determine the size and activation energy of STZs in an Au–Ag–Pd–Cu–Si bulk metallic glass (BMG) by analyzing the high temperature nanoindentation pop-in data according to Argon’s classical constitutive equations [1]. Although a good first attempt, this study did not consider the wide variability in the pop-in data, which points to the stochastic nature of STZ

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activation and, hence, warrants a model that is intrinsically statistical in nature. We attempt such in this paper, with the objective of estimating the STZ size by statistical analysis of the pop-in data within the premise of the CSM model. From a series of nanoindentation experiments on a Zr-based BMG with a spherical indenter we have obtained the maximum shear stress at the elastic to plastic transition and its dependence on the loading rate. This data is utilized to ascertain the STZ size.

An ∼7 mm diameter and ∼70 mm long rod of Zr-based BMG, $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ (commercial designation Vit 105) was examined. No crystalline peak was detected in the X-ray diffraction (XRD) spectra of the specimen [16]. Experiments were conducted at room temperature using a Nanoindenter-XP (formerly MTS-now Agilent, Oak Ridge, TN) instrument equipped with a spherical tip. Hertzian contact analysis [17] of indentations made on fused quartz was utilized to estimate the tip radius R as 31.5 μm based on the assumption that the sample surface, which was polished to a mirror finish prior to testing, is flat. It is notable that R in the original Hertz analysis is the "relative" radius of the sphere-to-sphere contact and thus determined as $1/R = 1/R_i + 1/R_s$, where R_i and R_s are the radius of the indenter and sample, respectively. For a flat surface $1/R_s = 0$. Tests were conducted in load control mode at loading rates of 0.5, 1, 5, 10, and 20 mN s^{-1}. More than 120 tests were conducted at each rate so as to obtain statistically significant data sets. Thermal drift was maintained below 0.05 nm s^{-1} in all experiments.

Figure 1 shows a representative P–h curve exhibiting pop-ins. Figure 1 also shows the P–h curve obtained at low load (before the first pop-in), with the loading part of the curve retraced by the unloading curve, indicating that deformation is solely elastic before the first pop-in. This elastic behavior of the material during spherical indentation can be described by Hertzian contact theory [17]:

$$P = \frac{4}{3} E_r \sqrt{R} \cdot h^{\frac{3}{2}} \tag{1a}$$

and

$$\frac{E_s}{1 - \nu_s^2} = \left(\frac{1}{E_r} - \frac{1 - \nu_i^2}{E_i} \right)^{-1} \tag{1b}$$

where E and ν are the elastic modulus and Poisson ratio, with the subscripts s and i indicating the sample and the indenter. The reduced modulus E_r accounts for the fact that elastic deformations occur in both the indenter and the sample. Since a diamond tip is used ($E_i = 1141$ GPa, $\nu_i = 0.07$) [18]. By fitting the loading part of the P–h curve to Eq. (1a) the indentation modulus of the samples $E_s/(1 - \nu_s^2)$ was estimated to be ∼100 GPa, which is in agreement with the reported value in the literature (∼103 GPa based on $E_s = 89$ GPa, $\nu_s = 0.37$) [19].

The maximum shear stress at the first pop-in τ_{max} represents the critical shear strength for the onset of plasticity in the indented material. In spherical indentation τ_{max} occurs at a distance approximately half the contact radius directly below the rotational axis of the contact and is given as [17]:

$$\tau_{max} = 0.31 p_0 = 0.31 \left(\frac{3}{2} p_m \right) = 0.31 \left(\frac{6 E_r^2}{\pi^3 R^2} P \right)^{\frac{1}{3}} \tag{2}$$

where p_0 and p_m are the maximum and mean pressures of the contact, respectively. Even for indentations conducted under identical testing conditions τ_{max} estimated from the first pop-in load is distributed over a wide range of ∼1.2–3.4 GPa, as seen in Figure 2. An important feature in Figure 2 is that τ_{max} is rate dependent; a higher loading rate generally results in a higher τ_{max}. These results are in contrast to some of the published literature. Ng et al. [20], who have examined the serrated flow (i.e. a series of pop-ins) behavior of a (Cu–Mg–Y)–Be BMG at room temperature using a sharp indenter, concluded that the flow is insensitive to strain rate. We wish to point that they used a sharp indenter, which means that the strains imposed are large and, hence, plastic flow is dominated by shear band kinetics [20]. In contrast, we used a spherical indenter and, hence, the plastic strains at the first pop-in event are rather small and capture the elastic-to-plastic transition. Recently Packard et al. [15] reported that in a Pd- and a Fe-based BMG the first pop-in stresses are rate- and temperature-independent. This observation led them to suggest that the distribution of the first pop-in stress predominantly originates from scattering in the local atomic structure rather than thermal fluctuations. However, at least in the Zr-based BMG examined here, detectable rate dependency of the pop-in stress exists, indicating that, with the inherent inhomogeneity of the atomic configuration in the amorphous state, thermal fluctuation could still have a role to play in the pop-in event. Therefore, it is likely that the rate dependency of the first pop-in event depends on the BMG composition (and thus

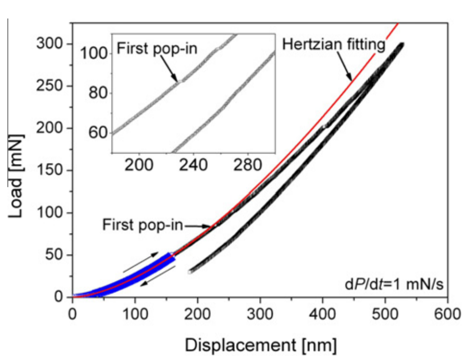

Figure 1. A representative P–h curve showing pop-ins and a Hertzian curve. (Inset) The loading part near the first pop-in.

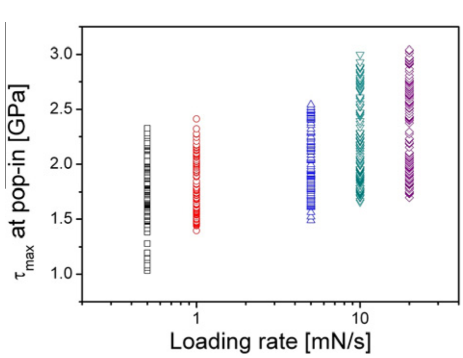

Figure 2. Variation in the maximum shear stress for the elastic to plastic transition τ_{max} with loading rate.