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Constitutive model for inhomogeneous flow in bulk metallic glasses

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Abstract

This study presents results on the mechanical properties of Zr-based bulk metallic glasses over a wide range of temperatures (77–673 K) and strain rates ($3.3 \times 10^{-5} - 0.2\ s^{-1}$), and provides novel information on the kinetics of inhomogeneous deformation in metallic glasses. The disappearance of serrated flow, i.e. temporal homogenization, below a critical temperature and above a critical strain rate corresponds to a transition of the asymptotic strain rate sensitivity (SRS) from negative to positive values. Based on thermally activated shear processes a temperature and strain rate dependent deformation model is presented that includes a time-varying state variable which represents the local degree of relaxation in the shear band after a shear event has taken place. The model makes possible a theoretical description of the observed SRS and the spatio-temporal evolution of plastic deformation in metallic glasses.

Keywords: Bulk metallic glasses; Inhomogeneous deformation; Strain rate sensitivity; Deformation kinetics; Activation volume

1. Introduction

Bulk metallic glasses (BMGs) exhibit excellent mechanical properties, such as high strength, elastic strain limit and fracture toughness, and therefore represent a promising class of materials for structural and functional applications [1–5]. The lack of long-range order in the atomic structure of amorphous alloys makes their plastic deformation fundamentally different from that in crystalline solids, where deformation can generally be described in terms of the underlying dislocation dynamics. The corresponding underlying mechanism in the deformation process of amorphous alloys is, however, not yet fully understood.

Phenomenologically, the mechanical behavior of monolithic BMGs can be classified by either homogeneous or inhomogeneous deformation. Homogeneous deformation usually takes place at elevated temperatures (>0.7 T_g, where T_g is the glass transition temperature) and can be described by Newtonian viscous flow at low strain rates or as non-Newtonian at higher strain rates [6–8]. Although the macroscopic mechanical response of a metallic glass often displays elastic–perfectly plastic behavior at low temperatures ($T << T_g$), deformation is inhomogeneous and localized in narrow shear bands with a thickness of ~10 nm [9], limiting macroscopic plasticity. The activation and propagation of individual shear bands gives rise to serrated flow, which manifests itself as small displacement bursts or stress drops in the stress–strain curve during plastic deformation [10–13]. Similarly, the appearance of new surface steps (shear bands) has recently been correlated with displacement bursts ("pop-ins") in the load displacement curve during nanoindentation testing [14,15].

The inhomogeneous deformation behavior of BMGs is strongly influenced by the imposed strain rate. Mechanical tests at room temperature have shown a slight decrease in fracture strength with increasing strain rates (negative strain rate dependence) [12,16–20]. At high strain rate, contrasting behavior (positive strain rate dependence) has also recently been observed [21]. In addition, its influence on plastic strain to failure is controversial: both an enhancement [20] and a reduction [17] upon an increase in the strain rate have been reported. However, there is strong

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experimental evidence that the temporal intervals and the load variations of the flow serrations and pop-ins diminish with increasing compressive strain rate [12,17,21–25] and nanoindentation load rate [14,15,26–28], respectively. Strain rate also exerts a significant influence on the formation of shear bands in BMGs. Jiang et al. [22–25] recently reported that the number of shear bands in a Zr-based BMG observed ex situ on uniaxially compressed specimens decreases with increasing strain rate (in agreement with preliminary observations we made on Zr-based BMGs). This trend contrasts with previous indentation studies that reported that higher strain rates promote more shear bands [14,27,29,30]. Based on this, Schuh et al. [27,28] proposed a new "homogeneous II" regime in the deformation map of BMGs, where the deformation rate exceeds the characteristic rate of shear band nucleation. Consequently, the simultaneous operation of multiple shear bands (i.e. homogenization in time) leads to macroscopically more homogeneous flow (disappearance of pop-ins). These nanoindentation experiments led to the development of a model describing the dependency of an increasing shear band density with increasing strain rate [31]. Here it is, however, important to highlight the differences between uniaxial compression and nanoindentation tests: the deformation in nanoindentation is complex due to the fact that the strain gradient constantly changes during penetration, so that reactivation of the same shear band is less likely for geometrical reasons than in uniaxial compression. Thus an increase in shear band density with increasing strain rate is not expected in uniaxial compression tests. Despite this discrepancy, the different studies agree on an apparent homogenization in time at higher strain rates. Similarly, it has been shown that a decrease of testing temperature leads to a decrease in the magnitude of stress drops and the associated time intervals between two serrations [11,24,32,33], and at the same time to an increase in strength (without loss of ductility) [13,34].

It has been known for some time that shear bands in BMGs are susceptible to preferential etching [10], indicating a structural change in the deformed material (i.e. increased free volume content and/or chemical reordering). Over the last few years new insights into this structural evolution during shear banding have underlined the importance of formation and coalescence of free volume. Higher free volume contents have been measured after deformation, e.g. by positron annihilation spectroscopy (PAS) [35–37], X-ray diffraction (XRD) and differential scanning calorimetry (DSC) [38,39]. The PAS investigations also indicate that the content of certain alloying elements around free volume sites is significantly enhanced at the expense of the other, remaining constituents of the BMG [35–37]. The coalescence and spontaneous formation of nanometer-scale voids during deformation has also been experimentally observed [40] via high-resolution transmission electron microscopy, and confirmed according to thermodynamic considerations [41]. Depending on the stress state and composition of the metallic glass, even nanocrystallization in shear bands has been seen in aluminium-based amorphous alloys [42] and explained by dynamic flow dilatation and the attendant enhancement in atomic diffusional mobility [43].

The serrated flow phenomenon and the associated inhomogeneous deformation are not unique to BMGs. In certain crystalline solids they can be observed in distinct temperature, strain and strain rate ranges and are commonly known as the Portevin–Le Châtelier (PLC) effect [44]. The physical basis for appearance of the PLC effect is a negative strain rate sensitivity, which originates from dynamic strain aging (DSA) caused by the interaction of moving dislocations with mobile solute atoms [45–48]. Lower strain rates or higher temperatures allow for faster impediment of the dislocation movement by diffusing solutes, generating a stronger material and thus negative strain rate sensitivity.

The present investigation elucidates the effects of strain rate and temperature on the inhomogeneous deformation behavior of Zr-based BMGs. Building on our experimental results (strain rate sensitivity and activation volume measurements at temperatures of 77–673 K and strain rates of 3.3×10^{-5}–$0.2\ s^{-1}$), we develop a constitutive formalism which rationalizes the inhomogeneous flow kinetics and in particular the correlation between presence/absence of serrated flow and strain rate sensitivity. In this respect, Section 2 gives a short overview about deformation models in metallic glasses and provides the basis for the theoretical discussion (Section 5) of our experimental findings (Sections 3 and 4).

2. Deformation models

While the homogeneous flow of metallic glasses can be described relatively well using rate theory and rheological models that average the operation of many local atomic-scale events, a fundamental description of inhomogeneous flow remains elusive. Phenomenologically, shear band formation in amorphous alloys is a strain localization phenomenon and can be related to a local change of viscosity in narrow bands near planes of maximum shear stress. Basically, two models have emerged in the past to explain the viscosity drop within shear bands: temperature rise [49] and dilatation of the atomic structure (i.e. generation of free volume) [50].

Many authors have discussed the possibility of adiabatic temperature rise during shear band propagation, and possibly even local melting during fracture. From theory and (thermographical) experiments, estimates of the temperature rise in shear bands have varied from ~0.1 to hundreds of K [51–53]. By applying a fusible coating it has recently been shown that the heat content in shear bands corresponds to a maximum temperature rise of several thousand K [54]. Since the observed shear band thicknesses are less than can be explained by thermal diffusion lengths, however, the authors concluded that the temperature rises are a consequence of shear band localization or frictional