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Shear transformation zone dynamics model for metallic glasses incorporating free volume as a state variable

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Abstract

A mesoscale model, shear transformation zone dynamics (STZ dynamics), is employed to investigate the connections between the structure and deformation of metallic glasses. The present STZ dynamics model is adapted to incorporate a structure-related state variable, and evolves via two competing processes: STZ activation, which creates free volume, vs. diffusive rearrangement, which annihilates it. The dynamical competition between these two processes gives rise to an equilibrium excess free volume that can be connected to flow viscosity via the phenomenological Vogel–Fulcher–Tammann relation in relaxed structures near the glass transition temperature. On the other hand, the excess free volume allows glasses to deform at low temperatures via shear localization into shear bands, even in the presence of internal stress distributions that arise upon cooling after processing.

Keywords: Metallic glasses; Shear transformation zone; Free volume; STZ dynamics model; Shear bands

1. Introduction

Metallic glasses exhibit a great variety of deformation behaviors, depending upon conditions [1]. At low temperature and high stress, the deformation is highly inhomogeneous, and the plastic strain is localized into shear bands. In contrast, at high temperature and low stress, deformation is homogeneous, exhibiting Newtonian flow rheology that gives way to exponential rheology as the stress rises. Homogeneous flow is also associated with significant transients that follow any change in conditions (i.e., stress). The details of how all these diverse behaviors are connected in metallic glasses pertain to the underlying deformation mechanism of local shear shuffling in the amorphous structure. In particular, the shear transformation zone (STZ) originally proposed by Argon [2] and further observed and studied widely in computational simulations [3–7], has been accepted as a "unit process", which underlies the deformation of metallic (and other) glasses. STZs are essentially local clusters of a few atoms that can rearrange collectively in response to a shear stress. This local rearrangement involves not only the displacement of atoms, but also an anelastic reconfiguration of atomic neighbors and a redistribution of free volume within the atomic cluster. This free volume redistribution is not only a transient process, but is also believed to involve local, permanent changes to the excess free volume. The local accumulation of excess free volume is believed to facilitate shear localization through local softening in the vicinity of previously deformed regions [2].

The concept of free volume as an important internal state variable for glasses pre-dates the STZ description of glass deformation, beginning with developments by Cohen and Turnbull [8,9]. They laid the groundwork for scalar free volume evolution equations that have been widely and successfully applied by experimentalists to explain relaxation behavior during glass formation from the super-cooled liquid regime [10–12]. In addition, Spaepen [13] extended this approach to glass deformation on the

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basis of two competing processes mediated by free volume: free volume creation due to atomic motion driven by a shear stress vs. free volume annihilation via diffusive rearrangement. This extended phenomenological model is capable of capturing the homogeneous and heterogeneous deformation modes in metallic glass, and is especially useful for describing deformation transients.

Both the STZ (as a fundamental unit process for glass deformation) and free volume (as an internal state variable that evolves with time, temperature, stress and position) are simplified pictures of what actually happens in a glass. There is a vast spectrum of STZs and STZ-like atomic rearrangements that occur in deforming glasses, and this spectrum is only poorly understood at present [14–16]. And "free volume" is often used as a catch-all internal variable that subsumes a far greater complexity of internal states, including chemical and topological short- and medium-range order, atomic stresses and strains [17–19]. Neither picture (STZ or internal state variable) is yet fully worked out, and it is for this reason that simulation work on metallic glasses is essential to progress on understanding of glass deformation [20]. There is an inherent lack of direct experimental approaches to these events in atomic glasses and, as such, atomistic simulations provide many details of plasticity in metallic glasses that would otherwise be unavailable [21]. Yet, even though these simulations provide significant detail, they suffer from a limitation in both length and time scales (typically a few tens of nanometers during a few tens of nanoseconds). Atomistic methods are therefore less useful for investigating large-scale events that occur over a long time scale, e.g., glass formation by cooling from the melt at realistic rates.

In this context, mesoscale modeling methods are needed to bridge the gap between atomistic simulations and experiments by averaging out atomistic effects and accounting only for an ensemble of characteristic events. Such approaches, on the one hand, enable simulations to access larger scales and, on the other hand, retain a description of elementary deformation physics. For example, Baret et al. [22] proposed a *depinning* model to investigate long-range spatial and temporal correlations in "depinned" local shear transition events. The critical stresses that lead to depinning can involve a stochastic distribution in order to account for the structural disorder of metallic glass. By introducing an age parameter that can vary the yield stress distribution, the authors were able to show an age-dependent shear banding behavior. A *fluidity* model developed by Picard [23,24] associated the characteristic transition times with deformation and relaxation and produced complex spatio-temporal patterns of transformation events.

"STZ dynamics" is another mesoscale model proposed by Homer et al. [25–27]. In this model a simulated metallic glass is partitioned into an ensemble of potential STZs that are mapped onto a finite element mesh. The activation of individual STZs is selected via the Kinetic Monte Carlo (KMC) algorithm originally introduced by Bulatov and Argon [28–30], which uses an energy-based yield criterion according to a Boltzmann probability, with local biasing from the stress state. As a consequence, the model incorporates a time scale and at the same time thermal effects, which are missing from other mesoscale models. The activation of STZs leads to stress and strain redistribution, which is solved accurately via the finite element method (FEM). The updated elastic field resulting from STZ activations in turn affects the rate of subsequent activation events and thus the dynamical evolution of the system. With this framework, the model is able to capture the basic behaviors of metallic glasses, including high-temperature homogeneous flow, and low-temperature strain localization into shear bands.

One limitation of STZ dynamics as it has been implemented to date, however, involves the use of a single, fixed energy barrier for STZ activation, which cannot capture the mechanical effects of structural evolution (e.g., local structural hardening or softening) as the system evolves. One consequence of this limitation is that autocatalytic accumulation of structural change (as in the dynamic softening that leads to shear localization) is difficult to trigger. In fact, when disorder pre-exists in the internal stress fields of the system (e.g., due to cooling from the liquid state), the dynamical correlations between STZ activations are not large enough to overcome the internal stress fields (structural disorder) to allow STZ activations to correlate and form shear bands [26,31,32]. In prior STZ dynamics simulations shear bands are only seen when the initial configuration is uniform and free of structural noise [26], which is neither realistic for a glassy material nor in line with the perception that local structural softening is responsible for shear banding in glasses in the first place.

In this paper, our purpose is to continue the development of the STZ dynamics modeling framework by incorporating a structural state variable into it. In line with the above discussion, we introduce *excess free volume* into the STZ dynamics model of Homer and Schuh [26] as a simple internal parameter that can track local structural change and allow for dynamical softening and hardening. We allow system evolution via two competing processes: free volume creation via STZ activation vs. free volume annihilation via diffusive rearrangement; the treatment is in the spirit of the original works by Argon [2] and Spaepen [13]. In what follows, we first present our basic modeling framework followed by a two-dimensional (2D) FEM implementation. Then we explore the interplay between evolution of excess free volume and metallic glass deformation over a range of thermal and mechanical loads. In particular, we show that excess free volume-assisted shear banding occurs at low temperatures, even in the presence of structural disorder in "processed" samples.

2. Modeling framework

The STZ dynamics model is built upon that of Homer et al. [25–27] and uses the same general apparatus, consisting of a finite element mesh and a KMC algorithm.