



The role of time in activation of flow units in metallic glasses



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ABSTRACT

Flow units, which accommodate deformations and initiate transformation from glass to supercooled liquid state, have been proved to significantly influence the properties of metallic glasses (MGs). We study the time dependent activation of the flow units in MGs, and find that the size and the fraction of the flow units increase with the applied time, which lead to the time dependences of the mechanical behaviors, flow phenomenon and relaxations of MGs. A diagram for the flow in glass based on the concept of flow unit, activation time, and imposed energy (stress or temperature) is constructed to understand the deformations and flow mechanisms of MGs.

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1. Introduction

Substances can be divided into solids, liquids and gases based on their macroscopic properties. However, the pitch drop experiment [1] by Parnell has shown that some solids and liquids are hard to be distinguished through their appearance. So another classification, which distinguishes the substances by the orders in their microstructures, is proposed [2,3]. Crystals are the substances with both short-range and long-range orders, while gases have neither of them. The liquids and glasses, which can be called amorphous substances, do not have long-range orders but have short range orders, and this leads to some similar features of glasses and liquids [2].

Unlike freezing or boiling, the transition from liquid to glass has no substantially detectable change in structure. Although it is still disputed whether the glass transition is a slow kinetics phenomenon or a thermal precursor of an obscured “ideal” phase transition [3], experimental results [4] have shown that the glass transition temperature (T_g) is time dependent. Besides, experiments and simulations [5–7] have shown that the slow flow features and mechanical properties of a glass is also time dependent. When a glass is deformed with a low enough strain rate, the glass can behave like a Newtonian liquid [5,6]; while with a very high strain rate, liquid can show fracture behavior similar to that of brittle glass [5].

Metallic glasses (MGs) are the simple atomic glasses among the wide spectrum of glass-forming systems [7–9], because their random disordered structure can be considered as the dense random packing of spheres [10]. The MGs then offer a model system to study the structural origin of flow behavior, deformations, and the glass transition [12–16]. Recently, experimental and simulation evidences [17–27] have shown that the deformation behavior and the mechanical properties of MGs are determined by the intrinsic heterogeneous structure, especially the liquid like regions or flow units in MGs. The flow units play a crucial role in the elastic and plastic deformations, relaxations, rejuvenation, and mechanical and physical properties of MGs. For the features and properties of MGs are time dependent, it is supposed that the flow units are also dependent on time. In this paper, we attempt to characterize the time dependent feature of the flow units in MGs. We find that the activation process, size and distribution of the flow units in MG are indeed time dependent, which is in accordance with experimental results of time dependent macroscopic properties of MGs. A time-relevant diagram for the flow in metallic glasses is proposed.

2. Theory and model

The microstructure of MGs is heterogeneous with nano-scale localized liquid-like sites called flow units embedded in elastic matrix, which strongly affect the features and properties of the glasses [18,22,23]. When stress or temperature is applied, the flow units in MGs will be activated, dissipate the applied energy and flow inelastically. According to the elastic model [9], the activation

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energy W_i of a flow unit in a glass is mainly elastic energy, and is controlled dominantly by its shear modulus G_i [9,28]:

$$W_i = aG_iQ_i, \quad (1)$$

where a and Q_i are the constant coefficient and volume of the flow unit, respectively. The activation energy and activation time have an Arrhenius relation [9]: $t = \tau_i \exp(W_i/kT)$, or $W_i = kT \ln t / \tau_i$, where T is temperature, k is the Boltzmann constant, and τ_i is the intrinsic relaxation time of the flow unit. Therefore, the relation between activation time and Q_i is:

$$Q_i = \frac{kT}{aG_i} \ln \frac{t}{\tau_i}. \quad (2)$$

Eq. (2) indicates that the flow units are not activated spontaneously because the activation process needs sufficient time, and the larger (smaller) flow units with longer (shorter) intrinsic relaxation times need longer (shorter) time to be activated. In other words, the dynamic and energy distribution of the atoms in a flow unit are inhomogeneous, and the flow unit is actually activated gradually: the softest region in a flow unit will be activated first and the harder ones later. Fig. 1(a) is a schematic illustration of a flow unit in MG. The blue spheres represent elastic matrix of MG. The region in the center with red spheres contains the most loosely packed atoms, and these atoms have higher mobility and shortest intrinsic relaxation time τ_1 , and the orange and yellow sphere regions surrounding the red zone are atoms with lower mobility and

longer intrinsic relaxation time τ_2 and τ_3 ($\tau_2 < \tau_3$). Fig. 1(b) shows the energy distribution of the atoms in the flow unit. The energy from the center of the flow unit to the surrounding harder regions is a Gaussian-like distribution and the energy decreases quickly with the increase of the distance from the center. When the stress or thermal energy is applied, and if the activation time (t), $\tau_1 < t < \tau_2$, the central region with red atoms in the flow unit will be activated; If $\tau_2 < t < \tau_3$, the relative harder orange region will also be activated; and if $t > \tau_3$, the whole flow unit will be fully activated.

Due to the heterogeneous structure of MGs, each flow unit in MG has different average intrinsic relaxation time and activation energy, and the average relaxation time and activation energy of flow units of an MG have a broad distribution [29–31]. The average volume of flow units is $\bar{Q} = \Sigma Q_i / n$, and the average flow unit activation energy is $W = \Sigma W_i / n$, the average intrinsic relaxation time is $\tau_0 = \Sigma \tau_i / n$, where n is the number of flow units in an MG. So the $\bar{Q}$ and W are also a function of the activation time t . Based on the cooperative shearing model [28], the relation of the W and $\bar{Q}$ is [28,29]: $W = (\pi^2 / \xi) G \gamma_c^2 \bar{Q}$, where ξ is a constant, γ_c is the yield strain limit, G is the average shear modulus of the MG, and the relation of $\bar{Q}$ and time of the activated flow unit is:

$$\bar{Q} = \frac{\pi^2 kT}{8G\xi\gamma_c^2} \ln \frac{t}{\tau_0}, \quad (3)$$

the volume fraction of flow units is:

$$c = n\bar{Q}/V = \frac{\pi^2 kTn}{8VG\xi\gamma_c^2} \ln \frac{t}{\tau_0}. \quad (4)$$

Eq. (4) indicates that the density or the volume fraction of the flow units in an MG is activation time dependent.

3. Results

Fig. 2(a)–(c) schematically illustrate time dependence of size and density of flow units in an MG. For short activation time (or high strain rate) as shown in Fig. 2(a), only few and small flow units with higher energy and atomic mobility are activated; with extended time, more and larger flow units are activated as indicated in Fig. 2(b); and if the activation time is long enough, more and more flow units with higher activation energy would be activated [Fig. 2(c)], and when the density of the flow units reaches a critical value the MG will behave like liquids [6,11].

The flow units are found to correlate with properties P such as elastic moduli, hardness, and plasticity of MGs in the form of [21,30–34]: $P = P_m / (1 + \alpha)$, where α correlates to the effective fraction of flow units, and P_m is the property of glass without flow units corresponding to that of perfect crystal or ideal glass [21,31–34]. According to Eqs. (3) and (4), the time dependent activation and fraction of flow units results in the time dependent properties of MGs. We can then investigate the role of time in activation of flow units through the studying of time dependent mechanical properties and macroscopic flow behaviors in MGs.

We used a Sr-based MG to perform the uniaxial compression at room temperature (RT) to observe the yield strength changing under different strain rate ν . The rod of $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Zn}_{20}(\text{Li}_{0.55}\text{Mg}_{0.45})_{20}$ MG is 2 mm in diameter and 4 mm in length, and has a low T_g of 323 K [35]. The strain rate is equivalent to the activation time t . With a giving ν , the activation time at a given strain γ can be expressed as: $t = \gamma / \nu$. Fig. 2(d) shows that at RT and a higher $\nu = 3 \times 10^{-4} \text{ s}^{-1}$, the MG exhibits an elastic deformation followed by pure brittle behavior, and fractures into small pieces. This indicates that, due to few and small flow units in the MG are activated at RT at high strain rate or shorter activation time, no

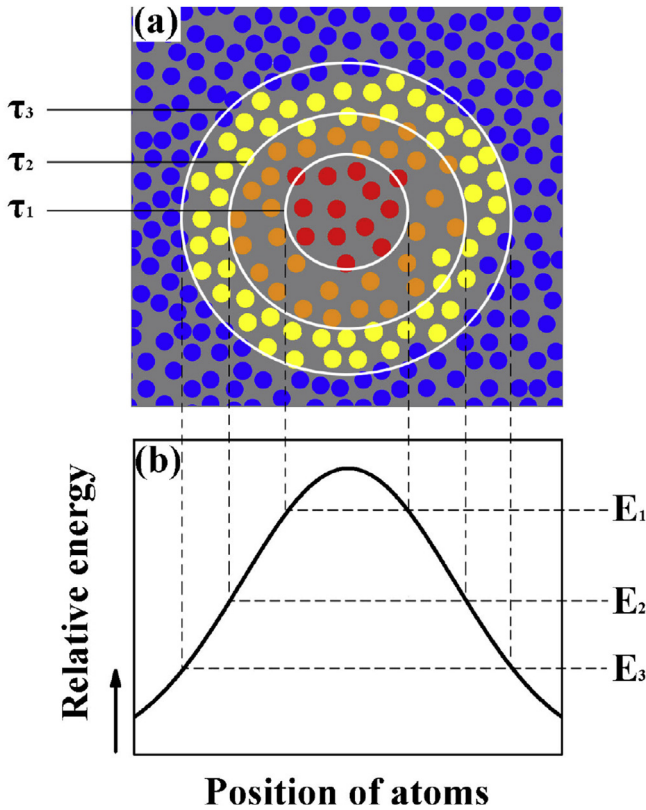


Fig. 1. (a) The schematic illustration of the atomic structure of flow unit in metallic glass. The blue spheres represent the atoms in elastic matrix region, the red spheres represent the atoms in the flow unit with shortest intrinsic relaxation time τ_1 and the orange regions and yellow regions respectively are harder atoms with lower mobility and larger intrinsic relaxation time τ_2 and τ_3 . (b) The energy distribution of the atoms in the flow unit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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