Theoretical and Applied Mechanics Letters 5 (2015) 200–204

Contents lists available at ScienceDirect

Theoretical and Applied Mechanics Letters

journal homepage: www.elsevier.com/locate/taml

Letter

# Understanding ductile-to-brittle transition of metallic glasses from shear transformation zone dilatation

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## HIGHLIGHTS

- Cooperative shearing of shear transformation zones (STZs) is assisted by free volume.
- STZ dilatational strain is introduced to understand the ductile-to-brittle transition (DBT) of metallic glasses.
- The DBT of metallic glasses is underpinned by the transition of STZs to tension transformation zones (TTZs).

## ARTICLE INFO

*Article history:*
Received 30 July 2015
Accepted 5 September 2015
Available online 28 September 2015
*This article belongs to the Solid Mechanics

*Keywords:*
Ductile-to-brittle transition
Metallic glass
Fracture
Shear transformation zone
Tension transformation zone

## ABSTRACT

A theoretical model that takes into account the free-volume aided cooperative shearing of shear transformation zones (STZs) is developed to quantitatively understand the ductile-to-brittle transition (DBT) of metallic glasses. The STZ dilatational strain is defined as the ratio of STZ-activated free volume to STZ volume itself. The model demonstrates that the STZ dilatational strain will increase drastically and exceed the characteristic shear strain of STZ as temperature decreases below a critical value. This critical temperature is in good agreement with the experimentally measured DBT temperature. Our results suggest that the DBT of metallic glasses is underpinned by the transition of atomic-cluster motions from STZ-type rearrangements to dilatational processes (termed tension transformation zones (TTZs)).



At temperatures well below the glass transition temperature, metallic glasses usually share an avenue to mechanical failure by shear-induced dilatation or free volume generation [1–5] resulting from a cascade of inelastic rearrangements of local atomic clusters, commonly called shear transformation zones (STZs) [6–8]. Macroscopically, such a shear-softening process appears as a runaway of shear bands of about 10 nm in thickness into crack propagation through the Saffman–Taylor flow instability [9], leaving microscale vein-like fracture patterns. However, recent experiments [10–13] and simulations [14,15] have revealed that the dilatation itself, whether induced by shear or hydrostatic tension, can dominate brittle fracture of metallic glasses. In this case, the crack tip propagates via cavitating events that involve a series of nanovoids nucleation and coalescence with very limited plastic growth [11,14], which retains the tip atomically sharp during propagation [16,17]. The dilatation-mediated brittle fracture is strongly supported by the resulting fracture morphologies [10–12,18–21]: very-fine dimples and nanoscale periodic corrugations. At the atomic scale, such a brittle cavitating event has been originally defined by Jiang et al. [11,22] as a tension transformation zone (TTZ) that describes the atomic cluster motion undergoing significant dilatation. For convenience of understanding, the TTZ can be regarded as a dilatation-dominated STZ, but the later is usually shear-dominated [6,7,23].

It is therefore expected that the ductile-to-brittle transition (DBT) of metallic glasses will take place if the STZs at the crack tip are restrained, whereas the TTZs are activated simultaneously. More specifically, the DBT is trigged by the change in the nature of the "transformation zones" from STZ-type to TTZ-type [11,19,21,24–26]. Nevertheless, why and how an STZ could convert into a TTZ remains to be further clarified. Very recently, we performed systematic experiments on the fracture behavior of a typical Zr-based (Vitreloy 1) bulk metallic glass at decreasing temperature from room temperature (300 K) to liquid helium temperature (4.2 K) [27]. It was observed that the cryogenic temperature can incur a sharp DBT at about 20 K, which motivates us to explore

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http://dx.doi.org/10.1016/j.taml.2015.09.002

the critical condition for the STZ-to-TTZ transition. In the present letter, we propose a STZ dilatational strain to quantitatively characterize the STZ-to-TTZ transition by taking free volume into the cooperative shearing of STZs. This model is supported by the good agreement between the predicted DBT temperature and that determined experimentally [27].

It has been recognized that plastic flow of metallic glasses occurs as a result of a series of STZ operations [2,6,7,28]. However, unlike the unit glide of a dislocation in a crystalline material that is only driven by the deviatoric stress, the potential STZs require local dilatation or free volume for the shear transformations to take place [2,29]. In fact, the STZs initiate easier in regions where the free volume is higher [8], although the STZ operation is shear-dominated [2,23]. We could thus envision an extreme situation in which an STZ experiences a remarkable dilatation rather than shearing, so that the STZ actually becomes a cavitation event, i.e., the TTZ operation per se defined previously [11]. The TTZs ahead the crack tip are widely believed to be responsible for the brittle fracture of metallic glasses [11,19,24,25,27]. Hence, how to characterize the dilatation degree of STZ holds a key to the microscopic STZ-to-TTZ transition or the macroscopic DBT in metallic glasses.

From a mechanistic point of view, the STZ dilatation should involve two characteristic scales: one is the absolute scale of dilatation and the other is the STZ volume itself, $\Omega$. It has been well accepted that the generation of free volume stems from dilatation of STZs under applied shear stress [2,8,29]. Therefore, we introduce the concept of active free volume, $v_{\mathrm{f}}^{\mathrm{a}}$, to measure the absolute dilatation of an STZ. Here we address that the active free volume is different from the static free volume defined commonly. The latter is determined at the glass transition temperature, $T_{\mathrm{g}}$, and frozen at temperatures $T < T_{\mathrm{g}}$ [30]. However, the active free volume can be mobile via stress-driven STZs even at $T < T_{\mathrm{g}}$. Reasonably, we may define the STZ dilatational strain, $\vartheta$, as the ratio of the two characteristic volumes:

$$\vartheta = \frac{v_{\mathrm{f}}^{\mathrm{a}}}{\Omega}. \tag{1}$$

This definition means that if the active free volume or the absolute dilatation of an STZ is comparable to the STZ volume itself, the STZ will be prone to undergo a significant dilatation. When the STZ dilatational strain exceeds a threshold, the STZ becomes a TTZ-type cavitation operation. It should be pointed out that the STZ dilatational strain defined here is somewhat different from the cavitational strain defined by Guan et al. [15]. The former mainly addresses the nucleation of a void due to shear (STZ)-induced dilatation, while the latter describes the void nucleation and plastic growth under a direct hydrostatic tension.

The cooperative shearing model (CSM) of STZs points out that the plastic flow occurs in metallic glasses when the barrier crossing rate of STZs reaches a critical value comparable to the applied strain rate [31]. Considering that the STZ activation is usually assisted by free volume [2,6,29], we generalize this onset condition to involve the configurational (or free volume) probability, $F_\xi$, of the activation of STZs that is statistically related to the free volume by [8,32]

$$F_\xi = \exp\left(-1/\xi\right), \tag{2}$$

where the free volume concentration $\xi$ is defined as the active free volume, $v_{\mathrm{f}}^{\mathrm{a}}$, normalized by the critical volume $\chi\bar{v}_0$ for flow, here $\chi$ is a material-specific constant, and $\bar{v}_0$ is the average atomic volume. Thus, metallic glasses will flow when the following condition is satisfied [19,33]:

$$\gamma_0\omega_0 \exp\left(-\frac{W_\tau}{k_{\mathrm{B}}T}\right) F_\xi = C\dot{\gamma}, \tag{3}$$

where $\gamma_0$ is the characteristic shear strain of an STZ, $\omega_0$ is an attempt frequency, $W_\tau$ is the activation energy for an STZ at finite shear stress, $k_{\mathrm{B}}$ is the Boltzmann constant, $C$ is a dimensionless constant of order unity, and $\dot{\gamma}$ is the applied shear strain rate. Following Johnson and Samwer [31], the activation energy for an STZ to flow in a stressed metallic glass can be written as

$$W_\tau = 4R\zeta G_T\gamma_{CT}^2\left(1-\frac{\tau_{CT}}{\tau_{C0}}\right)^{3/2}\Omega. \tag{4}$$

In this equation, $R$ is the "fold ratio", $\zeta$ is a correction factor arising from the matrix confinement. $G_T$ is the temperature-dependent shear modulus that can be obtained by [34]

$$G_T = G_0 - \frac{s}{\exp\left(\theta_{\mathrm{D}}/T\right)-1}, \tag{5}$$

where $G_0$ is the athermal shear modulus, $\theta_{\mathrm{D}}$ is the Debye temperature, $s$ is a fitting parameter that can be determined as $s = 0.15G_0\left[\exp\left(\theta_{\mathrm{D}}/T_{\mathrm{g}}\right)-1\right]$ for metallic glasses. The temperature-dependent yield strain $\gamma_{CT} = \gamma_{C0} - \gamma_{C1}\left(T/T_{\mathrm{g}}\right)^{2/3}$, amounts here to $\gamma_{C0} = 0.036$ and $\gamma_{C1} = 0.016$ [31]. The applied shear stress at yielding $\tau_{CT} = 2G_T\gamma_{CT}/\pi$ where the nonlinear elastic response of an STZ is considered as $\tau_{CT}$ increases from 0 to $\tau_{C0} = G_0\gamma_{C0}$. Using Eqs. (2)–(5), the STZ volume can be derived as [19–21]:

$$\Omega = \frac{k_{\mathrm{B}}T}{4R\zeta\Phi\left(T\right)}\left(\ln\frac{\omega_0}{C\dot{\gamma}} - \frac{1}{\xi}\right), \tag{6}$$

where $\Phi(T) = G_T\left[\gamma_{C0}-\gamma_{C1}\left(T/T_{\mathrm{g}}\right)^{2/3}\right]^2\left\{1-\left[2G_T/\left(\pi G_0\right)\right]\left[1-\left(\gamma_{C1}/\gamma_{C0}\right)\left(T/T_{\mathrm{g}}\right)^{2/3}\right]\right\}^{3/2}$. It can be found from Eq. (6) that the STZ volume mainly depends on three factors: (i) the environmental temperature, (ii) the applied loading rate, and (iii) the active free volume within material. In the following, we only focus on the effect of the environmental temperature.

As for the temperature dependence of active free volume, we borrow from a result of the simulation by Starr et al. [35]. It is suggested that the evolution of the active free volume with STZ operations at $T < T_{\mathrm{g}}$ can be well approximated by a power law

$$v_{\mathrm{f}}^{\mathrm{a}}\left(T\right) = \Lambda\left(\frac{T}{T_0}\right)^{\eta}, \tag{7}$$

where the fitting parameters are chosen as $\Lambda = \chi\bar{v}_0, T_0 = 2.73\times10^5$ K and $\eta = 0.338$. Eq. (7) then indicates that the active free volume is about $0.1\chi\bar{v}_0$ at room temperature, a typical value for metallic glasses [7,36]. Additionally, Eq. (7) predicts that the active free volume will remain larger than zero at all temperatures and only vanishes at absolute zero temperature. This behavior agrees with the extended model of the free volume by Cohen and Grest [37] and the Adam–Gibbs entropy model for viscosity that is well defined for all temperatures [38]. Combining Eq. (6) with Eq. (7) indicates that the STZ dilatational strain (1) is also a function of the environmental temperature.

Next, we examine the effect of the environmental temperature on STZ volume (6), active free volume (7), and STZ dilatational strain (1), by using Vitreloy 1 as a model material. The relevant mechanical and physical parameters of Vitreloy 1 are derived from the recent literature [3,8,29,31,39–41] and listed in Table 1. By combining Eqs. (6) and (7), the evolution of active free volume and STZ volume with temperature is calculated in Fig. 1. The calculated free volume and STZ volume are, respectively, comparable to the experimentally determined values [39,42,43]. It can be seen that both STZ volume and STZ-activated free volume decrease monotonously and approach zero with decreasing temperature. This result implies that low temperatures render plastic flow more difficult. It is