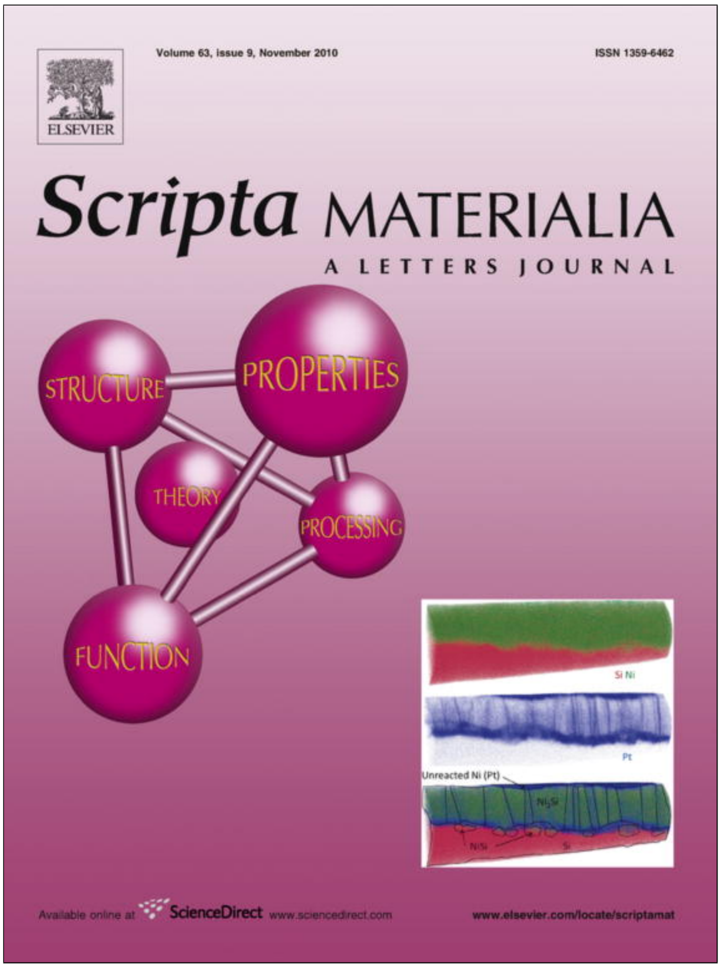
Volume 63, issue 9, November 2010
ISSN 1359-6462
ELSEVIER
Scripta MATERIALIA
A LETTERS JOURNAL
STRUCTURE
PROPERTIES
THEORY
PROCESSING
FUNCTION
Unreacted Ni (Pt)
Available online at ScienceDirect www.sciencedirect.com
www.elsevier.com/locate/scriptamat

Available online at www.sciencedirect.com
ScienceDirect
Scripta Materialia 63 (2010) 945–948
Scripta MATERIALIA
www.elsevier.com/locate/scriptamat

Intrinsic correlation between dilatation and pressure sensitivity of plastic flow in metallic glasses

L. Sun,[a] M.Q. Jiang[a] and L.H. Dai[a,b,*]

[a]State Key Laboratory of Nonlinear Mechanics, Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190, People's Republic of China

[b]State Key Laboratory of Explosion Science and Technology, Beijing Institute of Technology, Beijing 100081, People's Republic of China

Received 1 June 2010; revised 3 July 2010; accepted 7 July 2010
Available online 24 July 2010

Taking shear-induced dilatation into consideration in shear transformation zone (STZ) operations, we derive a new yield criterion that reflects the pressure sensitivity in plastic flow in metallic glasses (MGs), which agrees well with experiments. Furthermore, an intrinsic theoretical correlation between the pressure sensitivity coefficient and the dilatation factor is revealed. It is found that the pressure sensitivity of plastic flow of MGs originates in the dilatation of microscale STZs.

Keywords: Metallic glasses; Shear transformation zone; Dilatation; Pressure sensitivity

The pressure dependence of plastic deformation in metallic glasses (MGs) has been a topic of active research for over a decade [1–7], since it reflects the basic flow mechanism, which differs from that of their crystalline counterparts. Although the precise physical picture of how this dependence arises from the internal structure of MGs remains elusive, it is plausible that it originates from atomic-scale dilatation [8,9]. In crystalline solids, unit glide of a dislocation, which is the basic unit of plasticity, does not require significant dilatation; hence, plasticity involves only deviatoric stress. However, the fundamental carriers of plastic deformation in MGs are shear transformation zones (STZ) or flow defects [10–12], i.e. a local cluster of atoms undergoing an inelastic shear distortion to produce a local shear strain. Due to the long-range disorder structure, MGs—unlike crystalline solids—cannot find a slip plane when they experience shear deformation. As a result, STZs change into a loose configuration with a larger volume, resulting in dilatation [13,14]. Such dilatation results in hydrostatic stress during STZ formation, and thus the resultant macroscopic plastic flow should depend on pressure or normal stress. However, the precise correlation between dilatation during STZ formation and the pressure sensitivity of macroscopic plastic flow in MGs is unclear. In this paper, we present a theoretical derivation of the intrinsic correlation between pressure sensitivity and dilatation; the underpinning physics is briefly discussed as well.

There is a general consensus that the behavior of STZs in MGs can be treated as an Eshelby-type inclusion problem [10,15]. In this case, the STZ operation takes place within the elastic confinement of a surrounding glass matrix. Since there is a shear-induced dilatation during shear transformation, the STZ would shove aside the surrounding atoms [14], making the operation of the STZ harder. In order to highlight the essential physics, the initial configuration of a STZ is assumed to be a sphere of radius R. If the origin of coordinates (x, y, z) lies at the centre of sphere, the initial configuration of a STZ can be described by $x^2 + y^2 + z^2 = R^2$, as shown in Figure 1a. After a shape distortion, the configuration of the STZ changes into an ellipsoid described by $x^2/\left[(1+\beta)^2R^2\right] + y^2/\left[R^2(1-\beta)^2\right] + z^2/R^2 = 1$, as shown in Figure 1b. At the same time, an accompanied bulk dilatation takes place, as shown in Figure 1c. Thus, the final configuration of the STZ can be described by $x^2/\left[(1+\alpha)^2(1+\beta)^2R^2\right] + y^2/\left[(1+\alpha)^2R^2(1-\beta)^2\right] +$

* Corresponding author at: State Key Laboratory of Nonlinear Mechanics, Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190, People's Republic of China. Tel.: +86 10 82543958; fax: +86 10 82543977; e-mail: lhdai@lnm.imech.ac.cn

1359-6462/$ - see front matter © 2010 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.
doi:10.1016/j.scriptamat.2010.07.011

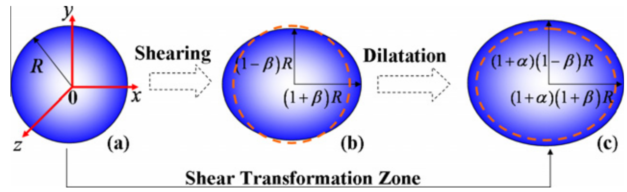

Figure 1. Schematics of Configuration change of an STZ operation.

$z^2/\left[(1+\alpha)^2R^2\right]=1$. It is then easy to obtain that the shear strain is 2β and volume strain is 3α during such a STZ operation. The relationship between shear-induced volume strain and shear strain is assumed to be linear [16], $3\alpha=c(2\beta)$, where c is the dilation coefficient measuring the ratio of dilatation to shear strain.

In order to make STZ produce such a shear and accompanying bulk deformation, the activation free energy ΔG is given by $\Delta G(\alpha,\beta)=\Delta F(\alpha,\beta)-W(\alpha,\beta)$, where ΔF represents the energy required to deform a STZ embedded in the matrix, and W is the work of applied stress at distant boundaries. $\Delta F(\alpha,\beta)$ can be expressed as the sum of a distortional part $\Delta F_s(\beta)$ and a dilatational part $\Delta F_d(\alpha)$. Prior to calculating the energy required to deform a STZ, the potential energy landscape of a STZ must be known. Analogous to the case of close-packed atoms, the shear resistance of a free STZ via shear strain can be assumed to be a form of sinusoidal curve [10,15]. Furthermore, for ease of operation, we replace it with:

$$\tau=\begin{cases}2\mu'\beta & \beta\leqslant\beta_C\\ 2\mu'(2\beta_C-\beta) & \beta_C<\beta\leqslant 3\beta_C\\ 2\mu'(\beta-4\beta_C) & 3\beta_C\leqslant\beta\leqslant 4\beta_C\end{cases}\quad\left(\beta_c=\frac{\hat{\tau}}{2\mu'}\right),\tag{1}$$

where $\hat{\tau}$ is the barrier shear resistance of a STZ and μ' has the mean of shear modulus of a STZ. Then, the energy necessary to shear a free STZ by an increment of shear strain is $\Delta\phi_s=2\mu'\Omega_0\left(4\beta\beta_c-2\beta_c^2-\beta^2-\beta_0^2\right)$, where Ω_0 is the volume of a STZ, and $\beta_0=\left(\sigma_1^0-\sigma_3^0\right)/4\mu'$ represents the initial deformation of the STZ before shear transformation takes place with the principal stress components $(\sigma_1^0,\sigma_2^0,\sigma_3^0)$ of applied stress σ_{ij}^0. Since the STZ is constrained by the matrix, the energy caused by such a shear deformation is $\Delta F_s=\xi_s\Delta\phi$, where $\xi_s=15(1-v)/(7-5v)$ is the Eshelby factor for pure shear deformation [17,18], and v is the Poisson's ratio. As for the dilatation part, the potential energy landscape is assumed to be $\Delta\phi_d=9/2K\alpha^2\Omega_0$, the same as the elastic deformation, and K is the bulk modulus. Considering the elastic constraint of the matrix, the potential energy change associated with dilatation is $\Delta F_d=9/2K\alpha^2\Omega_0\xi_d$, where $\xi_d=1.5(1-v)/(1-2v)$ is the Eshelby factor for a pure dilatation [18,19]. As given by Eshelby [17], the work of the applied stress σ_{ij}^0 at the boundaries is $W(\alpha,\beta)=2\tau(\beta-\beta_0)\xi_s+2pc(\beta-\beta_0)\xi_d$, where $\tau=(\sigma_1^0-\sigma_3^0)/2$is the maximum shear stress, and $p=(\sigma_1^0+\sigma_2^0+\sigma_3^0)/3$ is the hydrostatic stress. Thus, we obtain the whole activation free energy for such an STZ operation:

$$\Delta G(\beta)/\Omega_0=-A\mu'(\beta-\beta_0)^2+B(\beta-\beta_0)+D\mu',\tag{2}$$

where $A=2\xi_s-4\xi_dc^2(1+v)/[3(1-2v)]$, $B=-2c\xi_dp+4\xi_s(\hat{\tau}-\tau)$, and $D=2\xi_s\left(4\beta_0\beta_c-2\beta_c^2-2\beta_0^2\right)$. The free energy barrier for deforming an STZ is obtained by $\partial\Delta G_u(\beta)/\partial\beta=0$. Thus we obtain the energy barrier:

$$\Delta G_u(\beta)/\Omega_0=\left(4\xi_s^2/A-\xi_s\right)(\hat{\tau}-\tau-Qp)^2/\mu',\tag{3a}$$

with

$$Q=3c/\left[30(1-2v)/(7-5v)+2(1+v)c^2/(1-2v)\right].\tag{3b}$$

Note that a term with c^2, which makes it much less than other terms for $c^2\ll1$, is neglected.

It has been recognized that macroscopic flow of MGs occurs as a result of a series of STZ operations as described above. In other words, yielding occurs when the applied activation energy causes a critical density of STZs to become unstable [15,20,21]. According to the cooperative shear model (CSM) proposed by Johnson and Samwer [15], only when the barrier crossing rate of STZs reaches a critical value comparable to the applied strain rate, $\dot{\gamma}$, does plastic deformation takes place. This yields:

$$\omega_0\ \exp\left(-\Delta G_u/k_BT\right)=\vartheta\dot{\gamma},\tag{4}$$

where ω_0 is the attempt frequency, k_B is the Boltzmann constant, T is the temperature, and ϑ is a dimensionless constant of order unity. Then, substituting $\Delta G_u$with Eq. (3) into Eq. (4), one obtains:

$$\tau+Qp=k,\tag{5a}$$

with

$$k=\hat{\tau}-\left[\frac{1}{\Omega_0}\mu'k_BT\ \ln\left(\omega_0/\vartheta\dot{\gamma}\right)/\left(4\xi_s^2/A-\xi_s\right)\right]^{1/2}.\tag{5b}$$

Eq. (5) is a pressure-dependent criterion for the plastic yield of MGs. The coefficient Q, comparable to the friction coefficient in the conventional Mohr–Coulomb criterion, describes the pressure sensitivity, and k represents the strength threshold of yielding. Both Q and k involved in Eq. (5) are endowed with clear physical meaning: they are correlated with microscopic STZ operations. We find that the pressure sensitivity coefficient Q is a function of dilatation coefficient c and Poisson's ratio v. The strength k depends on not only the material parameters such as μ', Ω_0 and v, but also ambient temperature T and applied strain rate $\dot{\gamma}$. Here, $\hat{\tau}$ is the yield stress at absolute zero temperature, and the second term on the right-hand side of Eq. (5b) is the softening of strength induced by thermal assistance in the STZ operation. The temperature dependence of strength is described by a power law with an exponent 1/2, i.e. the same as that of Schuh et al. [9]. However, the temperature dependence of strength is relatively weak, and hence a slightly different exponent of 2/3 derived by Johnson and Samwer [15] was also found to capture experimental data well. Furthermore, it is noted that both the enhanced effect of applied strain rates and the weakened effect of dilatation are involved in k.

Obviously, our criterion indicates that plastic yielding in MGs relies on the maximum shear stress, as well as hydrostatic pressure. To verify the criterion, shear strength as a function of hydrostatic stress over a wide range compiled in Figure 2 with data collected from previous works [1,4,22,23] for $Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$ (Vit.1). Eq. (5) describes the experiment data quite well while $Q = 0.158$ and $k = 823.2$ MPa. The pressure sensitivity coefficient 0.158 is very close to 0.12 fitted from indentation data [24]. And the pure shear strength $k = 823.2$ MPa is a little smaller than experiment value 1.03 GPa [22]. The reason is that the real experiment can hardly be in a pure shear state without any constraint in the shear plane. In this plot, two broken lines represent uniaxial compression (left) and uniaxial tension (right), respectively. Intersecting points between the solid line and broken lines, which represent uniaxial compression and uniaxial tension strength, also show that strength of compression is larger than that of tension. Furthermore, according to Eq. (3b), we can obtain the dilatation coefficient $c = 0.093$ for Vit.1, here taking Poisson's ratio $\nu = 0.352$[5,25]. The calculated value is in agreement with our previous analysis. Based on the systematic study on the correlation between fragility and elastic modulus for MGs, we revealed that, in a real flow event, the shear-induced dilatation strain is about 10% of shear yield strain [13].

The Eq. (3b) clearly indicates the relationship between the pressure sensitivity Q and the dilatation coefficient $c$with Poisson's ratio ν involved. If there is no dilatation in our microscopic model, that is, $c = 0$, the pressure dependency of plastic flow vanishes. In this case, Eq. (5) becomes the known Tresca criterion that describes plastic yield of crystalline solids. For a given Poisson's ratioν, the pressure sensitivity Q increases significantly with increasing the dilatation coefficient$c$, which is shown in Figure 3. Physically, the higher c, which means high volume dilatation when shear strain is the same, could make hydrostatic stress p do more work (see the expression of $W(\alpha, \beta)$). As a result, p plays a more important role in overcoming energy barrier of STZs. In other words, the plastic flow of the MG becomes more sensitive to pressure. From above analysis, one can conclude that it is the microscopic dilatation that causes pressure dependency of macroscopic plastic flow. And in other pressure-sensitive materials, such as granular materials, Massoudi et al [26] and Nemat-Nasser [27] have derived a correlation between friction angle in Mohr-Coulomb criterion and dilatation angle by analyzing the sliding and rolling of grains on a shear plane.

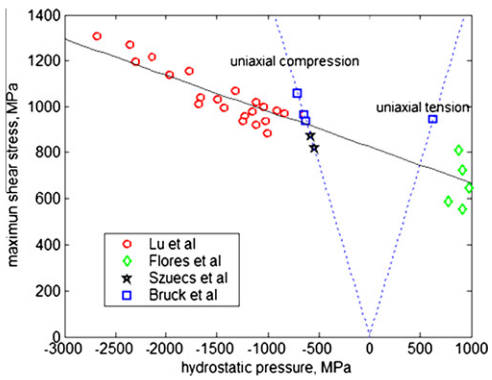

Figure 2. The maximum shear stress as function of hydrostatic pressure for Vit.1 with collected experiment data [1,4,22,23].

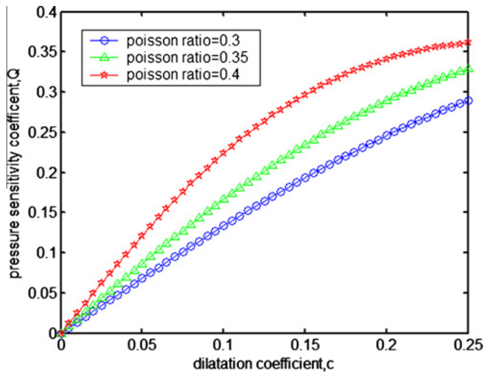

Figure 3. Relationship between pressure sensitivity coefficient and dilatation coefficient with different Poisson's ratio.

Their calculations also indicate that pressure dependency is resulted from dilatation in granular materials. In addition, it is noted from Eq. (3b) that Poisson's ratio ν also have an effect on pressure sensitivity coefficient Q. However, there is no clear trend between them for various MGs, since the dilatation coefficient is highly material-dependent. Our prediction was confirmed by the recent experimental data presented by Baricco et al. [28].

The intrinsic correlation of pressure sensitivity with the dilatation could be understood further within the content of the free volume. Recently, Dubach et al [29] have performed systematic indentation experiments on a Zr-based BMG with different structural states to examine the pressure sensitivity of plastic flow. They revealed that structural relaxed BMGs vis-à-vis as-cast samples have more enhanced pressure sensitivity. It is well known that atomic disorder or free volume within BMGs is reduced during structural relaxation [30,31]. In such surroundings with lower free-volume content, the STZ operations become more difficult, because it requires more significant dilatation of the surrounding matrix. According our foregoing analysis, such atomic-scale dilatation would lead to pressure dependence of macroscopic plastic flow in BMGs. Therefore, availability of free volume is important for the STZs to operate in a given volume of BMGs [29]. In fact, STZ operations occur preferentially in those regions being higher free volume as relatively less dilatation is required [10]. In the opposite case, that is, free volume goes zero, STZs could be restrained and tension transformation zones (TTZs) could be activated as they can be regarded as the counterpart of STZs suffering an extreme dilatation but only slight shearing [31–33].

In summary, the energy barrier for a STZ operation which takes shear-induced dilatation into account is derived by treating STZ as an Eshelby inclusion. And then following the physical picture of CSM, we obtain a yield criterion theoretically, which describes the pressure dependence of plastic flow of MGs quite well. In particular, the explicit relationship between pressure sensitivity coefficient and dilatation is obtained. Its validity is briefly discussed in the context of free volume.

Financial support is from the NSFC (Grants Nos. 10725211 and 10721202), the NSAF (Grant No. 10976100), the National Basic Research Program of

China (Grant No. 2009CB724401) and the Key Project of Chinese Academy of Sciences (No. KJCX2-YW-M04).

[1] K.M. Flores, R.H. Dauskardt, Acta Mater. 49 (2001) 2527.
[2] V. Keryvin, K.E. Prasad, Y. Gueguen, J.C. Sangleboeuf, U. Ramamurty, Philos. Mag. 88 (2008) 1773.
[3] C.A. Schuh, A.C. Lund, Nat. Mater. 2 (2003) 449.
[4] J. Lu, G. Ravichandran, J. Mater. Res. 18 (2003) 2039.
[5] M.Q. Jiang, L.H. Dai, J. Mech. Phys. Solids 57 (2009) 1267.
[6] P. Lowhaphandu, S.L. Montgomery, J.J. Lewandowski, Scripta Mater. 41 (1999) 19.
[7] R.T. Ott, F. Sansoz, T. Jiao, D. Warner, C. Fan, J.F. Molinari, K.T. Ramesh, T.C. Hufnagel, Metall. Trans. A 37 (2006) 3251.
[8] K.E. Prasad, R. Raghavan, U. Ramamurty, Scripta Mater. 57 (2007) 121.
[9] C.A. Schuh, T.C. Hufnagel, U. Ramamurty, Acta Mater. 55 (2007) 4067.
[10] A.S. Argon, Acta Metall. 27 (1979) 47.
[11] M.L. Falk, J.S. Langer, Phys. Rev. E 57 (1998) 7192.
[12] F. Spaepen, Scripta Mater. 54 (2006) 363.
[13] M.Q. Jiang, L.H. Dai, Phys. Rev. B 76 (2007) 054204.
[14] J.C. Dyre, N.B. Olsen, T. Christensen, Phys. Rev. B 53 (1996) 2171.
[15] W.L. Johnson, K. Samwer, Phys. Rev. Lett. 95 (2005) 195501.
[16] A.S. Argon, J. Megusar, N.J. Grant, Scripta Metall. 19 (1985) 591.
[17] J.D. Eshelby, Proc. Roy. Soc. Lond. A 241 (1957) 376.
[18] T. Mura, Micromechanics of Defects in Solids, Martinus Nijhoff Publishers, London, 1982.
[19] T. Egami, D. Srolovitz, J. Phys. F: Met. Phys. 12 (1982) 2141.
[20] D. Pan, A. Inoue, T. Sakurai, M.W. Chen, PNAS 105 (2008) 14769.
[21] Y.H. Liu, K. Wang, A. Inoue, T. Sakurai, M.W. Chen, Scripta Mater. 62 (2010) 586.
[22] H.A. Bruck, T. Christman, A.J. Rosakis, W.L. Johnson, Scripta Metall. 30 (1994) 429.
[23] F. Szuecs, C.P. Kim, W.L. Johnson, Acta Mater. 49 (2001) 1507.
[24] C.E. Packard, C.A. Schuh, Acta Mater. 55 (2007) 5348.
[25] M.Q. Jiang, W.H. Wang, L.H. Dai, Scripta Mater. 60 (2009) 1004.
[26] M. Massoudi, M.M. Mehrabadi, Acta Mech. 152 (2001) 121.
[27] S. Nemat-Nasser, J. Mech. Phys. Solids 48 (2000) 1541.
[28] M. Baricco, T.A. Baser, J. Das, J. Eckert, J. Alloy. Compd. 483 (2009) 125.
[29] A. Dubach, K.E. Prasad, R. Raghavan, J.F. Loffler, J. Michler, U. Ramamurty, J. Mater. Res. 24 (2009) 2697.
[30] P. Murah, U. Ramamurty, Acta Mater. 53 (2005) 1467.
[31] R. Raghavan, P. Murali, U. Ramamurty, Acta Mater. 57 (2009) 3332.
[32] M.Q. Jiang, Z. Ling, J.X. Meng, L.H. Dai, Philos. Mag. 88 (2008) 407.
[33] J.P. Escobedo, Y.M. Gupta, J. Appl. Phys. 107 (2010) 123502.