

On bifurcation of homogenous deformation in metallic glasses

Yan Chen & Lanhong Dai*

State Key Laboratory of Nonlinear Mechanics, Institute of Mechanics,
Chinese Academy of Science, Beijing, 100190, China

* Corresponding author: lhdai@lnm.imech.ac.cn

Abstract At temperatures well below the glass transition and at high stresses, the homogenous deformation in metallic glasses (MGs) usually develops to a critical point, at which the discontinuity in deformation rate is incipient across nano-scale shear bands. However, such a bifurcation condition of homogeneous deformation concerning the unique properties of MGs is still lacking for general stress state. In this paper, a new constitutive is introduced for MGs accounting for the pressure-sensitivity, dilatancy and structural evolution; the shear banding is regarded as the appearance of instability in the constitutive description of inelastic deformation. Tying the bifurcation theory to the new model, the general condition of deformation localization is derived. The shear band initiation and failure orientation are then precisely predicted for MGs by constructing a bridge between the microscopic origin and the loss-of-ellipticity instability in the constitutive law in continuum mechanics.

Keywords Metallic glasses, Bifurcation, Shear band

1. Introduction

Metallic glass (MG) represents a relatively young class of structural materials with a combination of excellent properties. Due to their random atomic structure, plastic deformation in MGs is usually accommodated by localized shear bands. These nanoscale shear bands as a precursor of crack lead to a fast fracture of material, which presents little plasticity at room temperature. Being a key process to understand the underlying plasticity of MGs, shear banding, including its origin and propagation, has attracted lots of attention for last decades[1-5].

Although the precise physical picture of how it originates from the internal structure remains elusive, it is well accepted that the shear banding of MGs occurs as a consequence of formations and self-organizations of shear plastic flow events [3, 6-8]. Those flow events are essentially local arrangements of atoms around free volume sites, termed shear transformation zones (STZs) or flow defects [9-12]. The transition from local plastic events to macroscopic shear-band instability is dominated by the stress-driven free volume softening and assisted by thermal softening [1, 2, 13-15]. Regardless of micro-mechanisms, at the continuum level, the shear banding, a physically material unstable event, can be regarded as the appearance of instability in the macroscopic constitutive description of inelastic deformation [16-19]. It is accepted that the homogenous deformation in MGs develops to a critical point, at which the discontinuity in deformation rate is incipient across nano-scale shear bands, at temperatures well below the glass transition and at high stresses. Tying the bifurcation theory to a pressure-sensitive dilatant constitutive model, Rudnicki and Rice [17] derived both the general conditions for shear localization and the orientation of shear band in the stress space. Based on this, the shear band direction as an important feature in shear band formation was predicted for MGs by Gao et al. [20], which closely depends on the pressure coefficient, the dilatancy factor, and etc. In nature, shear banding as a material instability is greatly correlated to atomic structural change. Such a picture is also realized by Ruan et al. [21] who embody the atomic structural change by the plastic strain and the associated dilatation for MGs. On the other hand, through perturbation analysis of constitutive instability, the conditions for the shear instability for a given stress state (usually simple shear) can be also obtained for MGs [1, 2, 13]. These analyses confirm the important role of free volume, as a state variable, in the constitutive instability. However, such a bifurcation condition of homogeneous deformation concerning the unique

properties of MGs is still lacking for general stress state. In this paper, we attempt to derive the critical condition of the shear band initiation and direction for MGs by constructing a bridge between the microscopic origin and the loss-of-ellipticity instability in the constitutive law in continuum mechanics.

2. Theoretical model

For initial homogeneously deformed material, the loss of constitutive stability will cause strain localization into a shear band [16-19]. Therein, the homogeneous deformation develops to a bifurcation point, at which the discontinuity in deformation rate is incipient across a band. As to MGs, two internal factors are critical for the shear instability, namely, free volume and thermo [1, 2, 13-15]. The onset of shear banding in MGs can be reasonably described as a result of constitutive instability induced by dramatic change of internal state variables.

2.1 Constitutive model

For the instability analysis, a proper constitutive is required for MGs. In this section, a new constitutive model is developed for MGs due to the following considerations. At macroscopic scale, MGs exhibit inherent pressure sensitivity and shear-dilatancy during plastic deformation, which usually renders a non-associated flow. Microscopically, the nucleation and coalescence of free volume decreases the flow stress of MGs and further leads to shear localization. This is actually quite similar to the void evolution mechanism in the continuum damage mechanics. One of the best known micro-mechanical models is that of Gurson [22], who studied the plastic flow of a void-containing material and established a yielding function reflecting the softening effect due to the presence of voids. Here, by introducing the free volume evolution into the framework of continuum mechanics, we can establish a new constitutive model to comprehensively and satisfactorily describe the deformation in MGs. “Free volume” as the topological disorder in MGs, can be simply considered as randomly distributed atomic voids in material. Treating those voids to be spherical, the yield function of Gurson is reasonably extended to MGs by taking into the pressure sensitivity of the matrix. The free volume evolution is assumed to obey the self-consistent dynamic free volume model proposed by Johnson et al.[23]. The thermal effect is neglected since the structural disorder induced softening precedes thermal softening at the origin of the shear banding [13], especially for the present quasi-static loading case.

For a pressure-independent material containing spherical voids, Gurson model gives the following yield function [22]:

$$F = \left(\frac{\sigma_e}{\sigma_y} \right)^2 + 2\nu_f \cosh\left(\frac{3\sigma_m}{2\sigma_y} \right) - \nu_f^2 - 1 = 0, \quad (1)$$

where $\sigma_e = \sqrt{3s_{ij}s_{ij}/2} = \sqrt{3J_2}$ ($s_{ij} = \sigma_{ij} - \sigma_m\delta_{ij}$) is the effective stress, $\sigma_m = \sigma_{ii}/3$ is the mean stress, ν_f is the current fraction of voids, equivalent to free volume concentration, and σ_y is the yield stress of the matrix. Numerous studies have demonstrated clearly that pressure affects the yield behavior of MGs [24-27]. This is easily reflected by the tension-compression asymmetry of failure [28]. Since the Gurson model considers the von Mises matrix, an additional pressure-dependent term i.e. $\alpha\sigma_m$, should be taken into account (analogous to the Drucker-Prager criterion) for a proper description of MGs. For simplicity, one can modify the above criterion as below by neglecting the minor term $(1 - \cosh^2(3\sigma_m/2\sigma_y))\nu_f^2$. We define the initial free volume in

MGs to be ν_0 . The corresponding shear strength σ_0 satisfies $\sigma_0 = \sigma_y (1 - \nu_0 \cosh(3\sigma_m/2\sigma_y)) / \sqrt{3}$. Therefore, the new yield criterion can be obtained from Eq.(1) as

$$\Phi = \sqrt{J_2} + \alpha\sigma_m - \sigma_0 \left(1 - \frac{\Lambda}{\operatorname{sech}(3\sigma_m/2\sigma_y) - \nu_0} \right), \quad (2)$$

where $\Lambda = \nu_f - \nu_0$ is the free volume increment. It is obvious that the shear strength is softening with increasing free volume concentration.

Following the self-consistent dynamic free volume model [23], the local time rate of change of the free-volume concentration is

$$\dot{\nu}_f = \chi \left(R \dot{\epsilon}_e^p - \frac{\Lambda}{\sigma_e / (G \dot{\epsilon}_e^p)} \right), \quad (3)$$

where χ is a material parameter of order unity, R is a free-volume creation function defining the free volume produced by a unit shear strain and is given by GA/σ_{ss} (σ_{ss} is the effective stress at steady state) [29], $\dot{\epsilon}_e^p = \sqrt{2\dot{\epsilon}_{ij}^p \dot{\epsilon}_{ij}^p / 3}$ ($\dot{\epsilon}_{ij}^p = \dot{\epsilon}_{ij}^p - \dot{\epsilon}_{kk}^p \delta_{ij} / 3$) is the effective plastic shear strain rate, σ_e is the effective stress, and G is the shear modulus at room temperature.

Considering the shear dilatancy inherent in the deformation of MGs, we introduce the dilatancy factor β and invoke Q as the plastic potential as below

$$Q = \sqrt{J_2} + \beta\sigma_m - \sigma_0 \left(1 - \frac{\Lambda}{\operatorname{sech}(3\sigma_m/2\sigma_y) - \nu_0} \right). \quad (4)$$

When $\beta \neq \alpha$, the deformation is non-associative. The deviatoric components of the plastic deformation strain tensor are obtained as

$$\dot{\epsilon}_{ij}^p = \frac{1}{H} \frac{\partial Q}{\partial \sigma_{ij}} \frac{\partial \Phi}{\partial \sigma_{kl}} \dot{\sigma}_{kl}, \quad (5)$$

where H is the plastic hardening modulus.

If we adopt the spineless strain rate, the generalized constitutive relation is recast as

$$D_{ij} = \frac{s_{ij}^\nabla}{2G} + \frac{s_{ij}}{2H\sqrt{J_2}} \left(\frac{s_{kl}}{2\sqrt{J_2}} \sigma_{kl}^\nabla + \frac{\alpha'}{3} \sigma_{kk}^\nabla \right) + \frac{\beta'}{3H} \left(\frac{s_{kl}}{2\sqrt{J_2}} \sigma_{kl}^\nabla + \frac{\alpha'}{3} \sigma_{kk}^\nabla \right) \delta_{ij} + \frac{\sigma_{kk}^\nabla}{9K} \delta_{ij}, \quad (6)$$

where $\sigma_{ij}^\nabla = \dot{\sigma}_{ij} + \Omega_{ik} \sigma_{kj} + \Omega_{ki} \sigma_{jk}$ is the Jaumann rate of the true stress σ_{ij} , $\alpha' = \alpha + \sqrt{3}\nu_f \sinh(3\sigma_m/2\sigma_y)/2$, $\beta' = \beta + \sqrt{3}\nu_f \sinh(3\sigma_m/2\sigma_y)/2$, and

$$H = \frac{\sigma_0 \chi GA}{\sqrt{3}(\operatorname{sech}(3\sigma_m/2\sigma_y) - \nu_0)} \left(\frac{1}{\sqrt{3J_2}} - \frac{1}{\sigma_{ss}} \right).$$

The instantaneous rate of the deformation and the spin tensor are respectively $D_{ij} = (\partial v_i / \partial x_j + \partial v_j / \partial x_i) / 2$, and $\Omega_{ij} = (\partial v_i / \partial x_j - \partial v_j / \partial x_i) / 2$. The stress rate and deformation rate are related by

$$\sigma_{ij}^\nabla = L_{ijkl} D_{kl}, \quad (7)$$

where L_{ijkl} is the elasto-plastic modulus tensor.

2.2 Constitutive instability analysis

We ascribe the initiation of shear band in MGs to bifurcation arisen from the constitutive description of the homogeneous deformation. The shear bands inclination is obtained at the onset of instability. An initially uniform deformation field of a homogeneous MG plate is considered, see Fig. 1. The constitutive instability is viewed as a formation of a narrow localized band of deformation within the plate under external load [17]. Use rectangular Cartesian coordinates (x_1, x_2, x_3) , such that the x_2 -direction is normal to the planes bounding the band. Outside the band the velocity field remains uniform and within it varies only in the direction normal to the band. Thus, the non-uniformities in the rate of deformation field are expressed as

$$\Delta(\partial v_i / \partial x_j) = g_i(x_2) \delta_{j2}, \quad (i, j) = 1, 2, 3, \quad (8)$$

where v_i is a velocity component, Δ denotes the difference between the local field inside the band and the uniform field outside, and the functions g_i of x_2 are nonzero only within the band.

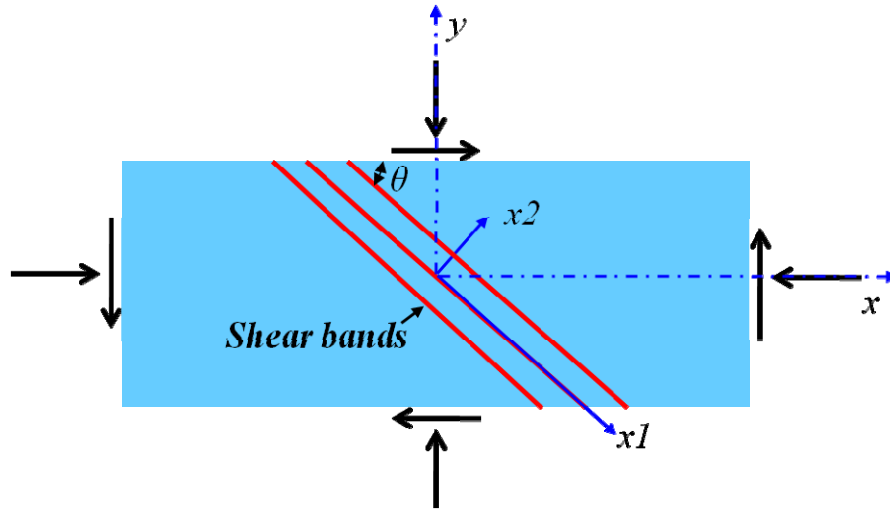


Fig.1 Illustration of shear band localization.

The condition $\partial \sigma_{ij} / \partial x_i = 0$ and $\partial(\partial \sigma_{ij} / \partial t) / \partial x_i = 0$ make sure that the stress equilibrium continues to be satisfied at the inception of bifurcation. The stress rates at incipient localization from the original uniform field is thus following

$$\partial \dot{\sigma}_{ij} / \partial x_i = 0, \quad (9)$$

where the superposed dot denotes its material time rate. The condition of the continuity of the stress rate at the band interfaces can be expressed as

$$\Delta \dot{\sigma}_{2j} = 0, \quad j = 1, 2, 3. \quad (10)$$

Since $\dot{\sigma}_{ij}$ is not invariant under rigid rotations, by introducing the Jaumann (co-rotational) stress rate $\overset{\nabla}{\sigma}_{ij} = \dot{\sigma}_{ij} - \sigma_{ip} \Omega_{pj} - \sigma_{jp} \Omega_{pi}$. Eq. (10) can be regarded as a set of three quasi-homogeneous equations in g_1, g_2, g_3 and the conditions for bifurcation are merely those for which solutions other than $g_1 = g_2 = g_3 = 0$ exist.

If at the bifurcation of deformation-rates, the values L_{ijkl} remain the same inside and outside the band, the following difference can be formed based on Eq. (7):

$$\overset{\nabla}{\Delta \sigma}_{ij} = L_{ijkl} \Delta D_{kl} = L_{ijk2} g_k. \quad (11)$$

Combining Eqs. (10) and (11) yields a set of linear, homogeneous equations in g 's,

$$L_{2jk}g_k = R_{jk}g_k, \quad j=1, 2, 3, \quad (12)$$

The above equation have nontrivial solution when

$$\det|L_{2jk} - R_{jk}| = 0. \quad (13)$$

Eq. (13) represents the condition for the deformation bifurcation when a non-uniform ($g_i \neq 0$) continuation of deformation is possible. The bifurcation condition relates the free volume concentration at localization A_c to the parameters G, K, α, β and to the prevailing stress state. Since the stress to modulus ratio is significantly small, R_{jk} introduced by the co-rotational stress rate are neglected in Eq. (13). We can then obtain the instability condition as below

$$A_c = \left(-\sqrt{B^2 - 4C_1^2 D} - B \right) / 2C_1^2, \quad (14)$$

where $B = \sqrt{3}C_1 \left(\frac{2(\alpha + \beta)}{3} - \frac{s_{22}}{\sqrt{J_2}} \right) - \frac{\chi\sigma_0 C_2}{\sqrt{3}} \left(\frac{4G}{3K} + 1 \right) \left(\frac{1}{\sigma_{ss}} - \frac{1}{\sqrt{3J_2}} \right) + 2\nu_0 C_1^2,$

$$D = \left(\frac{4G}{3K} + 1 \right) \left(1 - \frac{s_{21}^2 + s_{23}^2}{J_2} \right) - \frac{G}{K} \frac{s_{22}^2}{J_2} + (\alpha + \beta) \left(\frac{2\nu_0 C_1}{\sqrt{3}} - \frac{s_{22}}{\sqrt{J_2}} \right) + \frac{4\alpha\beta}{3} - \sqrt{3}\nu_0 C_1 \frac{s_{22}}{\sqrt{J_2}} + \nu_0^2 C_1^2 \quad \text{and}$$

$$C_1 = \sinh(3\sigma_m/2\sigma_y), \quad C_2 = 1/(\cosh^{-1}(3\sigma_m/2\sigma_y) - \nu_0).$$

It means that, shear localization starts when the free volume increment A reaches a critical value A_c given by Eq. (14). A_c is a function of the orientation of the potential plane of localization, indicating it would vary from place to place. At a fixed stress state, the shear instability will first occur at the location or along the direction where the minimum of A_c is required. For the stress state as illustrated in Fig. 1, the preferential direction for shear banding should exist, which satisfies $\partial A_c / \partial \theta = 0$ and the corresponding θ denotes the shear band angle. The hardening modulus H is a function of A . The increase of free volume causes a gradual loss of load-carrying capacity of MGs, corresponding to a decreasing H . The minimal A_c for preferential shear localization is actually consistent with the maximum H first meeting the instability condition as proposed by Rudnicki and Rice [17].

3. Discussion

According to the theoretical model above, the constitutive instability occurs in MGs when the free volume increment A satisfies the condition as described by Eq. (14), which gives birth to shear band in original homogenous materials. The onset of condition and the incipience direction for MGs under pure bending will be specifically discussed next, where both tension and compression cases can be included. The related material properties of Vit-1 MGs and parameters used in the subsequent analysis have been adopted from elsewhere [23, 29, 30]: Shear strength $\sigma_0 = 1GPa$, Young's modulus $E = 96GPa$, Poison ratio $\nu = 0.36$, pressure sensitive factor $\alpha = 0.1$, density $\rho = 6120Kg/m^3$, $\chi = 1.5$, $d_1 = 46981$, $d_2 = 162$, $T_{ref} = 672K$, $T = 300K$, and $\sigma_{ss} = 0.36GPa$.

The critical free volume increment A_c versus θ for $\alpha=0.1$ and different β can be plotted by Fig. 2. Under both tension and compression, the critical A_c is found to reach a minimum at the directions around $\theta = 45^\circ$ (within the green circle), indicating that the free volume required to cause a shear instability is smallest along these directions and thus shear band will preferentially initiate. With a gradual deviation from these angles, the A_c for shear banding increases, denoting a smaller

possibility of shear band formation. As is known, $\theta = 45^\circ$ corresponds to the direction of the maximum shear stress, which is usually the shear direction of crystalline alloys. However, shear bands for MGs are predicted to form at the angles normally $>45^\circ$ in tension while $<45^\circ$ in compression (see Fig. 2), consistent with the experimental observations in MGs [28]. This deviation of shear direction from the maximum shear stress can be partly attributed to the pressure effect which usually cannot be ignored in MGs [24-27]. In addition, the A_c also shows an obvious dependence on dilatancy factor β . With increasing β , the minimum value of A_c decreases in tensile case while increases in compressive case. It means that shear dilatancy promotes the shear banding in tension while impedes that in compression. In Fig. 2, the minimum point of A_c in tension is relatively lower than that in compression. Due to this asymmetry, strain localization in tensile part is usually easier than that in compressive part.

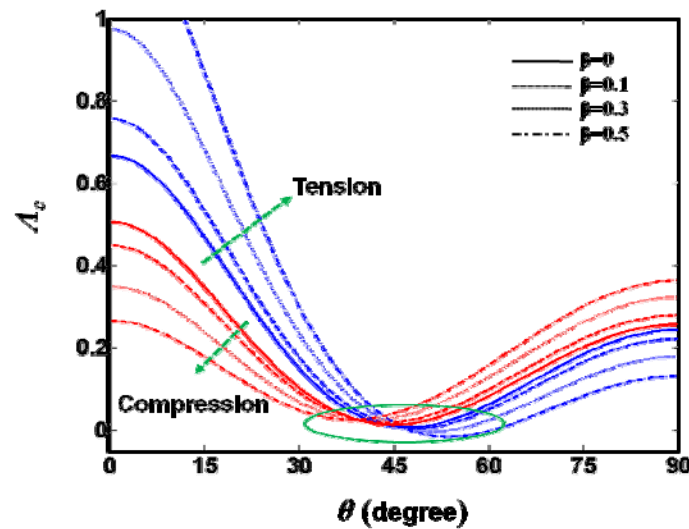


Fig. 2 Dependence of A_c on θ for $\alpha=0.1$ and different β both on tensile side (the blue curves) and on compressive side (the red curves).

The inclination angle of shear banding θ versus the dilatancy factor β for different α is plotted by Fig.3. We interestingly find that θ is a little bigger than 45° for both tension and compression even excluding the pressure sensitivity and dilatancy ($\alpha \rightarrow 0$ and $\beta \rightarrow 0$). This is attributed to the specific stress state and depends on the Poisson ratio [20]. Increasing β or α , the inclination angle increases in tensile part while decreases in compressive part, showing a asymmetrical deviation from 45° . This prediction is similar to those results of uniaxial loading (tension and compression) from continuum and molecular simulations [31, 32]. For Vit-1, the pressure sensitive factor is around 0.1 [33]. Ranging β from 0 to 0.5, the shear band angles are predicted to be $47^\circ \sim 54^\circ$ and $38^\circ \sim 44^\circ$, respectively in tension and compression. Actually, the shear band angles observed in the experiment are 55° on tensile side and 40° on compressive side. In the above discussion, the Poisson ratio ν is set to be a constant 0.36 for Vit-1.

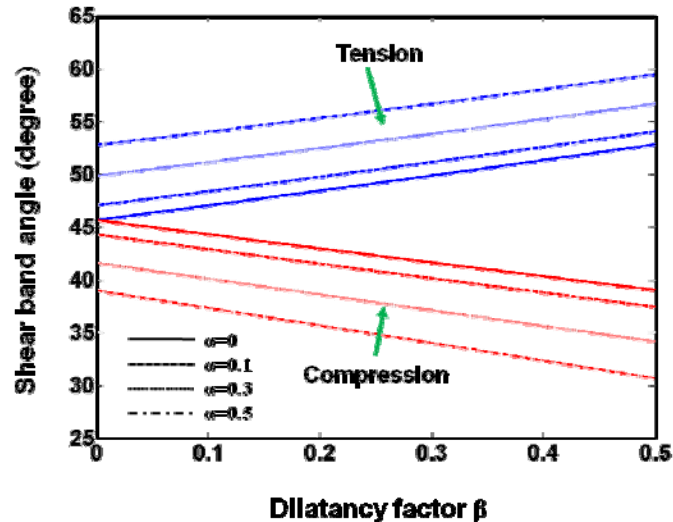


Fig. 3 Dependence of Shear band angle on β for different α both on tensile side (the blue curves) and on compressive side (the red curves).

4. Conclusions

The critical condition for bifurcation of homogenous deformation in MGs has been derived in terms of free volume. It has been found that pressure sensitivity and shear dilatancy exert important effects on the initiation and direction of shear banding. Pressure sensitivity and dilatancy, which are usually coupled together, have the similar trends of influence on the shear band birth and propagation direction. With the enhancement of these two properties, the tension-compression asymmetry of shear banding would be intensified. The instability of constitutive occurs with a smaller \mathcal{A}_c in tension, and the plastic flow is therefore easier to localize into shear bands. This highly localization may lead to a poor tensile ductility at room temperature. The asymmetrical deviation of shear band angle from 45° has been revealed to be mainly ascribed to these two properties.

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