Materials Science and Engineering A 527 (2010) 2613–2620

Contents lists available at ScienceDirect

Materials Science and Engineering A

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

A modified criterion for shear band formation in bulk metallic glass under complex stress states

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ARTICLE INFO

Article history:
Received 11 September 2009
Received in revised form
10 December 2009
Accepted 23 December 2009

Keywords:
Bulk metallic glasses
Shear bands
Yielding criterion

ABSTRACT

A new criterion for shear band formation in metallic glasses is proposed based on the shear plane criterion proposed by Packard and Schuh [1]. This modified shear plane (MSP) criterion suggests that a shear band is not initiated randomly throughout the entire material under stress but is initiated at the physical boundaries or defects and at locations where the highest normal stress modified maximum shear stress occurs. Moreover, the same as in the shear plan criterion, the shear stress all over the shear band should exceed the shear yield strength of the material. For a complete shear band to form, both requirements need to be fulfilled. The shear yield strength of the material is represented by the shear stress of the point at which the shear band stops. The new criterion agrees very well with experimental results in both the determination of the shear yield strength and the shear band path.

1. Introduction

Bulk metallic glasses (BMGs) have drawn intensive interest of many researchers due to their impressive properties including high strength, large elastic strain, good corrosion resistance and high wear resistance [2,3]. However, the high strength of BMGs is always accompanied by remarkably small plastic deformation in comparison to crystalline metallic materials. The deformation and fracture behavior of metallic glasses have been extensively investigated in the past few decades [1,4–10]. A general observation is that the plastic deformation of metallic glasses is localized in very narrow regions of shear bands whose rapid propagation often leads to sudden fracture of the material [11–13]. The underlying micro-mechanisms of the plastic deformation of metallic glasses are fundamentally different from those of crystalline metals primarily due to the absence of long-range ordering in metallic glasses [11]. The flow localization in metallic glasses is considered to be related to the local change of viscosity within shear bands and there are two kinds of hypotheses to explain this phenomenon [14]. The first suggests that it is the formation of free volumes during deformation that decreases the viscosity within the shear bands and thus decreases the density of the material [15]. Such geometry softening will lead to severe localization and subsequent fracture along the shear bands. The second considers the local adiabatic heating generated during shear banding [16], and an estimation of temperature rise up to the glass transition temperature or even the melting temperature has been made, which could decrease the viscosity by several orders of magnitude [11].

Therefore, paralleling dislocation activities responsible for the yielding of crystalline metals, shear band formation signals the yielding of metallic glasses. Several criterions have been proposed for shear band formation within metallic glasses. The first and simplest is the maximum shear stress criterion which is commonly used for crystalline metals [17–19]. Bei et al. [4] interpreted their nanoindentation data of metallic glasses (Vit 1, BAM11, both Zr-based, and two Fe-based systems) by means of this criterion. In their study, the objective is to find the "theoretical strength". However, the physical picture of such "theoretical strength" is not clearly defined in the paper of Bei et al. [4]. According to Bei et al., the theoretical strength appears to be the stress that kicks off the first shear band. But this interpretation of "theoretical strength" seems at variance with the widely accepted understanding of theoretical strength [20].

Unlike in crystalline metals, the plastic deformation of metallic glasses has been found to exhibit significant sensitivity to the normal stress or to pressure [21–25]. One piece of experimental evidence that supports this notion is the tension/compression asymmetry in the yield stress and the deviations of the shear fracture plane from the maximum shear stress plane under uniaxial loading [21–24]. The pressure-modified criterion can be approx-

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doi:10.1016/j.msea.2009.12.045

imated by the Coulomb–Mohr yield criterion, which is widely used to describe the constitutive behaviors of granular materials [26]. Both the maximum shear stress criterion and the pressure-modified maximum shear stress criterion are based on the belief that the process of shear band formation is controlled by shear stress (or pressure-modified shear stress). As such when the highest maximum shear stress (or pressure-modified maximum shear stress) all over the material exceeds the yield shear stress it will initiate shear band.

Different from the aforementioned criteria, Packard and Schuh [1] contended that since the formation of shear band is a cooperative process along a specific shear plane, it requires the yield stress be exceeded everywhere along the entire path. In other words, it is not the highest shear stress all over the material but the lowest shear stress on a potential shear path that controls the formation of shear flow localization.

Naturally, the yield strength of a metallic glass extracted from a certain experiment is directly related to the yield criterion applied. Thus the comparison of yield strengths from different experiments will be meaningless if inconsistent yield criteria are used. This is particularly the case if the stress state is complex. It becomes a rather important issue in interpreting the experimental results so as to pin down the underlying physics such as responsible for the so-called size effect, for example [4,6,7,9,10,27]. Moreover, a good understanding of how shear band forms in metallic glasses will not only shed light on the underlying mechanism and physics of fracture and fatigue, but will also facilitate the design and fabrication of structure devices made from metallic glasses.

In this paper, we will critically evaluate the aforementioned yield criterions (or criterions for shear band formation) in bulk metallic glasses by applying them to derive the yield strengths of different experiments with various stress states. Particularly, we will provide detailed analysis of a spherical indentation test which serves as an example of complex stress state but at the same time can be analyzed more or less thoroughly. Based on such efforts, a new criterion is proposed to describe the requirement of initiating a shear band and to determine the yield strength of metallic glasses under stress concentration.

In what follows, we will first provide a detailed review of the shear plane criterion of Packard and Schuh [1]. A critical assessment of the maximum shear stress criterion and the pressure-modified criterion has been given by Packard and Schuh, and will not be detailed here. Then we will lay out a modification to the shear plane criterion. Finally we will use the modified shear plane criterion to re-examine various experimental data.

2. Review of the shear plane criterion

Recently, Packard and Schuh [1] have analyzed the stress state in metallic glasses under spherical indentation using the Hertzian contact theory. They also compared the theoretical results with their experiments on Pd-, Zr- and Fe-based BMGs. Their goal is to interpret the first stage of plastic deformation and to derive the yield stresses using the maximum shear criterion or pressure-modified maximum shear criterion. However, it turned out that neither of the two criteria agreed with the theoretical and experimental results. Based on the nature of shear banding processes in metallic glasses, and in order to reconcile the discrepancy between the much too high yield strengths from indentation experiments vis-à-vis the values derived from uniaxial loading, Packard and Schuh proposed the shear plane criterion briefly mentioned in the preceding section. Here we will provide a more detailed review on the shear plane criterion so as to lay the foundation for the new criterion to be developed later in this article.

The stress states arising from the contact between two elastic bodies was first studied by Hertz in 1881 [28]. Based on the Hertzian contact theory, the stress components, the principal stresses and maximum shear stresses under spherical indentation can be easily determined [28]. A nondimensionalized solution for the stresses was given in Ref. [1]. With the assumption that the angular direction of the shear path at every point is determined by the local shear angle, the shear trajectories within the material could also be obtained [1].

Fig. 1 is the distribution of the maximum shear stress and the modified maximum shear stress from Fig. 4 of Ref. [1]. The same stress contour map can also be found in the work of Bei et al. [4]. In Fig. 1, the contact radius is a and is used as a characteristic length of the indentation problem. P_m is the mean pressure on the contact surface and is used as characteristic stress. These two characteristic quantities are given by [1].

$$a = \left(\frac{3PR}{4E_r}\right)^{1/3} \tag{1}$$

$$P_m = \frac{4E_r a}{3\pi R} \tag{2}$$

where P is the applied load, R the radius of the indenter tip, E_r the reduced modulus [29].

We also made the following definition. τ' is the maximum shear stress normalized by the mean pressure P_m and is related to the maximum ($\sigma_1{}'$) and the minimum ($\sigma_3{}'$) principal stresses. p' is the normalized hydrostatic pressure and is calculated from the principal stresses. A pressure-modified shear stress is then defined as ($\tau' - \alpha p'$), where α is pressure coefficient (or the internal friction coefficient in the sense of the Coulomb–Mohr yield criterion). We have

$$\tau' = \tfrac{1}{2}(\sigma_1' - \sigma_3') \tag{3}$$

$$p' = -\frac{\sigma_r' + \sigma_z' + \sigma_\theta'}{3} \tag{4}$$

As can be seen from Fig. 1, the highest values of the maximum shear stress and the pressure-modified maximum shear stress are about $0.45P_m$ and $0.41P_m$, respectively. Meanwhile, in Ref. [1] the authors obtained P_m experimentally from the critical indentation load defined as the load at which the first shear banding event occurs. The yield strengths determined by the maximum shear stress criterion and pressure-modified maximum shear stress criterion are thus derived. A comparison of these results with the values of the expected shear yield stresses is shown in Fig. 2. The discrepancy is apparent and quite significant. Moreover, the shear

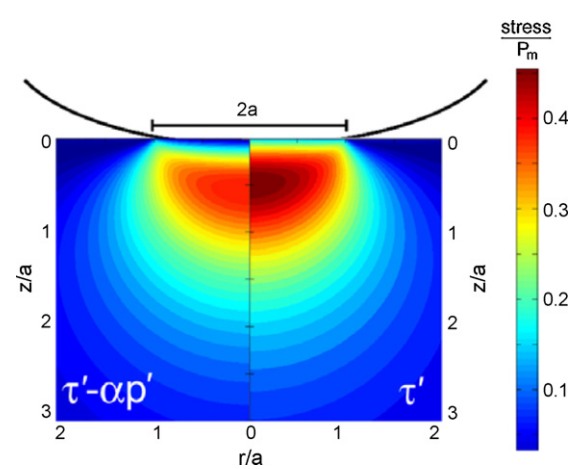

Fig. 1. Contours of maximum shear stress (τ', on the right) and pressure-modified maximum shear stress ($\tau' - \alpha p'$, on the left ($\alpha = 0.12$)) associated with a spherical indentation calculated using the Hertzian contact theory, from Ref. [1] (also to be found in the work of Bei et al. [4]). The stresses are plotted in P_m, the mean pressure under the spherical indenter.

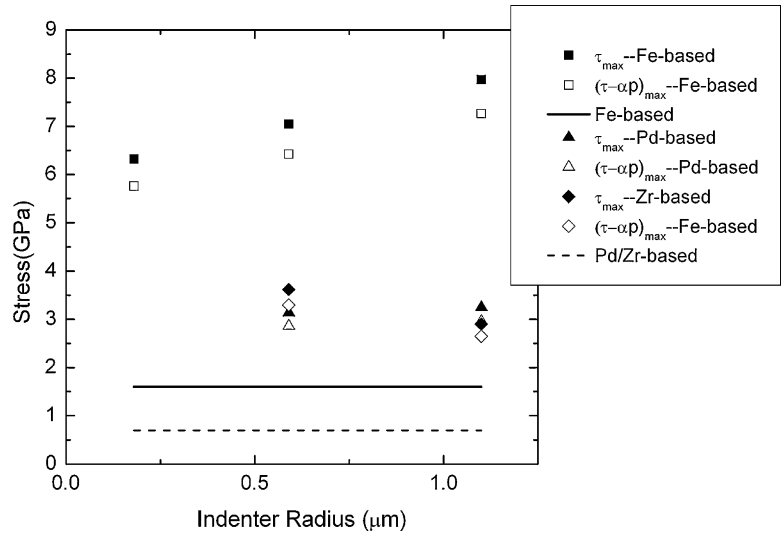

Fig. 2. Comparison of the shear yield stress calculated through the critical indentation loads by using the maximum shear stress criterion (τ_{max}) and the pressure-modified maximum shear stress criterion ($(\tau - \alpha p)_{max}$) with the expected values [1] (horizontal lines are values from macroscopic experiments).

paths under these two criterions could also be predicted by the Hertzian contact theory, as path D in Fig. 3 (the two shear paths are almost superposed, and hence they are denoted as one line). Fig. 3 also gives some other shear trajectories originated from $z = 0$, i.e. the surface of the specimen. The shear bands from a cylindrical indentation experiment are shown in Fig. 4 [30]. Although they might not be exactly the same with the spherical indentation, the mechanical similarities are sufficient to make some comparisons (the similarities between cylindrical indentation and spherical indentation will be discussed later in this article). There are three main shear bands in Fig. 4, denoted as A–C. However, none of them is even close to path D in Fig. 3 predicted by the maximum shear stress criterion or the pressure-modified criterion. Therefore, the seemingly irreconcilable differences between the predicted shear yield stresses/trajectories and the experimental results indicate that these criterions are not appropriate for metallic glasses under indentation conditions.

Packard and Schuh [1] then examined the stresses along all the potential shear paths and proposed the shear plane criterion for metallic glasses. There are two key features associated with the shear plane criterion. The first is that the plane which undergoes the highest shear stress along its entire path is the preferred shear plane. The second is that the lowest stress along that path defines the yield strength of the metallic glass. Based on these two points, Packard and Schuh reevaluated their experimental shear yield stresses using the shear plane criterion and observed much better consistency with the expected values, as shown in Fig. 5. Comparison between Figs. 2 and 5 reveals more clearly the improvement brought about by the shear plane criterion. Furthermore, based on the shear plane criterion, the preferred shear plane is now along path B in Fig. 3, as it has the highest stress along its entire path (curve B in Fig. 6, to be detailed later). This predicted shear plane is also consistent with the experimental observations, namely, shear band B in Fig. 4.

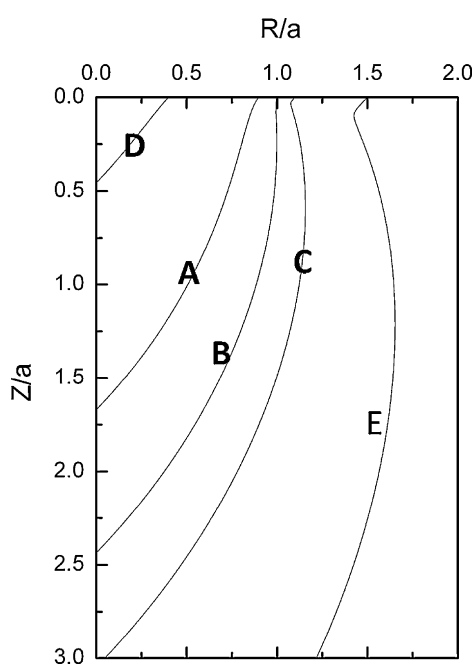

Fig. 3. Representative potential shear trajectories originated from the specimen surface, calculated using the Hertzian contact theory.

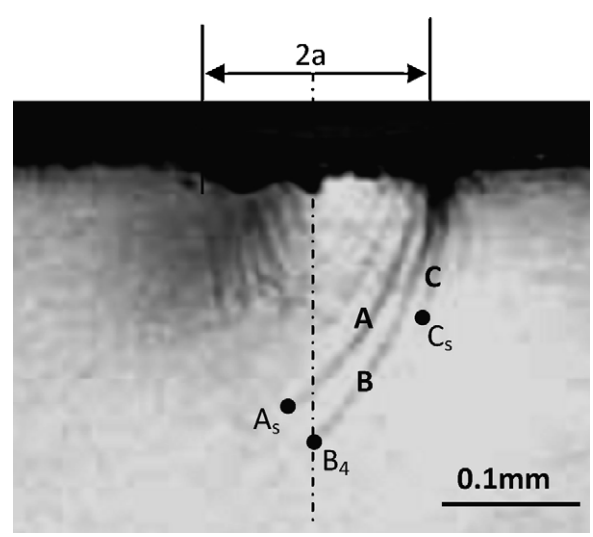

Fig. 4. Shear band paths under cylindrical indentation of a metallic glass, from Ref. [30]. The right half of this image has been used by Packard and Schuh [1]. Notice the major bands on the right-hand side, particularly the starting points of these bands which are at or at least close to the edge of the indent where large tensile stresses are expected.

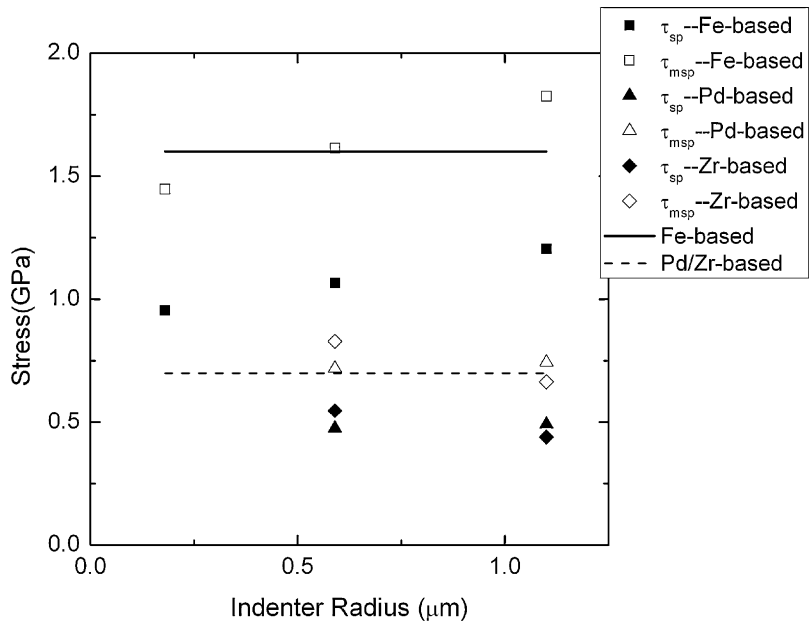

Fig. 5. Comparison of the shear yield stresses calculated through the critical indentation loads using the shear plane criterion (τ_{SP}) and the modified shear plane criterion (τ_{MSP}) with the expected values [1] (horizontal lines are values from macroscopic experiments).

3. Modified shear plane criterion

The shear plane criterion proposed by Packard and Schuh appears to be a much improved method for defining the shear banding behavior of metallic glasses. However, a closer examination suggests that there are still a few problems that need to be addressed. For example, Fig. 5 shows that the shear plane criterion consistently underestimates the yield stresses of all the three metallic glasses investigated by Packard and Schuh. In what follows, we attempt to provide a critical assessment of the shear plane criterion and to put forward a modified method that results in better predictions of the shear yield stress under complex stress state such as associated with indentation.

3.1. Determination of a complete shear band

The shear trajectories calculated using the Hertzian contact theory are just potential candidates along which the shear band may propagate. In other words, the shear band does not have to occupy the entire trajectory; it may instead stop at certain point within the path (see band C in Fig. 4 vis-à-vis curve C in Fig. 3). One may notice that the two boundaries of a shear trajectory calculated here and in Ref. [1] by the Hertzian contact theory are defined by $z=0$ and $r=0$, i.e. the contact surface and the symmetry axis of the (spherical) indentation problem, respectively. The symmetry axis ($r=0$) is, however, just a mathematical boundary, which means that the shear band does not have to stop at this axis but may go beyond it (see band A in Fig. 4, as compared with curve A in Fig. 3). The above two cases indicate that the actual shear band may not be identical to the theoretical shear trajectory with mathematically defined boundaries; it could be shorter or longer. This observation implies that determining the lowest shear stress along an actual shear band by using the stress on a mathematically confined trajectory is inappropriate. A more reasonable way is to take into account the start and end points of the shear trajectory.

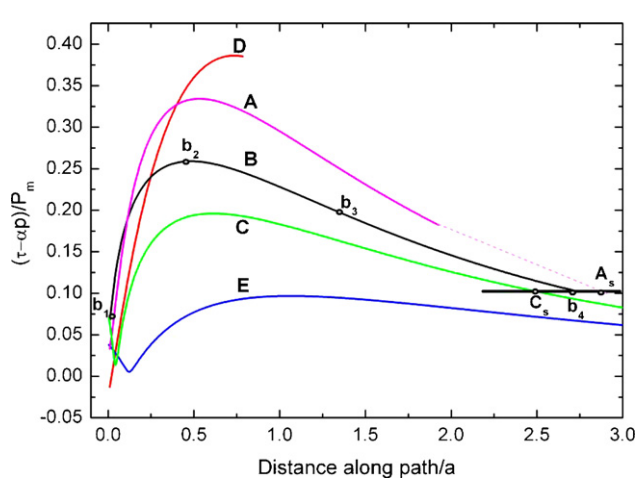

Fig. 6. The stresses along the shear trajectories in Fig. 3, from the starting points (on $z=0$) to the end points (on $r=0$). Notice that the letters in the plot correspond to the same used to denote the various paths in Fig. 3.

3.2. Initiation of the shear band

We have calculated the pressure-modified shear stresses along different shear trajectories displayed in Fig. 3, and the results are shown in Fig. 6. Suppose path B in Fig. 6 is the preferred shear plane. We have marked out four points, b_1–b_4 on path B for detailed consideration. From the shear plane criterion, the shear stress at b_1 under the load is roughly $0.07P_m$, and is the lowest stress point along path B. According to the shear plane criterion, it should therefore be taken as the shear yield strength of the material [1]. However, this yield stress has been exceeded earlier between points b_2 and b_3. Then a natural question arises: why dose not the shear banding event just occur earlier between b_2 and b_3? If it does, the yield strength should be changed to the stress at b_3 accordingly. This paradox could only be resolved if the requirement for shear band initiation is considered.

One may notice that in our calculation (as well as in Ref. [1]) of the shear trajectories by the Hertzain contact theory, an assumption is made that all potential shear paths originate from the surface of the material. To the best of our knowledge, under mechanical loading the shear localization of BMGs always starts at the physical boundaries of (under compression or tension) or defects in (usually under tension) the material. Since the material mostly

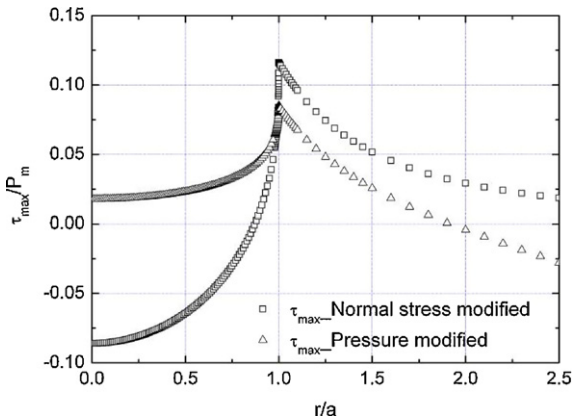

Fig. 7. Variation of the normal stress modified maximum shear stress on the specimen surface under a spherical indenter. Note the stress reach a peak value at $r = a$.

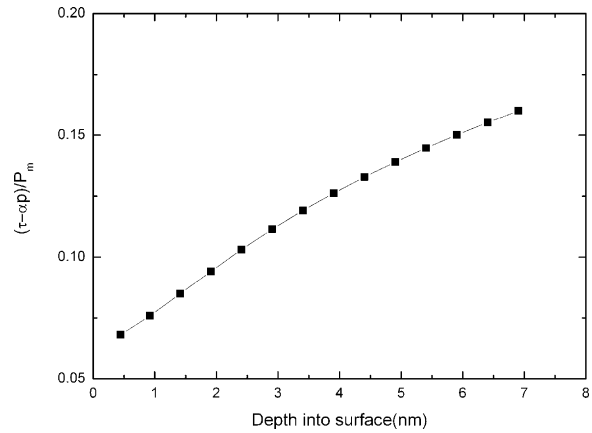

Fig. 8. Pressure-modified shear stress distribution from the surface to the inner part of the specimen, corresponding to path B in Fig. 3, a characteristic length corresponding to 0.59 μm indenter radius in Ref. [1] is used. Note the large stress gradient into the surface.

undergoes compression and the specimen surface is the only physical boundary of the indentation problem, the above assumption seems reasonable. In what follows we will focus on the stress analysis of the sample surface under indentation.

As discussed before, the tendency to shear localization of BMGs is greatly influenced by normal stress components. Moreover, its sensitivity to tensile stresses is quite different from that to compressive stresses. Flores and Dauskardt [5] found that the mode II fracture toughness of the Zr-based BMG exceed its mode I fracture toughness by four times, suggesting that the shear stress itself is not as effective as tensile stress to initiate a shear band. Zhang et al. [21] compared the fracture behavior of $Zr_{59}Cu_{20}Al_{10}Ni_8Ti_3$ under compression and tension and they also found that the shear localization is more sensitive to tensile loading. Lund and Schuh [22,24] analyzed the previous experimental data on the asymmetry characteristics of BMGs and found that the Mohr–Coulomb criterion is a more suitable criterion to describe the yield behavior of BMGs. The Mohr–Coulomb criterion says that the critical shear fracture stress, τ_y is expressed as $\tau_y = \tau_0 - \mu\sigma_n$ where σ_n is the normal stress acting on the shear plane, τ_0 is the critical shear fracture stress on a plane without normal stress and μ is a material constant. Based on previous experiments, they obtained an average value of $\mu = 0.26$ for tensile loading and $\mu = -0.11$ for compressive loading. Using the Mohr–Coulomb criterion, we analyzed the shear stress distribution on the specimen surface under indentation by Hertzian contact theory, and the results are shown in Fig. 7. The distribution of the pressure-modified maximum shear stress on the specimen surface is also plotted in Fig. 7. The stresses have peak values at $r = a$ by both criterions, although the peak stress values are slightly different. This stress distribution indicates that the shear band will probably initiate at the edge of the contact. Since the Mohr–Coulomb criterion considers the difference of the coefficients between tension and compression, it is preferred as the criterion for shear band initiation. The critical initiation shear stress is about $0.11P_m$ from Fig. 7.

It should be noted that the fulfillment of the initiation criterion is not sufficient for the formation of a complete shear band. Another requirement, which is also the key component of the shear plane criterion, is that the shear stress over the whole shear band should exceed the yield strength of the material. One will not observe a shear band or a load drop until both of the above two requirements are satisfied.

3.3. Determination of shear yield strength

The shear yield strength of BMGs could be determined by evaluation of the aforementioned two requirements. Similar to Ref. [1], we believe that the lowest shear stress over a complete shear band (*not* necessarily the potential shear path) represents the shear yield strength of the material.

Theoretically, the shear stress required to initiate a shear band should be the same as the shear yield strength. However, this stress was not adopted in the process of determining the strength. The reasons are listed below.

Firstly, as we have discussed before, the shear band may not come into being even when the initiation stress has been achieved. In other words, by the time when a complete shear band has been formed the stress at the initiation site may have changed and is no longer the shear yield strength of the material. Since the experimental results are always corresponding to the condition for the formation of a complete shear band, taking the stress at the initiation point of shear band as the shear yield strength could be misleading.

Secondly, the physical boundaries and defects are the preferred locations of the shear band initiation, but the stress state at those locations is not easy to analyze as it is affected by many factors such as surface quality or roughness. For example, Fig. 8 gives the variation of the pressure-modified maximum shear stress from the surface to the interior of the material along path B of Fig. 3. At the very top surface, the pressure-modified shear stress gets its lowest value, about $0.07P_m$, which is also the lowest throughout path B in Fig. 6. Since the lowest pressure-modified shear stress ($0.07P_m$) at this point of path B is the highest among all the trajectories, by the shear plane criterion it should be taken as the shear yield strength of this metallic glass. However, this value rises to $0.15P_m$ at about 5 nm deep into the surface, which indicates if the surface roughness is at this level (5 nm in the work of Packard and Schuh [1], for example), none of the stresses will be practical in the domain $0 \le z \le 5$ nm. Thus there are some other factors such as surface defects that affect the initiation of shear bands. For instance, surface defects are clearly visible in Fig. 4 on the left part of the indentation surface. Such surface defects might be the reason for the asymmetric pattern of the shear bands under the indenter. The above discussion and Section 3.2 suggest that the initiation of shear band is a rather complicated event and can be affected by many factors. The method of using the maximum shear stress at the initiation of shear band to predict the shear yield stress is apparently inadequate.

Unlike the initiation, the propagation of the shear bands is mainly related to the maximum shear stress (or the pressure/normal stress modified maximum shear stress) [5]. The shear band will grow along its maximum shear direction until the shear stress drops down below a critical value. Flores and

Table 1
Comparison between different shear band criterions (all stresses in GPa).

Metallic glass	τ_y [1]	τ_{max}		$(\tau-\alpha p)_{max}$		τ_{SP}		τ_{MSP}	
		M	E (%)	M	E (%)	M	E (%)	M	E (%)
$Pd_{40}Ni_{40}P_{20}$	0.7	3.20	357	2.91	316	0.48	−31.4	0.73	4.3
$Fe_{41}Co_7Cr_{15}Mo_{14}C_{15}B_6Y_2$	1.6	7.12	345	6.48	305	1.08	−32.5	1.63	1.9
$Zr_{49}Cu_{45}Al_6$	0.7	3.26	366	2.97	324	0.49	−30.0	0.75	7.1

M: magnitude; E: error. The magnitudes of the stress are all from the average of the spherical indentation results with different indenter radius.

Dauskardt [5] studied the critical shear stress required for continued propagation of the shear band by examining where the shear band stops and found a critical shear stress of 1075 MPa for $Zr_{41.25}Ti_{13.75}Ni_{10}Cu_{12.5}Be_{22.5}$, similar to the reported shear yield strength [31]. Therefore, it may be more reasonable to determine the shear yield strength of metallic glasses by studying where it stops instead of where it starts. Based on this, we have recalculated the shear yield strengths of the three BMGs studied by Packard and Schuh in Ref. [1] using the new criterion (hereafter referred to as modified shear plane (MSP) criterion). Our results are plotted in Fig. 5 to be compared with the experimental results as well as the results derived from the shear plane criterion. The same major shear band as that of Ref. [1], i.e. band B in Fig. 4, is chosen for calculation.

The first and critical step to use the MSP criterion is to determine where the shear band stops and then find the stress at this point by theoretical calculation. It is relatively easy when the shear band ends at the surfaces of specimen, such as those in uniaxial tension, compression or micro-compressions. When it comes to indentation where the shear band ends *in* the bulk, the exact location at which shear band stops is not easy to identify. However, in the later stage of the shear banding process, the variation of shear stress along the shear path is not so intense (see Fig. 6), which means that small discrepancy between the real location and the picked point will not introduce significant error. The values of shear yield strength derived based on different criterions are given in Table 1 to facilitate quantitative comparison. Clearly, the results from the MSP criterion have reduced the errors by two orders of magnitude vis-à-vis the maximum shear criterion; and by about one order of magnitude vis-à-vis the shear plane criterion.

Band B in Fig. 4 stops at B_4, and the shear stress at B_4 is adopted as the shear yield strength of this material by the MSP criterion. The shear stress at the last point of band A (i.e. at $r=0$) is larger than this yield strength, indicating that shear band A will not stop at $r=0$ but will continue to propagate. For band C, as the shear stress is smaller than the yield stress at $r=0$, the shear band will not go farther and it should stop at C_s as indicated in Fig. 6. Actually, the pressure-modified shear stress at point A_s and point C_s are also calculated using the Hertzian contact theory. Results show that $\tau_{A_S}=0.14P_m$ and $\tau_{C_S}=0.17P_m$, significantly larger than the shear yield strength ($\tau_{b_4}=0.10P_m$). However, the real values of these stresses should be lower than those calculated, because the calculation is based on an elastic assumption whereas the growth of secondary shear bands is under the precondition of the formation of the primary shear band. Since the applied load has been released by the first shear banding, the stresses along those secondary shear bands are greatly decreased.

It should be noted that our reference experiment is cylindrical indentation (the same as that in Ref. [1]), while all the theoretical calculations are based on the spherical indentation. The pressure distributions beneath the indenters can be expressed as [28]:

$$\frac{\sigma_z}{P_m}=-\frac{3}{2}\left[1-\frac{r^2}{a^2}\right]^{1/2} \quad \text{for spherical indenter and} \tag{5}$$

$$\frac{\sigma_z}{P_m}=-\frac{4}{\pi}\left[1-\frac{r^2}{a^2}\right]^{1/2} \quad \text{for cylindrical indenter} \tag{6}$$

These expressions are almost identical except for the slight difference (about 15%) between the magnitude of the coefficient ($-3/2$ for spherical indenter vs. $-4/\pi$ for cylindrical indenter). Considering the close similarities between the two cases, we believe that our calculation results should not be far from the real condition.

4. Comparison of the MSP criterion with the shear plane criterion

As we have pointed out, the MPS criterion derived in this work is inspired by the shear plane criterion proposed by Packard and Schuh [1]. Both criterions have been based on the premise that the shear plane is along one of the potential shear trajectories decided by the orientation of maximum shear stress, and that the formation of shear band in metallic glasses is a cooperative process. In what follows, however, we would like to discuss differences between the two criterions.

Firstly, the shear plane criterion is based on the notion that it is only the lowest shear stress along a potential shear trajectory that determines whether such a trajectory will be eventually selected as the shear plane. The way to obtaining the lowest shear stress is by checking the stresses along all the potential trajectories from the start point to the end point. As such, for example, all the paths in Fig. 3 have to be evaluated. One potential issue associated with this method is the determination of the start point and the end point. The two axis, i.e. $r=0$ and $z=0$, were chosen as the boundaries for calculation. Although the plane corresponding to $z=0$ represents the surface of the specimen and might be justifiably taken as a boundary, the position of the symmetry axis, $r=0$, is somewhat arbitrary. Moreover, as pointed out in Section 3.2, if the shear band initiation is only determined by the lowest shear stress along the shear plane, say b_1 on path B in Fig. 6, why does not the shear banding event occur earlier between b_2 and b_3, as the shear stresses between them have exceeded the stress at b_1 already? This suggests that the shear band initiation suggested by the shear plane criterion is debatable. The MSP criterion deems that the initiation of shear band is affected by many factors (defects and surface roughness for example) and always occurs on the physical boundaries of the problem. For the case of indentation, the shear band tends to initiate on the specimen surface at which the highest normal stress modified maximum shear stress is found.

Secondly, the shear plane criterion suggests that the shear yield strength of the material should be determined by the lowest shear stress along the shear plane. The MSP criterion is in keeping with that but it has further taken into account the surface roughness or defect effect. The extremely large stress gradient and the complicated conditions on the specimen surface (see Fig. 8) makes the local stress values unrealistically high. Alternatively, the MSP criterion chooses to examine the stress at the point where the shear band ceases and believes that the shear band will not stop until the shear stress drops down to a critical value, which we consider as the shear yield stress. In this sense, the critical shear stress *is* the lowest all over the shear plane.

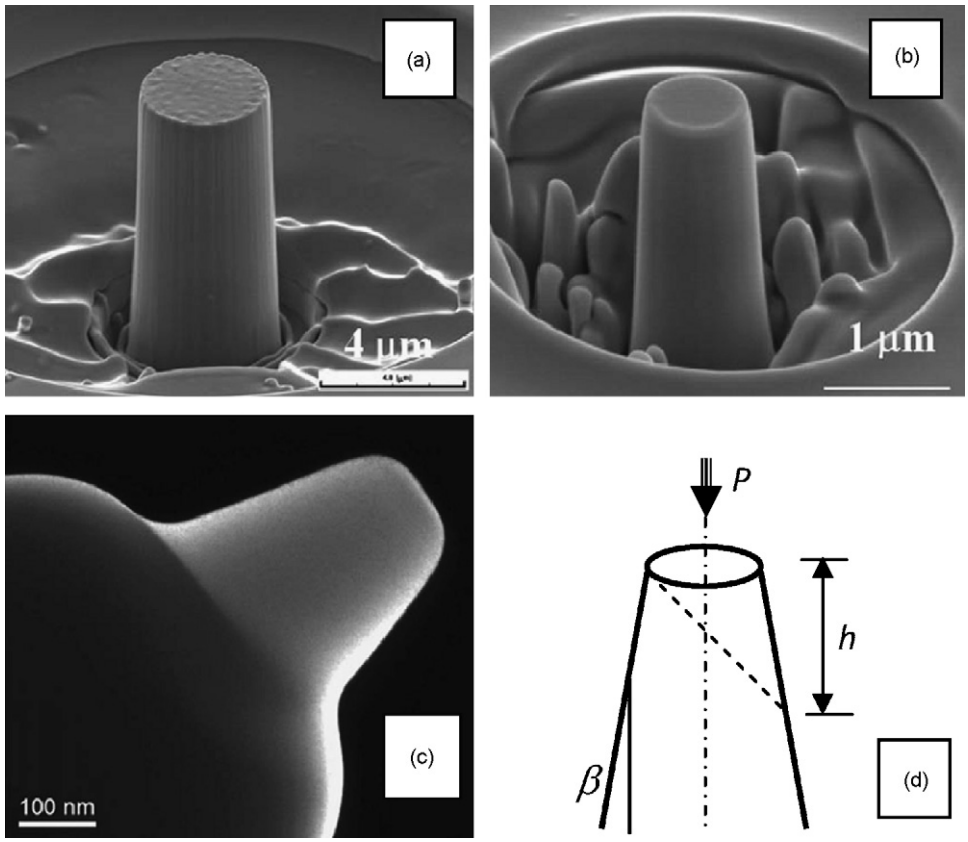

Fig. 9. Micropillars of different diameters: (a) 3.8 μm [7], (b) 1 μm [7], (c) top diameter 75 nm, bottom diameter 320 nm [27]; (d) definition of parameter pertaining to the calculation of shear yield stress using the shear plane or modified shear plane (MSP) criterion. Note the exacerbation of the taper angle and the irregular geometry of the pillars as the pillar size decreases.

5. Implications to the size effect

The so-called specimen size effect of BMGs has been intensively studied recently [1,4,6,7,9,10,27] and is still an issue of strong debates. While some researchers have reported that the strength of BMGs increases with decreased sample size [4,6,7,10], others have observed that the strength enhancement is at least in part extrinsic [1,9,27]. Bei et al. [4] and Wright et al. [10] have both studied the mechanical behavior of Zr-based metallic glass under nanoindentation using the Hertzian contact theory. The yield stresses they reported are more than three times larger than the shear yield stress of bulk specimens. They attributed this high yield strength to the "defect free" characteristic of the material due to the small test volume. However, the methods used in those studies to determine the shear yield stress are the maximum shear stress criterion or normal stress modified shear stress criterion, which could be misleading as pointed out by Packard and Schuh [1]. As the yield stress of BMGs is deemed to be close to the theoretical limit, an additional increase by three times seems physically unreasonable [1]. Moreover, in those studies the shear yield stresses measured by nanoindentation were just compared with those from uniaxial compression tests. The comparison between experiments with radically different stress states seems not as convincing as comparison between similar experiments, as done in Ref. [1]. Three indenter sizes were used in Ref. [1] and no apparent size effect were observed (see Figs. 2 and 5 of this work). All the above discussions indicate that those so-called "size effect" might be an artifact.

The size effect of BMGs has also been investigated by micro-compression [6,7,9,27,32]. Lai et al. [6] reported that the yield strength of micropillars of a Zr-based metallic glass with diameters from 3.8 μm to 0.7 μm is 25–86% larger than their bulk counterparts. Similarly, Lee et al. [7] claim that their micropillars of a Mg-based metallic glass are 60–100% stronger than the bulk specimens. They both calculated the yield stress by using the maximum shear criterion and both attribute the strength enhancement to the decreased defect population of the smaller specimens. One

Table 2
Shear yield strengths (τ_y, GPa) for different bulk metallic glasses based on different experiments and comparison against theoretical predictions.

Test method	Material	τ_y from Refs.	τ_y (uniaxial test)	τ_y (shear plane)	τ_y (MSP)
Nanoindentation [4]	$Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$	3.1 [4]	0.85 [33]	0.49	0.70
Nanoindentation [4]	$Zr_{52.5}Al_{10}Ti_5Cu_{17.9}Ni_{14.6}$	2.9 [4]	0.82 [34]	0.46	0.65
Nanoindentation [10]	$Zr_{40}Ti_{14}Ni_{10}Cu_{12}Be_{24}$	2.9 [10]	0.95 [10]	0.60	0.91
μ-Compression [7]	$Mg_{65}Cu_{25}Gd_{10}$	0.81 [7]	0.46 [7]	0.56[a]	0.56[a]
μ-Compression [6]	$Zr_{63.8}Ni_{16.2}Cu_{15}Al_5$	1.20 [6]	0.92 [6]	0.96	0.96
μ-Compression [27]	$Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$	2.32[b] [27]	0.85 [33]	0.68	0.68

[a] The values were calculated through direct measuring the top diameter and the diameter where lowest shear stress occurs along a shear band in Fig. 1(a) in Ref. [7] (42° shear direction were assumed).
[b] This value was calculated by using the maximum shear stress on the top of the micropillar.

issue associated with the fabrication of the micropillars is that those pillars are usually tapered because of the divergence of the ion beam (see Fig. 9). Schuster et al. [9] have compared the compression results of tapered micropillars with non-tapered ones for a Pd-based metallic glass. No significant dependence of strength on specimen size from ~2 μm to 20 μm for non-tapered pillars was convincingly established. An extrinsic size effect is observed for tapered specimens if the maximum shear stress criterion was applied. Their observations again suggest that the maximum shear stress criterion (or pressure/normal stress modified maximum shear stress criterion) is not adequate for metallic glasses. They reevaluate the experimental data using the shear plane criterion and estimate the shear yield stress by the following equation (with a shear band angle ~42°):

$$\tau_{SP} \approx \frac{P}{2\pi(r + h\tan\beta)^2} \tag{7}$$

where P is the applied compressive load, r the radius of the pillar top, h the distance from the end of the shear band to the top of the pillar and β the taper angle, see Fig. 9(d). By using Eq. (7), the geometry influence is eliminated and the yield shear stress of tapered specimens is comparable with those of the non-tapered ones. The shear yield strength of the pillars with diameters of 3.8 μm in Lai et al.s' paper was recalculated with the shear plane criterion and the results showed that they might have overestimated the shear yield strength by 20%. A 20% decrease of the shear yield strength makes it very close to that of its bulk counterparts, suggesting that there is no intrinsic size effect. Fabrication of small, taper-free pillars becomes more challenging with decreased pillar size. Furthermore, since irregular geometry may be produced with very small, nanometer sized pillars such as shown in Fig. 9(c), caution must be exercised when trying to derive yield stress from such pillars. Some of these experimental results are listed in Table 2. Applying the shear plane criterion, an over-all softening effect is observed. No remarkable size effect is established if the MSP criterion is used, indicating that conclusions regarding experimental evidence for the size effect in bulk metallic glasses need to be welcomed with due caution.

6. Summary and concluding remarks

A new criterion for shear band formation in metallic glasses, the modified shear plane (MSP) criterion, has been proposed in this work through analyzing the stress field of spherical indentation using the Hertzian contact theory. This criterion suggests that the shear band always initiates at physical boundaries or defects of the material and at locations where the highest normal stress modified maximum shear stress occurs. Moreover, as in the shear plane criterion, the shear stress all over the shear band should exceed the shear yield strength of the material. For a complete shear band to form, both of these two requirements need to be satisfied. The MSP criterion also suggests that the shear stress at the point where the shear band stops determines the shear yield strength of the material. This criterion has improved accuracy in predicting the shear yield stresses of some typical metallic glasses under spherical indentation.

For uniaxial tests, the MSP criterion converges with the shear plane criterion and the pressure/normal stress modified maximum shear stress criterion, because the maximum shear stress all over the specimen is considered to be uniform and the potential shear paths predicted by MSP criterion are identical with the maximum shear planes. For micro-compression tests, especially when the pillars are fabricated with tapered geometry, the maximum shear stress criterion should not be used, but the MSP criterion and the shear plane criterion could be equally applied. This is because in such cases the shear bands usually initiate on one surface, and propagate along the maximum shear path until reaching another surface, where the lowest shear stress is located. When it comes to indentation tests, the incipient shear band tends to form on the specimen surface which is the only physical boundary of this problem, and tend to cease *within* the material. The absence of physical boundaries makes it improper to use the shear plane criterion, and the MSP criterion appears to be a better choice.

Acknowledgements

The authors would like to thank Packard CE and Schuh CA for their pioneer work on this subject. This work is supported by the 111 Project of China (Contract No. B07050) and by the National Science Foundation of China (Contract No.10932008).

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