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Intrinsic factor controlling the deformation and ductile-to-brittle transition of metallic glasses

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The energetic driving force and resistance for shearing and cracking in metallic glasses (MGs) are quantitatively evaluated. A universal thermodynamic criterion is proposed for better understanding the intrinsic correlations between fracture toughness and Poisson's ratio, the competitions between various deformation modes and the ductile-to-brittle transition in MGs and other materials. A new cooperation parameter δ is also introduced to depict quantitatively the relative propensity of shearing versus cracking. This work could provide insights into the long-standing issues of deformation mechanisms of glassy materials, and be helpful in searching for ductile and tough MGs.

Keywords: metallic glass; thermodynamics; Poisson's ratio; ductile-to-brittle transition; shear bands

1. Introduction

Metallic glasses (MGs) usually exhibit extraordinarily high strength two times or even higher than that of their crystalline counterparts, owing to their amorphous nature, yet their fracture toughness covers a wide range of over almost three magnitudes [1–3]. For instance, the toughness of Mg-based MGs may approach that of ideally brittle solids [4], while Pd-based MGs may exhibit outstanding damage tolerance or even a rising crack growth resistance (*R*-curve) behaviour [2]. Meanwhile, the deformation modes beyond the elastic limits may also vary considerably among different MGs. Generally, macroscopic plastic deformation can usually be observed in intrinsically plastic MGs, while quasi-cleavage fracture or cracking may dominate in the brittle ones at room temperature [5–7]. The plastic deformation of MGs is normally mediated by shear bands induced by microscopic shear plastic flow [7]. In comparison, the brittle fracture of MGs is closely associated with the microscopic cavitation process, which necessitates volume dilatation [8]. As a consequence, the intrinsic plasticity or brittleness as well as the ductile-to-brittle (DTB) transition of MGs can be interpreted in terms of the competition between the shear plastic flow (or shearing) and volume dilatation (or cracking) processes. At the microscopic scale, both shearing and cracking originate fundamentally from the local rearrangement of atoms, i.e. the former corresponds to the cooperative

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shear motion of the atomic clusters, termed as the shear transformation zones (STZs), and the latter is caused by the separation of constituent units or the activation of the proposed tension transformation zones (TTZs) [5–7].

Fundamentally, the intrinsic plasticity or brittleness and the macroscopic deformation modes of MGs are closely related to their atomic-level structures, such as the atomic topological arrangements and bonding states [9,10]. For instance, it is revealed by atomistic simulations that decreasing the degree of full-icosahedral order facilitates the initiation of plastic flow and alleviates strain localisation, thus contributing to good ductility/toughness of MGs [9]. However, it is still extremely difficult to experimentally characterise and accurately describe the complicated atomic structures of MGs [10]. As a consequence, the underlying physics behind the diverse mechanical behaviours, the various deformation modes and the DTB transition of MGs still remains a challenge. Also, it is difficult to establish quantitative relations between the structures and mechanical properties [9]. The mechanical properties have so far been commonly correlated

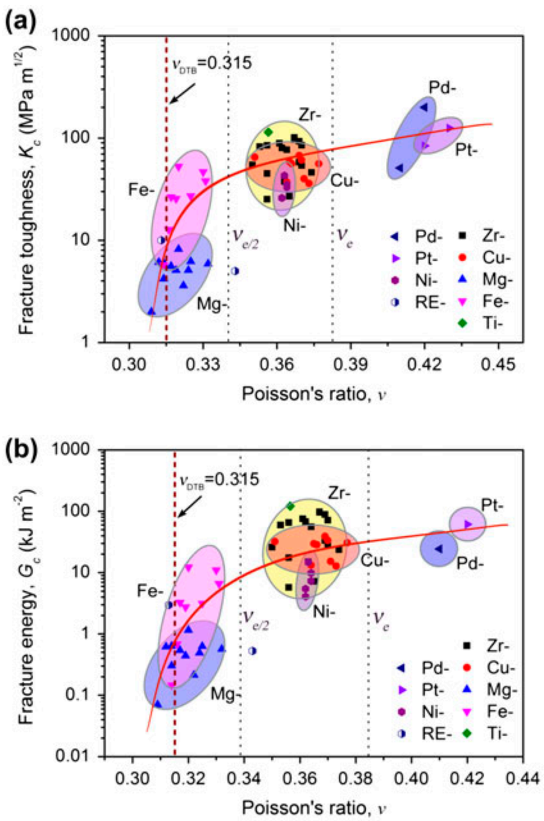

Figure 1. Variations of the (a) fracture toughness and (b) fracture energy as a function of Poisson's ratio ν among different alloy systems of MGs. A DTB transition occurs at $\nu_{DTB} = 0.315$. RE represents the rare earth elements. The MGs can be roughly categorised into four groups with different levels of toughness and different deformation modes according to ν as separated by the dashed lines.

with several basic physical parameters related to the elastic constants, which can be readily measured in MGs owing to their macroscopic isotropy and homogeneity [2,3,11–13]. For example, the Poisson's ratio v, which depicts the relative transverse strain compared with the longitudinal strain of a material when subjected to longitudinal elastic loading, has been regarded as a measure of the intrinsic toughness and plasticity of MGs [2,11,12]. As shown in Figure 1, both the fracture toughness and fracture energy of MGs increase monotonically with increasing v, and an obvious DTB transition occurs in the range of v from 0.31 to 0.32 [12]. The beneficial effects of a higher Poisson's ratio in a Pd-based MG with pronounced plasticity and toughness have been explained in terms of the competition between shear banding and cracking [11]. Also, it is supposed that a higher Poisson's ratio causes the tip of a shear band to extend rather than to initiate a crack, thus favouring good plasticity/toughness. Although the empirical relation between Poisson's ratio and toughness/ductility has been widely accepted and even applied to explore tough/ductile MGs [2,14,15], a deep and quantitative understanding on the physical mechanisms behind this relation as well as the diverse mechanical behaviours and various deformation modes in MGs is still required. Two crucial questions need to be further addressed: (i) why and in what way can the normally plastic-related toughness be dominated by an elastic parameter? and (ii) what are the physical origins of the DTB transition at the critical v and the origins of the various deformation modes in MGs and other materials?

Here, we propose a universal thermodynamic criterion that uncovers the underlying mechanisms for the Poisson's ratio-toughness/ductility correlations, the DTB transition and the various deformation modes in MGs by analysing the energetic driving force and resistance for shearing and cracking. It is found that the deformation modes of different materials can be unified by this criterion using a newly introduced cooperation parameter δ. We also show that the criterion is helpful for developing tough/ductile MGs.

2. Thermodynamic analysis

When subjected to external forces, MGs may in principle deform beyond their elastic limits through either shearing or cracking processes that are intimately associated with the mutually competing shear plastic deformation and volume dilatation at the microscopic scale, respectively [5–8]. Indeed, cracking may arise in MGs no matter whether preceding shear plastic deformation has occurred. The preferential occurrence of cracking relative to shearing in brittle MGs has been predicted by atomistic simulations and observed experimentally [8,16]. In addition, it has been revealed by theoretical calculations and atomistic simulations, as well as by experimental observations, that preparatory shear plastic deformation can also lead to cracking [8,17,18]. It is supposed that the competition between the preferring occurrence of shearing versus cracking reflects whether the MGs have an intrinsic capability to undergo plastic deformation, or whether there is a tendency for them to deform plastically rather than to fail in a brittle manner [6]. Here, we mainly focus on the intrinsic plasticity or brittleness of MGs by evaluating the inherent propensity for shearing relative to cracking by analysing their respective thermodynamic driving force and resistance.

Considering the stress condition of uniaxial loading shown in Figure 2, the total elastic strain energy per unit volume created by the external stress can be written as

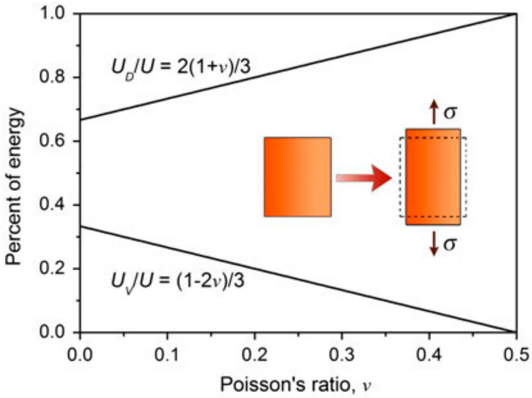

Figure 2. Dependences of the energy partitions for deformation U_D/U and volume dilatation U_V/U within the total input mechanical energy density on Poisson's ratio v. U_D/U increases while U_V/U decreases both linearly with the increase in v in an elastically strained sample shown in the inset.

$U = \sigma^2/2E$, where σ and E denote the elastic stress and Young's modulus of the sample, respectively. According to the theory of elasticity [19,20], the total mechanical energy per unit volume can be further divided into two individual parts – one arising from the change in shape and the other from the change in volume of the material. These two energy inputs will contribute to shear and volume changes, which correspond well to shear plastic deformation and volume dilatation, respectively. Thus, the corresponding energies per unit volume, which can be termed as the deformation strain energy density U_D and volume strain energy density U_V, can be regarded as the thermodynamic driving forces for the competing processes of shearing and cracking in MGs, respectively. In isotropic materials, U_D and U_V are tightly associated with the Poisson's ratio v and can be calculated by $U_D = (1+v)\sigma^2/3E$ and $U_V = (1-2v)\sigma^2/6E$, respectively [19,20]. As shown in Figure 2, both the energy partitions for shearing, U_D/U, and cracking, U_V/U, within the total mechanical energy density depend linearly on v but in opposite ways. With an increase in v, more energy will be consumed in an attempt to change the shape rather than the volume of the material. Here, only the general range of v for most isotropic materials from 0 to 0.5 is considered despite its theoretical numerical limit of $-1 \leq v \leq 0.5$. [13]. Accordingly, the reduced driving force for shearing relative to cracking can be described by

$$U_D/U_V = 2(1+v)/(1-2v). \tag{1}$$

As shown in Figure 3, U_D/U_V increases monotonically with increasing v, implying that the mechanical energy input by the applied stress is prone to contribute to shearing rather than cracking in materials with higher v.

With respect to the thermodynamic resistance, shear plastic flow can occur only when the driving energy for shearing U_D exceeds the critical energy threshold – the maximum shear elastic strain energy per unit volume W_D sustainable by the material. Analogously, the critical energy threshold for cracking induced by volume dilatation

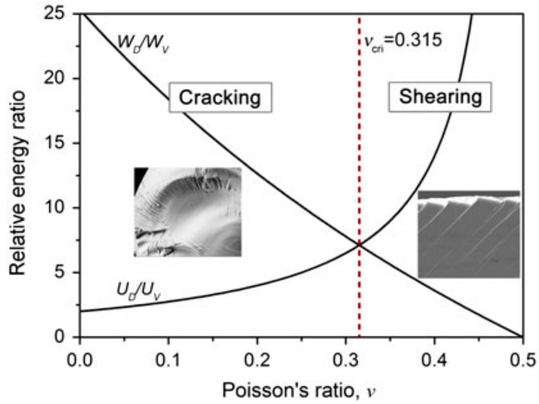

Figure 3. Variations of the reduced thermodynamic driving force U_D/U_V and resistance W_D/W_V of shearing versus cracking as a function of Poisson's ratio v. The dashed line indicates the transition in the deformation mode from cracking to shearing of which typical morphologies are shown in the insets.

equals the maximum volume elastic strain energy density W_V sustainable by the material. Thus, W_D and W_V can be regarded as the thermodynamic resistances for shearing and cracking in MGs, respectively. Neglecting the tiny changes in the elastic moduli with elastic strain, the resistances can be expressed as $W_D = G{\gamma_C}^2/2$ and $W_V = K{\chi_C}^2/2$, where G and K denote the shear modulus and bulk modulus, and γ_C and χ_C represent the maximum shear elastic strain and volume elastic strain of the material, respectively [19,20]. Thus, the reduced resistance to shearing relative to cracking can be expressed as

$$W_D/W_V = G{\gamma_C}^2/K{\chi_C}^2. \tag{2}$$

As the ratio between G and K can be described in terms of v in isotropic materials by $G/K = 3(1-2v)/2(1+v)$ [13,20], the relative resistance W_D/W_V can be further expressed as a function of v as

$$W_D/W_V = 3(1-2v){\gamma_C}^2/2(1+v){\chi_C}^2. \tag{3}$$

Therefore, both the thermodynamic driving force and resistance for shearing and cracking are closely related to the Poisson's ratio v. The competition between different deformation modes is clearly manifested by the relative values of U_D/U_V and W_D/W_V. From the thermodynamic viewpoint, shearing tends to prevail when $U_D/U_V > W_D/W_V$, while cracking will dominate when $U_D/U_V < W_D/W_V$. Therefore, a transition between these will occur at a critical v value (v_{cri}) at which U_D/U_V equals W_D/W_V. Considering the detailed cases for MGs, the transition from shearing to cracking corresponds well to the transition from intrinsically ductile/tough to brittle, i.e. the DTB transition. Thus, the critical values of Poisson's ratio for these two transitions should also be equal, i.e. $v_{cri} = v_{DTB} = 0.315$.

Quantifications of the critical elastic strain limits γ_C and χ_C have been stimulated by recent advancements in the field of MGs. It has been revealed that the mechanical failure, both plastic and quasi-cleavage, of MGs is closely associated with the rearrangement of atoms in local regions, i.e. the so-called STZs or TTZs [4–7]. This is supposed to be equivalent to the microscopic glass transition process or fluidization of the MGs induced by the input of mechanical energy [13]. Indeed, the equivalence between the onset of shear banding or local softening and the glass transition has been firmly verified by theoretical studies and experimental observations [13,21,22]. The onset of cracking or cavitation induced by volume dilatation is also related to the local softening process [6]. This is supported by experimental results revealing that characteristic patterns of plastic flow at different length scales can be observed on the fracture surfaces of both ductile and brittle MGs [4,6,16]. Therefore, γ_C and χ_C can be regarded as the critical shear and volume elastic strains, respectively, for individually activating the local softening or glass transition in MGs or fluidizing them.

It has been reported that different alloy systems of MGs invariably yield at nearly the same critical shear elastic strain γ_C at room temperature [23]. This is supported by the near linear relation between the yield strength (which is considered to be about twice of the maximum shear elastic stress) and the shear modulus for MGs [13]. The data have been fitted to a γ_C value of ~0.0267 ± 0.0020 [23]. In parallel, a critical value of the free-volume content ~2.4% has been proposed to be a sufficient condition for the onset of plastic flow in various MGs by theoretical calculation [24]. Independent careful experimental measurements have also shown that the free-volume content almost invariably approaches ~2.35% in MGs when the glass transition occurs [22]. The agreement between these two values further confirms that the yielding and glass transition are equivalent processes that can be induced by an increase in free volume. Accordingly, the critical free-volume content for yielding $V_f{}^*$ can be taken as 2.35%. As a consequence, the critical volume elastic strain χ_C can be quantified according to $\chi_C = V_f{}^* - V_f{}^0$, where V_f^0 denotes the content of the initially frozen-in free volume in MGs. This content is revealed to depend primarily on the cooling rates of the as-quenched samples. By measuring the volume changes in a Zr-based MG through positron annihilation and density measurements, Nagel et al. [25] have reported that an excess volume of the order of ~1% can be quenched into MGs at a cooling rate as low as 1–2 K/s. In comparison, for alloys cooled at a relatively higher rate of ~100 K/s, Wen et al. [26] and Xu et al. [27] have shown, by differential scanning calorimetry measurements, that the deduced free-volume content is higher than 2%. The initial free-volume content measured by other groups using different methods also varies [28,29]. Thus, it is difficult to determine an absolute value and there are corresponding deviations for V_f^0. Here, we have deduced the values of χ_C and V_f^0 in reverse, based on the observed critical Poisson's ratio of 0.315, i.e. determining their values to make U_D/U_V and W_D/W_V equal to each other at $v_{cri} = v_{DTB} = 0.315$, as shown in Figure 3. It turns out that an obtained free-volume content of $V_f^0 \approx 1.7\%$ fits well within the generally reported range of ~1–2% for MGs. Also, the corresponding critical volume strain of $\chi_C = V_f{}^* - V_f{}^0 \approx 0.65\%$ agrees well with the reported free-volume increase (~0.6%) within the shear bands of MGs [30], further supporting the chosen values of χ_C and V_f^0. Accordingly, the reduced resistance W_D/W_V can be described as a function of v by

$$W_D/W_V = 25.31(1-2v)/(1+v). \tag{4}$$

As shown in Figure 3, W_D/W_V decreases monotonically with the increase in v. This indicates a lowering energy barrier for shearing relative to cracking in MGs with higher v. It is noted that the global trend of W_D/W_V varying with ν holds independent of the particular values of χ_C and V_f^0 although the quantitative results may be slightly changed.

3. Discussion

Based on the above analysis, both the reduced thermodynamic driving force, U_D/U_V, and the resistance, W_D/W_V, of shearing versus cracking beyond the elastic limit are dominated by the Poisson's ratio ν in MGs. Quantitatively, U_D/U_V and W_D/W_V vary with ν in opposite ways and are equal to each other at the critical Poisson's ratio $\nu_{cri} = 0.315$. The Poisson's ratio-ductility/toughness relations and DTB transition in MGs have been widely accepted and successfully applied in the search for ductile/tough MGs [2,14,15]; yet, the physical mechanisms behind these relations are far from being understood. In this study, the mechanisms have been unravelled quantitatively in terms of the competition between shear plastic deformation and cracking: easy shearing concurrent with inhibited cracking favours higher toughness, and shear banding, as the main plastic deformation mechanism [5,31], is indeed the main factor improving the toughness/plasticity of MGs. The mechanical energy input can be readily consumed by shear band-mediated plastic deformation owing to the ease of shearing in MGs with a higher ν; while brittle fracture becomes dominant in MGs with a lower ν owing to the increased trend towards cracking. Thus, higher toughness and plasticity can be expected in MGs with a higher Poisson's ratio.

To describe more explicitly the deformation modes of MGs and other materials, we further propose here a new parameter δ by integrating the contrary thermodynamic aspects of driving force and resistance as

$$\delta = \frac{U_D/W_D}{U_V/W_V}. \tag{5}$$

From a thermodynamic perspective, shearing or plastic deformation is preferred in materials when $\delta > 1$ and cracking when $\delta < 1$. The two modes are comparable to each other at the critical point of $\delta = 1$ where the DTB transition occurs. Thus, the competition between the two fundamental processes of shearing or plastic deformation and cracking can be quantified by the parameter δ in MGs and other materials. Of the microscopic deformation mechanisms, shear plastic deformation originates from the cooperative motion of local atomic clusters in MGs [5–7,23], while cracking normally stems from the segregation of constituent units [6,8]. Therefore, the newly introduced parameter δ reflects the intrinsic capability of the local constituent units to coordinate their motion cooperatively rather than to separate from each other and can, thus, be termed as the 'cooperation parameter'. In MGs, δ can be readily determined from the basic material parameter ν using

$$\delta = \frac{U_D/W_D}{U_V/W_V} = (1+\nu)^2/12.65(1-2\nu)^2. \tag{6}$$

As shown in Figure 4, an increase in ν gives rise to a higher U_D/U_V concomitant with a lower W_D/W_V, and vice versa. Such effects contribute to an increased δ for an improved shearing trend over cracking and, as a result, favour higher intrinsic toughness and plasticity of MGs.

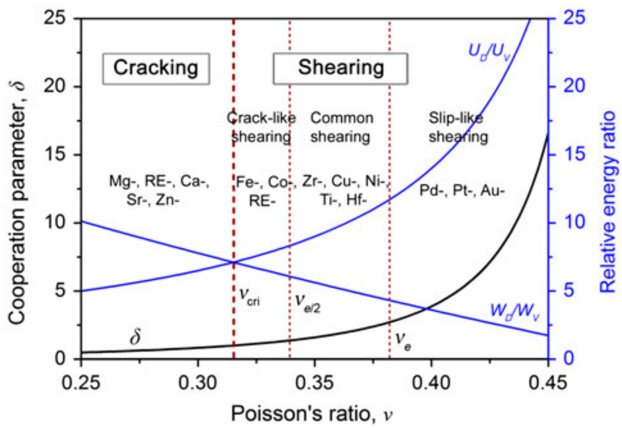

Figure 4. Reduced thermodynamic driving force U_D/U_V and resistance W_D/W_V of shearing relative to cracking as well as the cooperation parameter δ as a function of Poisson's ratio v in MGs. The detailed deformation modes of MGs are roughly categorised into four types according to δ with the increase in v, as separated by the dashed lines.

Next, we show that the parameter δ can characterise the DTB transition and classify the deformation modes in MGs. Careful examination of Figure 1 reveals that the toughness of MGs may vary significantly among different alloy systems, but is firmly associated with their Poisson's ratios even for those with $v > v_{cri}$. The toughness is also expected to be intrinsically correlated with the different shearing propensities relative to cracking in the deformation mode. In face-centred-cubic crystalline metals and alloys, the slipping modes of dislocations are usually characterised as planar slip or wavy slip, which exhibit different degrees of strain localisation and surface slip morphologies [32,33]. Different slipping modes usually give rise to varying plastic deformabilities of the crystals. In a similar manner, here, the detailed shearing modes of various MGs are also classified into different types depending on the parameter δ. For the Fe- and Co-based MGs which usually display a typical δ in the range $1 < \delta < \backsim e/2$, the shearing, although still prevailing over cracking, can be regarded as crack-like as it tends to develop through sparse shear bands with low stability and a high cavitation trend [16,34]. In contrast, for MGs with a high value of δ ($\delta > e$) such as those based on noble metals, the shearing typically advances through uniformly distributed multiple shear bands with high stability [2,11]. The shearing mode is analogous to some extent to the slipping of dislocations in crystals and can, thus, be termed as slip-like shearing. Therefore, the detailed shearing mode gradually shifts from a crack-like to a slip-like one with increasing δ for the shearing-dominated MGs. The demarcation points can be set as $\delta = e/2$ and $\delta = e$, whereby the values of v correspond to 0.339 and 0.382, respectively. In such a scenario, the detailed deformation modes of MGs can be roughly categorised into four types according to the parameter δ, i.e. cracking, crack-like shearing, common shearing and slip-like shearing, which depend fundamentally on their Poisson's ratio in the reported range of ~0.25–0.43 [13], as shown in Figure 4. It is noted that the characteristic δ values of $e/2$ and e were chosen mainly based on our observations of the general trends among different alloy systems of MGs. Although the determination is somewhat empirical, the competition between shearing and cracking is indicated well by δ or even $\ln \delta$ (the shearing propensity versus cracking increases with an increase in $\ln \delta$ from 0 to 1), and describes the experimental results reasonably well, as shown in Figure 1. Furthermore, the deformation modes in MGs and other materials may vary in a continuous manner and be not so distinctive among different alloy systems. We anticipate that the present categorisation can help in understanding the various shearing behaviours in a manner analogous to slipping modes (planner slip or wavy slip) of dislocations in crystalline materials.

Although the quantitative relations between δ and ν as described by Equation (6) may not be applicable to materials other than MGs, the parameter δ per se can act as an indicator of the relative propensity of plastic deformation versus cracking among various materials. It is also expected to be capable of describing the different deformation modes of materials. For instance, liquids with ν approaching 0.5 have a negligible resistance to deformation ($W_D/W_V \longrightarrow 0$), while all the input mechanical energy is expended in deforming or changing the shape at the same time ($U_D/U_V \longrightarrow \infty$) [35], as shown in Figure 2. Thus, when δ tends to infinity, the liquids flow quite easily. On the other hand, crystalline alloys usually exhibit a low resistance to dislocation-mediated plastic deformation compared to the shear banding in MGs, and thus display a high δ [32]. Consequently, the slipping of dislocations prevails and contributes, in general, to a high plastic deformability. In contrast, the deformation of ceramics and rocks necessitates the rupture of covalent bonds, which results in a high resistance to shearing [35]. Thus, a low δ is expected when cracking is preferred, causing the apparent brittleness of these materials. The difference among various deformation modes is supposed to lie fundamentally in their varying degrees of strain localisation, supporting the validity of δ in assessing the strain localisation tendency of materials. The toughness of MGs can be effectively improved by increasing δ to alleviate the strain localisation and increase the shearing propensity versus cracking. This can be readily achieved by increasing ν through designing the alloy compositions. Indeed, we have recently developed universal methods and relations capable of precisely predicting and designing the elastic constants of MGs [36]. The present thermodynamic criterion may aid in the development of MGs with improved toughness/ductility.

Finally, it is noted that the deformation and fracture behaviours of MGs are typically asymmetric under tensile and compressive conditions, which can be attributed to the normal stress effect [37]. Yet, we anticipate that the criterion presented here may be broadly applied to MGs because no items related to the stress state are involved in the thermodynamic analysis and calculations. Furthermore, it should be noted that the toughness/ductility and DTB transition of MGs can be affected by multiple factors, both intrinsic and extrinsic, in addition to the Poisson's ratio. For instance, a reduction in the free-volume content induced by thermal annealing can result in the embrittlement of MGs [12,38]. In contrast, the introduction of microstructural heterogeneities may help improve the ductility/toughness [39]. In terms of the extrinsic factors, the ductility/toughness of MGs may be degraded by defects introduced during processing, such as inclusions and porosities [40]. Therefore, the specific quantitative outcomes of the present study may be challenged when complex conditions are concerned. Nevertheless, the sensitivity of materials to extrinsic defects depends mainly on their intrinsic ductility/toughness. Thus, we anticipate that elucidation of the mechanisms underlying the relations between the Poisson's ratio and intrinsic toughness/ductility will greatly aid in the understanding of the deformation behaviours and mechanisms of MGs.

4. Conclusions

We have developed a universal thermodynamic criterion to describe the deformation modes of materials based on a newly introduced cooperation parameter δ, which quantifies the relative propensity of shear plastic deformation versus cracking. The criterion unravels from fundamental thermodynamics the underlying physics of the correlations between toughness and Poisson's ratio, the DTB transition and the various deformation modes of MGs in terms of the competition between shearing and cracking. The detailed deformation modes of MGs are classified into different types that can be quantitatively evaluated by the parameter δ. The validity of δ in describing various deformation modes and the propensity of strain localisation has been further verified qualitatively in other materials. The criterion has implications in understanding the deformation mechanisms and mechanical behaviours of MGs and may aid in the development of MGs with improved mechanical properties.

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