1. Introduction

Over the past two decades, bulk metallic glasses (BMGs) have attracted considerable attention because of their high strength and high elastic strain limit, as well as their advantage in shaping (via molding in supercooled liquid above glass transition temperature) [1,2]. For practical use of BMGs as structural components, fracture toughness, which characterizes the crack-propagation resistance of materials, is a vital indicator to consider in assessing their reliability in load bearing applications. For example, shafts transmitting torque (anti-plane shear) [3]. Under many complex loading conditions, cracking takes place on a plane oriented at an arbitrary angle with respect to the loading direction. Failure then occurs in a way of mixed mode fracture. Such events therefore require an understanding of material fracture toughness under loading conditions other than the basic mode I.

In particular, mode III fracture is encountered in many mechanical components. For example, shafts transmitting torque typically experience mode III loading of crack-like defects or notched features [4]. In such cases, the mode III fracture toughness is of primary importance, as shown for certain high-strength alloys [5–15], ceramics [16,17] and polymers [18,19]. Note that there is no well-defined correlation between the mode I fracture toughness and the mode III fracture toughness. For example, in the case of brittle alumina and polymer PMMA, the mode I fracture toughness, $G_{\text{IC}}$ (kJ/m$^2$), was reported to be about seven times and three times greater, respectively, than the mode I fracture toughness, $G_{\text{IC}}$ (kJ/m$^2$) [17,18]. However, for the Mn-containing 2034 aluminum alloys, when $G_{\text{IC}}$ increased from 16 kJ/m$^2$ to 121 kJ/m$^2$, the $G_{\text{IIIC}}/G_{\text{IC}}$ value decreased from 2.8 to 0.4 [12]. In other words, it is impossible to predict the mode III fracture toughness of a material simply based on its known mode I fracture toughness.

For BMGs under bending load, crack propagation possibly involves both mode II and mode III components, at least at the onset of crack extension. This was confirmed by Menzel et al. [20] in a study of the initial stage of fatigue crack growth in Vitreloy 1. Moreover, shear band formation is associated with collective shearing of atoms in a shear transformation zone (STZ) [21]. Since local dilation is required for STZ operation, normal or hydrostatic stresses necessarily play a role for the incipient flow and shear banding process [22–25]. Under mode I loading, a plastic zone forms ahead of the crack tip, due to numerous shear banding events under the constraint of a highly localized triaxial stress field. This contributes to the strain and fracture energy release as well as crack arrest. However, in the mode III situation, hydrostatic stress ahead of the crack tip becomes zero; it is expected that shear banding behavior in the plastic zone would then be quite different.
Consequently, it is clearly of interest to learn about the fracture behavior of BMGs under mode III loading.

The vast majority of studies conducted previously on the fracture toughness of BMGs focused on the mode I condition, with only a handful on mode II [26,27]. The mode I fracture toughness of BMGs has been found to depend strongly on alloy chemical composition and processing history, and varies over a wide range (spanning two orders of magnitude), from 2 MPa√m [28], which is as brittle as oxide glasses, up to 200 MPa√m [29], which is comparable to the toughest commercial alloys. Flores et al. [30] investigated the mode II fracture toughness of Vitreloy 1 BMG, showing that the \( K_{IC} \) is 75 MPa√m, a factor of -4 larger than the mode I fracture toughness. It suggests that the normal or mean stress played a significant role in the deformation process at the crack tip [22]. In contrast, Tandaiya et al. [31,32] found that the \( J_{IC} \) value of Vitreloy 1 was about three times higher than \( J_{IC} \). A similar finding was presented by Narayan et al. [33], who reported that the \( J_{IC} \) is about 2.5 times higher than \( J_{IC} \) for the \( Zr_{63}Ti_{32}Cu_{8.25}Be_{26.75} \) BMG. These limited findings suggest that the fracture toughness of BMGs can be highly dependent on loading mode.

Studies on mode III fracture toughness of BMGs are very few and far between, apart from several early studies using metallic glass ribbons by tearing the "trouser" specimens [34–37]. Such a tearing test probably does not reveal the intrinsic material property because the measured fracture energy was found to increase with increasing specimen thickness. With the BMGs, Varadarajan et al. [38] studied the mixed mode I/III fracture toughness, showing that the Vitreloy 1 exhibited significant increase in fracture energy under mixed mode loading. However, pure mode III loading was not achievable in their three-point bending tests. Thus, to our knowledge, mode III fracture toughness of BMGs remains an open question up to now.

In this work, a recently-developed \( Zr_{61}Ti_{13}Cu_{25}Al_{12} \) (denoted as ZT1) BMG with high mode I fracture toughness \( (K_{IC}) \) has been shown to reveal its intrinsic mode III fracture toughness based on both linear elastic fracture mechanics (LEFM) and elastic-plastic fracture mechanics. Moreover, fractography characterization has revealed crack-propagation micromechanism under mode III loading. Finally, the correlation between \( K_{IC} \) and \( K_{IC} \) of the BMGs is discussed, together with a comparison with available data of other engineering materials.

2. Experimental

2.1. Specimen preparation

The \( Zr_{61}Ti_{13}Cu_{25}Al_{12} \) (in atomic percentage, hereafter referred to as ZT1) BMG has robust glass-forming ability and high damage tolerance, providing the opportunity to reveal its intrinsic mechanical properties irrespective of the artifacts induced by confinement, as summarized in Table 1. Using elemental metals with purity better than 99.9 wt. % as starting materials, the master ZT1 alloy ingots were prepared by arc melting under a Ti-gathered argon atmosphere. The alloy ingots were re-melted several times to ensure compositional homogeneity. The BMG rods with a diameter of 7 mm and length of 105 mm were fabricated via copper mold casting of the re-melted master alloy. The amorphous nature of the rods was confirmed via X-ray diffraction (XRD, not shown here).

Mode III fracture toughness measurements were performed using circumferentially-notched cylinder specimens loaded under torsion, following the protocol widely used for many engineering materials [5–13,16–18]. Specimens were machined from the as-cast BMG rods, with final dimensions as shown in the schematic diagram in Fig. 1.

To produce annular concentric pre-crack at the machined V-notch root, specimens were fatigue pre-cracked in an Instron R.R. Moore rotating beam fatigue testing system under a load of 6.5 kg and rotation speed of 10 rpm. Samples were fatigued under several desired numbers of cycles in a range of \( 10^4 – 10^5 \), to achieve the desired depth of the pre-crack, which are required to be free of the strain-field effect at the notch tip during subsequent torsional tests. The minimum required pre-crack length in the mode III case was calculated based on a method proposed by Suresh et al. [44], as described as follows.

Considering an elastic cylindrical rod containing a circumferential semi-circle-shape notch with radius of \( \rho \) loaded under torsion, and a crack of length \( c \) emanating from the notch, the torsion loading causes a far-field anti-plane shear stress of \( \tau_{\infty} \). When \( c \ll \rho \), the stress intensity factor at the tip of the crack is given by

\[
K_t = k_t \tau_{\infty} \sqrt{\pi c}
\]

(1)

where the \( k_t \) is the elastic stress concentration factor of the notch under torsion. Note that in this Suresh's analysis, a free surface correction factor 1.12 was introduced for the calculation [44]. Under the current situation, however, the free surface correction factor is taken as 1 according to the calculation by Tada et al. [45]. If the crack length \( c \) is sufficiently long to exclude the influence of the notch tip field, i.e., for \( c \gg \rho \), the stress intensity factor ahead of the crack tip is given by

\[
K_c = F \tau_{\infty} \sqrt{\pi (\rho + c)}
\]

(2)

where the \( F \) is a dimensionless function of specimen geometry. The transition crack size \( c_0 \), which can be interpreted as the extent of the local notch field, is obtained by equating Eqs. (1) and (2):

\[
c_0 = \frac{\rho}{k_t \sqrt{F}} - 1
\]

(3)

For our specimen geometry, the stress concentration factor \( k_t = 2 \) [46], \( F = g(r) \cdot r^{1/2} \), where

![Fig. 1. Schematic diagram of cylinder specimen dimensions, with a V-60° notch and machined-notch root radius of ~0.03 mm.](image-url)
and \( e = a/b \) (\( a \) and \( b \) are defined in Fig. 2).

With Eqs. (3) and (4), the minimum required pre-crack length \( c_0 \)
calculated to be \(-10 \mu m\), which amounts to a small fraction of the
notch root radius \( \rho \) (\(-30 \mu m\)). Similarly, under mode I loading
conditions, the minimum fatigue pre-crack length is also required
to be much less than the notch root radius (in the range of \( \rho/20 \) to \( \rho/ 
2 \)) [44].

The pre-cracked specimens were subjected to quasi-static
torsional fracture on an Instron 8874 testing machine at a rota-
tion speed of \( 1^\circ/\text{min} \). The real length of the pre-crack was measured
under a scanning electron microscope (SEM) observation. On the
fracture surface, at least six typical sites were selected with an int-
erval of \( 60^\circ \) distributed along the circumferential direction, and
the attained average value was used to calculate the mode III
fracture toughness. The samples were invalid and abandoned if
their pre-cracks were either shorter than \( c_0 \) (the minimum required
pre-crack length) or eccentric. Six samples with pre-crack length,
c of 33, 37, 74, 348, 511 and 515 \( \mu m \) were used to determine mode III
fracture toughness. The pre-crack depth for all specimens was
required to be larger than the above-calculated \( c_0 \) to ensure the
validity of the data.

2.2. Linear elastic fracture mechanics analysis

For the circumferentially pre-cracked cylindrical rod, the mode
III stress intensity factor, \( K_{III} \), under small-scale yielding conditions,
is given by Ref. [45].

\[
K_{III} = \frac{2T}{\pi a} \sqrt{\pi a} \cdot \sqrt{1 - e \cdot g(e)} \tag{5}
\]

where \( T \) is the torque.

For an elastic perfectly-plastic material, the mode III plastic zone
size ahead of the crack tip is given by Ref. [47,48].

\[
r_y = \frac{1}{\pi} \left( \frac{K_{III}}{\tau_y} \right)^2 \tag{6}
\]

where the \( \tau_y \) is the shear yield strength. The validity of linear elastic
fracture mechanics (LEFM) requires that the plastic zone size, \( r_y \),
should be much less than the size of uncracked ligament, \( r_p \) [6],
typically between 1/12 and 1/15, as depicted in Fig. 2.

2.3. Elastic-plastic fracture mechanics analysis

Under large scale yielding condition of mode III loading, the
concepts of crack-tip displacement \( CTD_{III} \) or \( J \)-integral is difficult to
be applied to the case of circumferentially-cracked cylindrical
specimens, either computationally or experimentally. As such, a
new parameter termed the plastic strain intensity factor \( \Gamma_{III} \) was
used as a measure of fracture resistance under elastic–plastic
conditions [6,8,49,50]. This parameter is described as follows.

The distribution of plastic strain in the plane of the crack in a
circumferentially-cracked solid cylinder subjected to the torque \( T \)
can be expressed as

\[
\gamma = \frac{\tau_y}{\mu} \left( \frac{r}{r_p} \right)^2 \left( \frac{a - r_p}{a - r} \right) \tag{7}
\]

where \( \gamma \) is the plastic strain, \( \tau_y \) is the shear yield stress, \( \mu \) is the
shear modulus, and \( a, r, r_p \) are radii defined ahead of the crack tip in
Fig. 2. The plastic strain intensity factor \( \Gamma_{III} \) defined and expressed
in terms of the plastic zone is

\[
\Gamma_{III} = \lim_{a - r \rightarrow 0} \gamma(a - r) = \frac{\tau_y}{\mu} \left( \frac{a}{r_p} \right)^2 (a - r_p). \tag{8}
\]

Computation of \( \Gamma_{III} \) involves an analytical expression for \( r_p/a \), as
given in Ref. [49].

\[
\frac{r_p}{a} = \left[ 1 - \left( \frac{4}{3} \frac{T}{T_L} \right)^2 \left( \frac{a - r_p}{a} \right)^2 \left( 1 + \frac{5}{4} \left( \frac{4}{3} \frac{T}{T_L} \right)^2 \right) \right]^{1/6}
- \left( \frac{T}{T_L} \right)^6 + \left( \frac{T}{T_L} \right)^6 \sqrt{\frac{4}{1 - \frac{T}{T_L}}} \tag{9}
\]

where \( T_L = (2/3)\pi r_o a^2 \). Note that the Eq. (9) gives the expression for
the plastic zone size for polycrystalline von-Mises type materials
which are pressure insensitive and plastically incompressible.
However, BMGs are known to be pressure or normal stress sensitive
and plastically dilatant [2]. Regarding this consideration, we also
calculated the \( r_p \) and \( \Gamma_{III} \) value based on Mohr–Coulomb criterion
instead of von-Mises criterion. Then, it was shown that the \( \Gamma_{III} \)
value attained from Mohr–Coulomb criterion is at most 1.6%
greater than that from von-Mises criterion. Thus, the influence of
normal stress on the plastic deformation ahead of the mode III
crack tip is very marginal and can be ignored.

In this work, we choose \( \Gamma_{III} \) as a measure of the fracture resis-
tance under elastic–plastic mode III loading conditions for two
reasons. Firstly, the physical meaning of the \( \Gamma_{III} \) is well-defined
in terms of the crack-tip displacement, \( CTD_{III} \). In the case of small-

scale yielding, the relationship between \( \Gamma_{III} \) and \( K_{III} \) is [6,8]:

\[
CTD_{III} = 2\Gamma_{III} = \frac{2K_{III}^2}{\pi \mu \tau_y}. \tag{10}
\]

Such a relationship is substantially unchanged even under large-

scale yielding condition [8]. Secondly, as proven in a number of
previous studies [6,8,50,51], the \( \Gamma_{III} \) parameter is reliable to char-
acterize the fracture of crystalline alloys under either cyclic or
quasi-static mode III loading conditions.

2.4. Excluding frictional effects from fatigue pre-crack surfaces

Different from the mode I loading condition, the pre-crack easily
keeps closed under successive torsional loading. Then, frictional
abradion generated by asperities on mating pre-crack surfaces can
introduce an artifact to the apparent cracking resistance. As

\[
g(e) = \frac{3}{8} \left( 1 + \frac{e}{2} \right) \frac{3}{8} \left( \frac{5}{16} \right) \frac{3}{8} \left( \frac{35}{128} \right)^4 + 0.208 \cdot \frac{5}{5} \tag{4}
\]
observed for a 4340 steel previously [6], apparent fracture resistance exhibited a fourfold increase upon extending the pre-crack depth from 0.2 mm to 1.2 mm. To quantitatively determine such a contribution from abrasion, it is necessary to know the friction coefficient, \( f \), between mating crack surfaces. Unfortunately, directly measuring the \( f \) value is a challenge proposition. To resolve this issue, we measured the \( f \) value of the ZT1 BMG on CETR UMT-III Multi-Specimen Test System. Pin-on-plate approach of reciprocating sliding was taken, with a pin-specimen of 6 mm in diameter on a square ZT1 plate of 10 mm \( \times \) 10 mm. The surfaces of the pin and plate were mechanically grounded with SiC abrasive papers down to 2000 grit before tests. In this way, the counterpart surface roughness is approximately comparable to that of mating fatigue pre-crack surfaces. Then, the \( f \) was determined based on a relation of \( f = P/N \), where \( P \) is the measured friction force and \( N \) is the applied normal force (10 N, here).

2.5. Characterization of stable crack propagation

To interrogate stable mode III crack propagation during applied loading, two pre-cracked specimens were loaded to desired extent of crack growth prior to failure, and subsequently unloaded and subjected to a heat-tinted treatment at 300 °C for 60 min in air. These specimens were re-fatigued under a tension—tension loading of 160 N—1600 N at a frequency of 30 Hz until ultimate fracture. Fracture surfaces of the specimens were observed in Zeiss Supra 55 SEM and a 3-Dimensional LEXT OLS4000 laser optical microscope (LOM).

3. Results

3.1. Fracture initiation toughness

Fig. 3 displays representative torque twist plots, for three specimens with circumferential fatigue pre-crack depth, \( c \), of 33, 348 and 511 \( \mu \)m, respectively. The slight deviation from \( T-\theta \) linearity at the initial stage under applied loading is ascribable to the abrasion and frictional locking between the mated pre-cracked surfaces. On further loading, significant serrations appear until final failure occurs. These serrations are physically attributed to the occurrence of stable mode III crack propagation, which will be discussed in detail in Section 3.3.

As recommended by the ASTM-E 399 standard [52], when determining the mode I fracture toughness, the \( K_{IQ} \) can be defined from the 5% secant in a load-crack opening displacement (COD) curve. With a parallel consideration, the 5% secant method was also adopted to the \( K_{IIQ} \) cases [5,53]. Note that in mode I loading, the selection of 5% secant is to avoid the contributors from plastic zone size of the specimen. For a compact tension specimen, for example, the 5% secant corresponds to a plastic zone size that is 2% of \( W \) [54]. However, this is no longer adequate to the determination of \( K_{IIQ} \) because a one-to-one correlation between the plasticity extent and the secant is uncertain. Additionally, it is inconclusive to define the initial slope owing to a significant nonlinearity in the curve, as shown in Fig. 3. Since the stable mode III crack propagation is responsible for the serrations in the plot, it is physically reasonable to define the point where obvious serration starts as the onset of crack extension, as marked by \( T_Q \) in the inset in Fig. 3. This approach was used in this work to determine the mode III fracture initiation toughness.

Fig. 4 shows a plot of the \( K_{IIQ} \) of ZT1 BMG calculated in terms of Eq. (5), with several fatigue pre-crack lengths used in our experiments. The \( K_{IIQ} \) of ZT1 BMG was determined to be \( 30.3 \pm 1.4 \) MPa\( \sqrt{m} \), insensitive to the change of pre-crack length (\( c \)) at least in the range of 33—515 \( \mu \)m. Using Eq. (6), the corresponding plastic zone size was estimated to be approximately 320 \( \mu \)m. The LEFM theory requires that the ligament radius of specimen must exceed 12 times its plastic zone size. As such, the minimum ligament radius in our cases should be about 3.84 mm. This suggests that the LEFM theory is no longer valid in the current situation because the ligament radius of all the specimens we used was smaller than 2.4 mm. We therefore need to take crack-tip plasticity into account. The plastic strain intensity factor \( \Gamma_{III} \) was therefore used instead, to assess the mode III fracture toughness of ZT1, as shown in Fig. 4. Despite of some scatter in data, the trend is obvious that a higher \( \Gamma_{III} \) is associated with a larger fatigue pre-crack length. The \( \Gamma_{III} \) value increased by 42% as the \( c \) was enlarged from 33 \( \mu \)m to 515 \( \mu \)m.

This pre-crack length effect on cracking resistance may be related to the frictional sliding on mating pre-crack surfaces, which brings in an additional frictional torque, \( T_R \), in the opposite direction of the applied torque. This \( T_R \) plays a role to reduce the intrinsic fracture driving force, yielding a higher apparent \( \Gamma_{III} \), which would
increase with increasing crack surface contact area. In fact, this phenomenon was encountered also in crystalline alloys under mode III loading [6,55,56]. To exclude this effect, Gross [57] proposed a frictional model for calculating the magnitude of \( T_r \),

\[
T_f = \left( \frac{f \mu t}{1 - \nu} \right) \left[ \frac{c^2}{2} - 2ca + a^2 \ln\left( \frac{c}{a} \right) \right]
\]

(11)

where \( f \) is the frictional coefficient, \( \mu \) is the shear modulus, \( \nu \) is the Poisson’s ratio, \( t \) is theasperity height, \( c \) is the pre-crack length, and \( a \) is the radius of unbroken ligament. In the current situation, the height of asperities on pre-crack surface, \( t \), is of the order of 0.8 \( \mu \)m as measured with LOM, and \( f \) is measured to be approximately 0.2 as described in Section 2.4. Combining these data with Eq. (11), the \( I_{IIc} \) values were modified in Fig. 4. However, this model does not work well to eliminate the effect of pre-crack length. This is because only asperity height was taken into account in Eq. (11), without considering the influence of the wavelength of the asperities and the geometric interlock resistance between the mating asperities [55,56].

Therefore, to exclude pre-crack abrasion induced artifacts, we resorted to a method proposed by Tscheug et al. [6]: the intrinsic mode III fracture toughness is defined as the intercept with y-axis when extrapolating the pre-crack length to zero in the \( I_{IIc} \) vs \( c \) curve. As shown in Fig. 4, this extrapolation gives \( I_{IIc} = 29 \mu \)m, which reflects the intrinsic fracture initiation toughness of ZT1. Plugging our result of \( I_{IIc} = 29 \mu \)m into Eq. (10), the \( K_{IIc} \) of ZT1 would be of the order of 51 MPa\( \sqrt{m} \). Using Eq. (9), the plastic zone size ahead of the crack tip is then ~0.54 mm. It is noteworthy that when extrapolating the pre-crack length to zero in the intrinsic fracture toughness tests where a deformation of less than \( \pm 10^6 \) for the fatigue pre-crack to notch plane is acceptable [52]. It is interesting to observe that no remarkable crack deflection took place in the course of mode III crack growth, as shown in Fig. 7. This is different from the scenario of mode I fracture of the ZT1, where crack extension significantly deflected from the pre-crack plane [40].

3.2. Fractography observation

Fig. 5 displays a low-magnification SEM image showing the global appearance of the fracture surface of a specimen. An annular concentric pre-crack, ~348 \( \mu \)m in length, is seen in front of the notch root. There is also a clear contrast and well-defined boundary between the pre-crack and final fracture surface. For the latter, the crack propagation left three typical regions, denoted as I, II and III, from the outer border with the pre-crack inwards towards the center of the specimen. The enlarged views of the three boxed regions are shown in Fig. 6 (a), (c) and (d), respectively. Typical fatigue striations are present on the pre-crack surface, as shown in Fig. 6 (a). The flattening and elongation features along the twist direction on pre-crack surface reveal occurrence of frictional-sliding events under torsional stressing. Such events are responsible for the nonlinearity of the twist plot at initial loading stage and an additional extrinsic fracture resistance. The mode III fracture surface exhibits flat and vein pattern regions, denoted as A and B, respectively. Fig. 6 (b) shows a zoom-in image of the boxed region in (a), in which numerous parallel lines are present at an angle inclined by ~45° with respect to the shear stress direction, with a spacing of ~4 \( \mu \)m. This suggests that the mode III crack growth initiates in a stick slip manner, with discrete steps. With further crack advances, vein patterns are generated in region B, as well as in Fig. 6 (c). The ridges of veins are stretched along the shear stress direction, suggesting that shear banding events are involved in mode III fracture. As shown in Fig. 6 (d), dimple-like patterns in the center of fracture surface suggest that the final unstable fracture is dominated by cavitation under transient tensile stresses.

Fig. 7 shows a LOM 3D image of the fracture surface. It is observed that the fatigue pre-crack deviates slightly away from the initial notch bisection plane at an angle of ~4°. Such a minor deflection of pre-crack is negligible, in comparison with the mode I fracture toughness tests where a deflection of less than \( \pm 10^6 \) for the fatigue pre-crack to notch plane is acceptable [52]. It is interesting to observe that no remarkable crack deflection took place in the course of mode III crack growth, as shown in Fig. 7. This is different from the scenario of mode I fracture of the ZT1, where crack extension significantly deflected from the pre-crack plane [40].

3.3. Stable mode III crack growth

To reveal the path of stable crack propagation, two unloaded and subsequently heat-tinted samples were used; see Fig. 8 for the loading curve they experienced. Sample A was unloaded in the stage with obvious serration, whereas sample B was unloaded when the applied torque reached a visible plateau. These two sample were heat-tinted, and then fractured by tension—tension re-fatigue. Fig. 9 (a)–(d) show the LOM fractography images of these two samples. Fig. 9 (a) is a low-magnification view of the fracture surface of sample A. The fatigue pre-crack surface and the stable mode III crack surface are blue in color, whereas the final re-fatigue fracture surface is seen as the inner grey region. Fig. 9 (b) shows a zoom-in image of the boxed region in (a). It is noteworthy that the fatigue pre-crack surface is clearly distinguishable from the mode III fracture one, in terms of a major difference in heat-tinted blue colour, because fatigue striations are only present on the pre-crack surface. The mode III crack extension initiated at some favorable sites along the pre-crack front, and the initially straight front line evolves into a microscopically tortuous one, significantly different from the case of mode I loading [40]. Correlating with the loading curve of sample A, it is confirmed that the starting point of serration on the twist plot does correspond to the onset of mode III crack extension. This further supports that it is physically reasonable to define such a starting point for the calculation of the mode III fracture initiation toughness, as presented in Section 3.1.

As shown in Fig. 9 (c) for sample B, the subsequent crack propagation also moves forward in an inhomogeneous way. The crack extension at some sites has reached ~120 \( \mu \)m, but only at a level of 20 \( \mu \)m at other sites. Fig. 9 (d) displays a LOM zoom-in image of the box in (c). A number of parallel groove-like line patterns are visible at an angle of about 45° relative to the global shear-stress.
direction, with a spacing of around 3 μm. This is consistent with the finding in SEM observations, as shown in Fig. 6(b). It indicates that these regions, shown as the flat region in Fig. 6(b), are generated by stable crack propagation. In contrast, the vein pattern regions are associated with catastrophic fracture. The vein pattern suggests meniscus instability inside a zone with shear-banding-induced softening at the advancing crack tip.

4. Discussion

4.1. Phenomenological model of mode III crack propagation

Based on our findings above, the mode III fracture of ZT1 BMG consists of three stages: crack initiation (onset of crack extension), stable crack propagation (growth) and final catastrophic fracture. Such a process is schematically depicted in Fig. 10.

As the torsional loading increases, the mode III crack initiates from the pre-crack front at some favorable sites. The advancement of the initially straight crack front appears microscopically tortuous, in a zigzag pattern as shown in Fig. 10(a). This morphology develops because, under globally mode III loading on the specimen, the shear stress ahead of the crack tip, \( \sigma_{\text{global}} \), is parallel to the pre-crack front line. As such, the front is not being driven to march forward if it stays as a straight line [59]. Microscopically, however, for a slanted segment created via fluctuation,
the $\tau_{\text{global}}$ can be resolved into two components, $\tau_{\text{II,local}}$ and $\tau_{\text{III,local}}$, which are normal and parallel to the crack segment in question, respectively, as seen in Fig. 10 (b). $\tau_{\text{II,local}}$ drives the segment to extend by $\delta$. For segments at $45^\circ$, this driving stress is reasonably large and the probability of such segments is rather high, hence inducing the zigzag front observed.

Based on the LEFM mode III crack-tip stress field [45], the global shear stress ahead of crack tip in the crack plane is expressed as

$$\tau_{\text{global}} = \frac{K_{\text{III}}}{\sqrt{2\pi r}}$$

(12)

where $K_{\text{III}}$ is the stress intensity factor for the main crack, $\psi$ is the angle between the crack segment and global shear stress, which is $-45^\circ$ as measured from Fig. 9 (d). $\tau_{\text{II,local}}$ and $\tau_{\text{III,local}}$ will induce local mode II and mode III stress intensity factors at the crack segment tip, $k_{\text{II}}$ and $k_{\text{III}}$.

$$k_{\text{II}} = \tau_{\text{II,local}} \sqrt{\frac{2\pi r}{\pi}} = K_{\text{III}} \sin(\psi)$$

(14)

$$k_{\text{III}} = \tau_{\text{III,local}} \sqrt{\frac{2\pi r}{\pi}} = K_{\text{III}} \cos(\psi)$$

(15)

The effective energy release rate ahead of the crack tip is given by

Fig. 9. Laser optical micrographs of the fracture surface of samples subjected to loading and unloading as shown in Fig. 8: (a) and (b) for sample A. (c) and (d) for sample B. (a) Global view of fracture surface under visible light mode. Blue-colored outer regions are the fatigue pre-crack surface and stable mode III crack growth surface. Inner grey portion is the fracture surface after tension-tension re-fatigue. (b) Zoom-in image of boxed area in (a), showing a region of incipient mode III crack growth at some favorable sites as marked by arrows. (c) Fracture surface of sample B, showing that mode III crack growth is quite inhomogeneous. (d) Zoom-in image of boxed area in (c) under laser mode, showing a number of groove-like patterns with an interspacing of $\sim 3 \mu m$. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 10. Schematics showing the mode III crack propagation mechanism on microscopic scale. (a) Inclined crack segments initiate from some sites at the pre-crack front. (b) Crack segment extends by a distance of $\delta$ under a locally mixed mode II/III stressing, producing a macroscopic crack growth by a step of $\Delta c$. Such crack advancement leaves behind a sliding mark on the crack surface, at an angle of $\psi$ with respect to the global shear stress direction. (c) Successive crack propagation steps generate a number of groove-like patterns in region A. When the specimen ligament is reduced to below a critical value, catastrophic fracture takes place, generating the vein pattern in region B.
I/III fracture of solids, and used to establish a relationship between toughness under estimated here. This should be kept in mind when designing energy release rate. Based on Eq. (16), the effective energy release rate is 81% of $G_{II}$ for plane stress. Even though such an analysis is based on the LEFM, it generally suggests that the microscopically tortuous crack path provides the main resistance to crack growth.

Furthermore, when a microscopic crack segment advances by a step of $\delta$, as evidenced by the left-over mark on the crack surface as described earlier, the main crack propagates by a distance of $\Delta c$, as seen in Fig. 10(b). With continued crack growth, multiple groove-like patterns are generated on the crack surface. When the specimen ligament is gradually reduced to below a critical level, catastrophic fracture takes over along the shear band ahead of the crack tip, which is evidenced by the formation of vein patterns on crack surfaces, as shown in Fig. 6(a).

It should be emphasized that in the current work, we have experimentally measured only the crack-initiation toughness of the BMG, which corresponds to the driving force under a situation that the crack extension distance approaches zero, $\Delta c \rightarrow 0$. Since the advancing crack front becomes zigzag, it is difficult to determine accurately the $\Delta c$ during crack growth. This also makes it impractical to determine the crack-growth toughness. In other words, the toughness contribution from crack-growth events is not incorporated into the measured mode III fracture toughness. In this sense, the mode III fracture toughness of the ZT1 BMG is somewhat underestimated here. This should be kept in mind when designing BMG components under mode III loading.

4.2. Correlation between mode I and mode III fracture initiation toughness

Eq. (10) has been used as a general relation in crystalline alloys [8], although it is not strictly valid under the condition of large-scale yielding. Plugging our result of $t_{IIIC} = 29$ $\mu m$ into Eq. (10), the $K_{IIIC}$ of ZT1 would be of the order of 51 MPa$\sqrt{m}$. This converted value is $\sim$40% higher than the $K_{IIIC} = 30.3$ MPa$\sqrt{m}$ calculated using LEFM. It implies that the energy dissipation by plasticity at the crack tip has a significant contribution to the intrinsic fracture resistance of the ZT1 BMG. Using the 130 MPa$\sqrt{m}$ as the mode I fracture initiation toughness of ZT1 BMG [40], the ratio of $K_{IIIC}$ to $K_{IC}$, $K_{III}/K_{IC}$, is found to be $0.39$.

Several criteria [17,38,60,61] were proposed for the mixed mode I/III fracture of solids, and used to establish a relationship between $K_{IIIC}$ and $K_{IC}$. Among these theories, the maximum energy release rate criterion and the minimum strain-energy-density factor criterion are representative.

4.2.1. Maximum energy release rate criterion (MERR-criterion)

Based on the Griffith theory, the maximum energy release rate theory suggests that solid fractures along the plane which maximizes the energy release rate [62]. Under mixed loading mode, the energy release rate, $G$, can be expressed as

$$ G = \frac{K_{II}^2}{E} + \frac{K_{III}^2}{2\mu} + \frac{2\mu \sin^2(\psi)}{E} + \cos^2(\psi) \cdot G_{III} $$

(16)

where $\mu$ is the shear modulus, $E$ is equal to $E$ (in plane stress) or $E/(1 - \nu^2)$ (in plane strain), and $G_{III} = K_{III}^2/2\mu$ is the nominal energy release rate. Based on Eq. (16), the effective energy release rate is $\approx 55\%$ of $G_{III}$ for plane strain. $K_{III}$ is equal to $\sqrt{(\kappa - 3\sin^2(\theta))}$ $\cdot$ $K_{II}$, which is a material property and hence an invariant for various loading mode combinations. However, this criterion assumes a self-similar crack growth, i.e. a planar crack is assumed to remain planar and maintain a constant shape as it grows. In fact, this is usually not the case for mixed-mode fracture. To address this problem, Chang et al. [60] proposed a modified MERR criterion to predict the fracture angles $\theta_f$ with respect to initial crack plane. The $\theta_f$ is calculated by setting

$$ G = \frac{1}{2\mu \cos^2(\theta_f/2)} \left( \frac{k + 1}{8} \left[ K_{II}^2 \left( 1 + \cos(\theta_f) \right) - 4K_{II}K_{III} \sin(\theta_f) + K_{III}^2 \left( 5 - 3 \cos(\theta_f) \right) \right] \right) $$

(18)

and the corresponding energy release rate $G$ is given by

$$ G = \frac{1}{2\mu \cos^2(\theta_f/2)} \left( \frac{k + 1}{8} \left[ K_{II}^2 \left( 1 + \cos(\theta_f) \right) - 4K_{II}K_{III} \sin(\theta_f) + K_{III}^2 \left( 5 - 3 \cos(\theta_f) \right) \right] \right) $$

(19)

where $\mu$ is the shear modulus, $K_{II} = 3 - 4\nu$ for plane strain, and $(3 - \nu)/(1 + \nu)$ for plane stress. According to Eqs. (18) and (19), $K_{II}/K_{IC} = \sqrt{(\kappa + 1)/2}$. When applied to the ZT1, this gives a ratio of 0.80, which is twice the $K_{II}/K_{IC}$ value found in our experiments ($0.39$).

4.2.2. Minimum strain-energy-density factor criterion (the S-criterion)

The local strain-energy-density ahead of the crack tip can be expressed as $dW/dv = S/r$, where $r$ is the distance from the crack tip, $S$ is the strain-energy-density factor defined as [63],

$$ S = \frac{1}{\pi} \left( a_{11}K_{II}^2 + 2a_{12}K_{II}K_{III} + a_{22}K_{III}^2 + a_{33}K_{II}^2 \right). $$

(20)

The coefficients are given by

$$ a_{11} = \frac{1}{16\mu} [(3 - 4\nu - \cos(\theta)(1 + \cos(\theta))] $$

(21)

$$ a_{12} = \frac{1}{8\mu} \sin(\theta) \cos(\theta)(1 - 2\nu) $$

(22)

$$ a_{22} = \frac{1}{16\mu} [4(1 - \nu)(1 - \cos(\theta))(1 + \cos(\theta)(1 - 2\nu)) $$

(23)

$$ a_{33} = \frac{1}{4\mu} $$

(24)

where $\theta$ is measured from the line in front of the crack tip in the counter-clockwise direction and centered at the crack tip. The S-criterion assumes that (i) crack propagates in the direction where $S$ possesses a minimum value $S_{\min}$, and (ii) crack initiates when $S_{\min}$ reaches a critical value $S_{cr}$, which is a material constant and can serve as an indication of the fracture toughness of the material [64]. Using the S-criterion of $dS/d\theta = 0$ and $S_{\min} = S_{cr}K_{II}/K_{IC} = \sqrt{(\kappa + 1)/2}$, the predicted $K_{II}/K_{IC}$ is then 0.52 for the ZT1 BMG, which is 33% too large when compared with our experimental value.
4.2.3 Factors responsible for the gap between the experimental and the predicted $K_{IIC}/K_{IC}$

It is noteworthy that the above-mentioned theories are based on an assumption that fracture resistance under mode I loading and mode III loading remains nearly the same, i.e. $G_{IC} = G_{IIC}$ for the MERR-criterion and $S_{IIIC} = S_{IIC}$ for the S-criterion. Unfortunately, it is not the case as indicated by our experimental data of $G_{IIC} = 43$ kJ/m$^2$ and $S_{IIC} = 4.9$ kJ/m$^2$, which are 24% of $G_{IC}$ (177 kJ/m$^2$) and 42% of $S_{IIIC}$ (11.8 kJ/m$^2$), calculated based on Eq. (17) and Eq. (20), respectively. In other words, the mode III fracture resistance for the ZT1 is much lower than that of mode I. As a result, it is not surprising that the experimental value of $K_{IIC}/K_{IC}$ is much less than that predicated by either the MERR-criterion or the S-criterion.

In addition, Flores et al. [30] proposes that the stress state ahead of the crack tip plays a role on the fracture resistance of BMGs. Under mode I loading, a mean tensile stress exists ahead of the crack tip[65]. With this picture, the fracture resistance under mode III loading could be larger than that under mode I loading for BMGs. This conjecture is not supported by our finding. Consequently, we suggest that there exist additional operating mechanisms which are effective to elevate mode I fracture toughness but is absent in mode III fracture. Some possibilities are discussed in the following.

First of all, one possible source may be ascribed to an effect of crack trajectory deflection. As is well known, the crack trajectory is synergistically controlled by two factors, i.e., it tends to extend along the direction of maximum mechanical driving force and the path of weakest microstructure resistance, or a compromise if the two paths are not the same but in competition [66,67]. In either MERR- or S-criterion, however, only the maximum mechanical driving force direction is considered (the maximum-G or minimum-S direction). Then, the crack propagation would happen along the initial crack plane (i.e. $\theta = 0^\circ$) under either mode I or mode III loading. As revealed in our experiments, however, the real crack path of ZT1 does not simply follow such a picture. Under mode I loading, the shear band direction at the crack tip, as the path of weakest microstructure resistance, overpowers the maximum mechanical driving force direction, thus resulting in a remarkable crack deflection and elevated apparent mode I fracture toughness [40]. In contrast, significant crack deflection does not take place in mode III fracture, as seen in Fig. 7. It implies that the mode III fracture resistance would be much lower than that under mode I. This brings about a value of $K_{IIC}/K_{IC}$ much less than that predicted by the criteria.

Secondly, additional source may arise from the crack-tip blunting effect. Based on elastic-plastic fracture mechanics, initially sharp pre-crack undergoes blunting under mode I loading owing to crack-tip plasticity [3]. It happens indeed either in some polycrystalline alloys [68] or in BMGs [29,69]. The blunting radius of $\rho_1$ is given by Ref. [30].

$$\rho_1 = \frac{1}{4} \frac{K_1^2}{\sigma_y E} \quad (25)$$

where $\sigma_y$ is the tensile yield strength. Taking the 130 MPa $\sqrt{m}$ as $K_{IC}$ of ZT1, the $\rho_1$ is estimated at a level of 27 $\mu$m using Eq. (25). Compared with the initially sharp pre-crack, such a significant blunting effect necessarily mitigates the stress concentration, and retards crack propagation. In contrast, no crack blunting occurs under mode III loading because the displacement ahead of crack tip is parallel to the crack front [48,70]. Even if the asperities on pre-crack surfaces could probably introduce a mode I-type crack tip opening displacement under torsional loading, its magnitude is rather small, maintaining at the order of asperity height [58], such as ~0.8 $\mu$m in this work. Thus, the crack tip tends to maintain its sharpness under mode III loading, which leads to easier crack propagation and lower fracture resistance in comparison with the scenario of mode I.

Moreover, the plastic zone size of the ZT1 BMG under mode III loading is estimated to be ~0.54 mm, which is 54% larger than that under mode $I$, ~0.35 mm as estimated from $r_p = 1/6\pi(K_{IC}/\sigma_y)^2$ with $K_{IC} = 130$ MPa $\sqrt{m}$. But in the case of mode III loading, a sizable plastic zone does not necessarily correlate with a higher toughness. As usual, the plasticity at the crack tip of BMG is directly related to interactions between shear bands [31]. Unfortunately, it is experimentally difficult to monitor the shear-bandning events ahead of mode III crack tip for the specimens in the current work. As a result, whether there are multiple shear bands and the role played by shear bands in the plastic zone at crack tip remain unknown.

Finally, for a purpose of comparison, the $G_{IIC}$ of several engineering materials is plotted against their $G_{IC}$, as shown in Fig. 11. The $G_{IIC}$ of ZT1 BMG is observed to be comparable to that of high-strength aluminum alloys. It is about four fold higher than that of some steels such as HF1 [53] and 30Cr2MoV [71], let alone PMMA polymer and alumina ceramics. However, the ZT1 BMG exhibits a relatively low $G_{IIC}/G_{IC}$ value, implying that it is more susceptible to mode III fracture with respect to mode I fracture. Together with recent studies on mode II fracture of BMGs performed by Ramamurty et al. [31–33], where the ratio of $J_{IIIC}$ to $J_{IC}$ is also in a range of 0.3–0.4, it is evident that the BMG materials with pre-existing cracks are intrinsically more susceptible to fracture damage, when subjected to either in-plane shear or anti-plane shear, more so than tensile (opening) loading. This newly-uncovered feature of BMGs is of significance as a design variable for structural components. The mode III fracture toughness can provide a more reliable baseline for design against cracking.

5. Concluding remarks

Using circumferentially pre-cracked cylinder specimens, the mode III fracture initiation toughness of the Zr61Ti2Cu25Al12 (ZT1) BMG, which has a high mode I fracture toughness, was measured...
without the interference from inter-pre-crack abrasion. Based on the elastic–plastic fracture mechanics, the plastic strain intensity factor $I_{III}$ was used as a measure to characterize the fracture resistance under elastic–plastic conditions. The intrinsic mode III fracture initiation toughness determined as such for the ZT1 BMG, $G_{IC}$, is 29 $\mu$m. This converts to a $K_{IC}$ of 51 MPa$\sqrt{m}$ and a plastic zone size of $0.54$ mm at the crack tip under mode III loading.

Under an anti-plane shear loading, the crack propagation in ZT1 proceeds via subcritical crack growth prior to catastrophic fracture. The crack extension microscopically produces a zig–zag front pattern, which generates a local mixed mode II/III loading ahead of the crack tip. This microscopic path mitigates the effective crack driving force during crack extension and may be partly responsible for the toughness observed.

The ZT1 BMG exhibits a relatively lower ratio of $K_{III}$/$K_{IC}$ and $G_{III}$/$G_{IC}$, approximately 0.39 and 0.24, respectively. It indicates that with respect to the mode I fracture, the ZT1 BMG has a lower crack resistance under mode III loading. In other words, it is more susceptible to anti-plane shear loading in contrast to the Mode I tensile opening loading. As such, mode III fracture is sicsically more susceptible to anti-plane shear loading in contrast to the Mode I tensile opening loading. As such, mode III fracture is.

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