Size-dependent fracture toughness of bulk metallic glasses

Bernd Gludovatz [a], Steven E. Naleway [b], Robert O. Ritchie [a,c], Jamie J. Kruzic [b,*]

[a] *Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*
[b] *Materials Science, School of Mechanical, Industrial, and Manufacturing Engineering, Oregon State University, Corvallis, OR 97331, USA*
[c] *Department of Materials Science and Engineering, University of California, Berkeley, CA 94720, USA*

Abstract

The fracture toughness is a critical material property that determines engineering performance. However, as is well known for crystalline materials, if certain sample geometry and size requirements are not met, test results become sample-size dependent and difficult to compare between different studies. Here, the room-temperature fracture toughness of the Zr-based bulk metallic glass (BMG) $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ (Vitreloy 105) was evaluated using compact-tension, as well as single-edge notched-bend, specimens of different sizes to measure K_{Ic} values according to ASTM standard E399 and J_{Ic} values according to ASTM standard E1820. It is concluded that the ASTM standard E399 sample-size requirements should be cautiously accepted as providing size-independent (valid) K_{Ic} results for BMGs; however, it is also concluded that small-sized samples may result in a wider scatter in conditional toughness K_Q values, a smaller yield of valid tests and possibly somewhat elevated toughness values. Such behavior is distinct from crystalline metals where the size requirements of ASTM standard E399 are quite conservative. For BMGs, K_Q values increase and show a larger scatter with decreasing uncracked ligament width b, which is also distinct from crystalline metals. Samples smaller than required by ASTM standards for K_{Ic} testing are allowed by the J-integral-based standard E1820; however, in this study on BMGs, such tests were found to give significantly higher toughness values as compared to valid K_{Ic} results. Overall, the toughness behavior of BMGs is more sensitive to size requirements than for crystalline metals, an observation that is likely related to the distinct size-dependent bending ductility and strain softening behavior found for metallic glasses. It is concluded that toughness values measured on BMG samples smaller than that required by the K_{Ic} standard, which are common in the literature, are likely sample size- and geometry-dependent, even when they meet the less restrictive valid J_{Ic} requirements.

1. Introduction

Bulk metallic glasses (BMGs) are a class of engineering materials with unique properties, such as near-theoretical strength, low stiffness and the ability to be thermoplastically formed into precision-shaped parts with complex geometries [1–5]. Despite their useful combination of properties, the fracture toughness of these materials can sometimes be low and a limiting factor when considering BMGs for structural applications. For example, some early glasses are known to fail in a highly brittle manner, with K_{Ic} values as low as ~2 MPa m$^{1/2}$ [6]. In stark contrast, recent developments in specific Pd-based and Zr-based glasses have shown multiple shear band formation, subcritical crack growth and increasing fracture resistance with crack extension (i.e. rising fracture resistance curve (*R*-curve) behavior), with reported fracture toughnesses of up to ~200 MPa m$^{1/2}$ [7,8]. While such very low and very high fracture toughness values are certainly extremes for brittle

* Corresponding author. Tel.: +1 541 737 7027; fax: +1 541 737 2600.
  E-mail address: jamie.kruzic@oregonstate.edu (J.J. Kruzic).

and tough fracture behavior of BMGs, most metallic glasses are reported to lie somewhere between 10 and 100 MPa m$^{1/2}$ [9–17]. However, there is often significant variability in the results, even within a single study. While some variability between studies may be explained by factors such as the use of notched vs. pre-cracked samples[1] [19,20], the influence of other parameters, like processing history and/or sample geometry, remain less clear.

A commonality among the very high toughness BMGs is the ability to form significantly more shear bands as compared to lower toughness glasses [7,8]. Conner et al. have shown a similar positive correlation between high numbers of shear bands and high ductility for Zr-based metallic glass plates subjected to bending [21,22]. While thick plates of BMG are well known to fail catastrophically in bending without significant plastic deformation, in the Conner et al. studies plates with thicknesses, t, of less than 1.5 mm were bent in a ductile manner around dies of different radii, r, showing increased shear banding (smaller shear band spacing, λ) and increased ductility prior to fracture with decreasing plate thickness. The relevant dimensions are shown in Fig. 1. This leads to the conclusion that BMG plates below a certain critical thickness can achieve the needed number of shear bands to demonstrate significant bending ductility. This thickness-dependent bending ductility is a property of BMGs that is distinct from crystalline metal alloys.

Due to the often limited glass-forming ability of many BMGs, standard products like rods and plates can often only be produced with diameters or thicknesses less than ~10–15 mm; hence, most bending tests are done on relatively thin rectangular plates or square bars. Furthermore, fracture toughness tests are often solely done on single-edge notched-bend (SE(B)) samples of relatively small dimensions. Although SE(B) samples are among the recommended specimen geometries in the ASTM standards for measuring the fracture toughness of materials (E399, E1820) [23,24], the size requirements found in those standards are based on the behavior of common crystalline metals, such as steel, aluminum and titanium alloys. Furthermore, the minimum size limitations of both the K_{Ic} E399 standard and the J-integral-based E1820 standard do not distinguish between BMG samples that are above or below a certain critical bending thickness. Also, J-calculations of plastic contributions by E1820 assume strain hardening while metallic glasses typically show local strain softening behavior in tension and compression with strain localization often in a single shear band [20,25–27]. As metallic glasses clearly show very different deformation behavior from crystalline metals, the question arises whether current ASTM standard sample-size restrictions can be applied to determine a sample geometry-independent
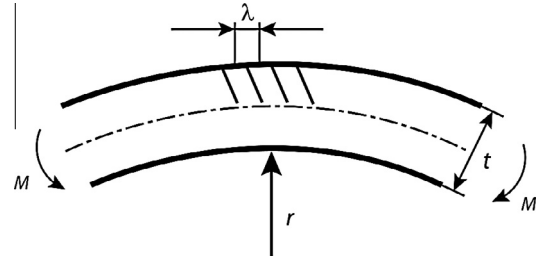

Fig. 1. Bending ductility of bulk metallic glasses. Conner et al. [21,22] have shown that BMG samples below a certain critical thickness, t, are capable of preventing catastrophic failure by the formation of multiple shear bands. The spacing of the shear bands, λ, decreases with increasing bending moment, M, and decreasing radius, r, leading to a more ductile behavior of the BMG.

measure of the fracture toughness for BMGs. Stated another way, are new sample-size requirements needed to account for the distinct size-dependent bending ductility behavior and strain softening behavior of BMGs?

To help answer these important questions, the present paper compares the fracture toughness of a $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$[2] bulk metallic glass using SE(B) samples with various sample sizes and compact-tension (C(T)) samples with dimensions well above the critical bending thickness of this material. Furthermore, results generated by applying the most stringent sample-size limitations of plane strain K_{Ic} testing, as dictated by ASTM standard E399, are compared with those that follow the less restrictive size criteria of ASTM standard E1820 for J-integral-based fracture toughness testing.

2. Background

The size requirements for a valid linear-elastic K_{Ic} test require that loading conditions are essentially elastic, i.e. that the crack-tip plastic zone size, r_y, is small enough to be ignored – at least an order of magnitude smaller than the in-plane dimensions of crack size, a, and uncracked ligament width, b – to guarantee a state of small-scale yielding with K as the appropriate description of the crack-tip field. Additionally, for a single-value characterization of toughness, a state of plane strain must prevail, which is achieved when r_y is at least an order of magnitude smaller than the out-of-plane dimension of the sample thickness, B. In the latter case, a recent study has demonstrated that plane stress conditions can lead to much higher fracture toughness values in BMGs [28].

Fig. 2 shows the relevant sample dimensions and, based on testing of various polycrystalline alloys (mainly steel, aluminum, and titanium alloys), the above considerations led to the empirically determined size requirements for K_{Ic} testing used in ASTM standard E399 [23]:

$$a, b, B \geqslant 2.5\left(\frac{K_Q}{\sigma_{YS}}\right)^2 \tag{1a}$$

---

[1] The effect of the notch root radius in artificially inflating the apparent fracture toughness of polycrystalline metals has been known since the 1970s [18]; however, this effect appears to be far more pronounced in BMGs [19,20].

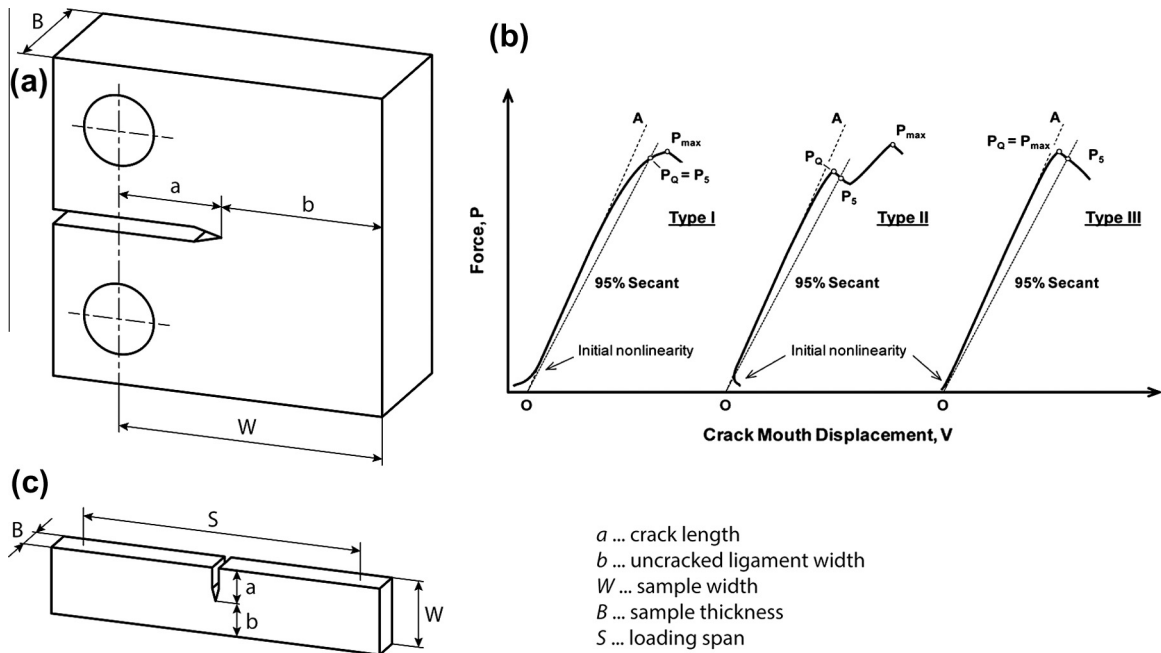[2] All compositions are given in terms of atomic percent.

Fig. 2. Tested specimen geometries, showing the nomenclature of their dimensions and failure types by ASTM standard E399 [23]. (a) C(T) specimens as well as (c) SE(B) specimens of $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ BMG were tested under monotonic loading in order to determine the fracture toughness of this material. (b) All samples failed catastrophically after either major plastic deformation – type I failure by ASTM standard E399 – or minor plastic deformation, thereby showing failure type III. Failure type II – a load-drop due to subcritical crack growth – did not occur for any sample in the present study. Panel (b) is adopted from the ASTM standard [23].

$$0.45 \leqslant a/W \leqslant 0.55 \qquad (1b)$$

where W is the sample width and σ_{YS} is the yield strength. For J_{Ic} toughness measurements the size requirements are relaxed somewhat, but must maintain valid J-field dominance ahead of the crack, with crack-tip zones of unloading and non-proportional loading that are small compared to the in-plane b dimension. The standard size requirement for J-integral-based fracture toughness testing found in ASTM standard E1820 [24] is:

$$B, b \geqslant M \frac{J_Q}{\sigma_Y} \qquad (2)$$

where σ_Y is now defined as the mean of the yield and ultimate tensile strengths, and for Prandtl-field geometries, i.e. C(T) and SE(B), M is an empirically determined coefficient in the range of 10–100, depending on whether the fracture occurs in a stable or unstable manner. While Eq. (1) is also found as the size requirement for plane strain K_{Ic} measurements in ASTM E1820, for clarity of differentiating the size requirements in this paper, ASTM E399 will be referred to exclusively when regarding the size requirements for plane strain K_{Ic} and ASTM E1820 exclusively when regarding the size requirements needed for J-integral-based toughness calculations.

ASTM standard E399 [23] distinguishes between three types of failure in a fracture toughness test based on the load–(crack mouth) displacement (P–V) curve, as shown in Fig. 2b. Type I describes a material which fails after significant plastic deformation; here, the intersection of the load with the 95% secant line (Fig. 2b) is used to

calculate a conditional fracture toughness, K_Q. Type II can be seen for a material that shows crack propagation ("pop-in") followed by a further increase in the load before catastrophic failure (Fig. 2b). Here, the peak load before crack propagation is used to calculate K_Q. Type III shows a material which behaves nearly linear-elastic, with just minor plasticity before catastrophic failure (Fig. 2b). In this case, the maximum load is used to calculate K_Q. Further details can be found in the standard [23].

3. Experimental procedures

Research-grade $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ (Vitreloy 105) BMG plates with nominal size $30 \times 30 \times 2.3$ mm³ were procured from Liquidmetal Technologies and machined into C(T) samples with nominal widths and thicknesses in the range of 20–22 and 2.0–2.3 mm, respectively (Fig. 2a). The faces of each sample were polished gradually to a 0.05 μm finish using silicon carbide grinding papers and alumina powders on cloth to allow accurate crack length measurements using optical microscopy. All samples were annealed at 300 °C (573 K) for 2 min in a flowing ultrahigh-purity nitrogen environment in order to relieve the residual stresses incurred by the thermal tempering during the casting process. These residual stresses have been shown to affect the fracture toughness and fatigue-crack growth behavior of Zr-based BMGs, and this stress relief procedure was used in previous studies without crystallization or structural relaxation [15]. It is important to note that this temperature is far below the reported glass

transition temperature of $T_g = 409–440\ ^\circ C$ [29]. The BMG samples were shown to be fully amorphous both before and after the annealing process by X-ray diffraction.

Prior to fatigue pre-cracking, the machined notches were extended by a razor micro-notching procedure. This was done by cycling a razor blade across the end of the machined notch in the presence of a 1 μm polycrystalline diamond compound to extend and sharpen the crack to a root radius of ∼5–10 μm. Subsequently, each C(T) sample had a 350 Ohm strain gage (Vishay Precision Group, Wendell, NC, USA) mounted on its back face. During fatigue pre-cracking, the crack size, a, was determined from the back-face strain gauge, as described in Ref. [30].

All C(T) samples were fatigue pre-cracked and tested to measure K_{Ic} values using a computer-controlled servo-hydraulic Instron 8501 mechanical testing machine (Instron Corporation, Norwood, MA, USA) in ambient air with a nominal test temperature of $23 \pm 2\ ^\circ C$ and relative air humidity in the range of 20–40%. Fatigue pre-cracks were created under load control using initial stress intensity ranges of $\Delta K = K_{max} - K_{min}$ of roughly 4–8 MPa m$^{1/2}$ at a constant frequency of 25 Hz (sine wave) with a load ratio $R = 0.1$, where R is the ratio of minimum to maximum applied load, P_{min}/P_{max}. Samples were fatigue pre-cracked in multiple increments of ∼0.5–1.0 mm and between increments the crack length was checked on both sides of the sample to ensure one straight crack grew from the notch until the crack length to width ratio, a/W, was ∼0.5 (Eq. (1b)). The length of the pre-cracks were well above 1.3 mm and within 10° parallel to the plane of the starter notch, as required by the standard. Final values of ΔK during fatigue pre-cracking were in the range of 4–5 MPa m$^{1/2}$. Four $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ BMG C(T) samples were prepared in this manner.

Fracture toughness tests were performed in accordance with ASTM standard E399 [23] under displacement control at a constant displacement rate of 1.33 μm s^{-1} ($\dot{K} \approx 0.12$ MPa m$^{1/2}$ s^{-1}). Load–displacement data were found to be linear to failure such that the value of K_Q was calculated from the peak load at fracture representing type III failure by the standard (Fig. 2b).

Beams for SE(B) fracture toughness tests were machined from both halves of the fractured C(T) specimens. Two sets of samples, in general accordance with ASTM standards E399 and E1820 [23,24], were made with different sizes: one set with thickness $B = 2$ mm, width $W = 4$ mm and loading span $S = 16$ mm, while the second set had $B = 2$ mm, $W = 2$ mm and $S = 8$ mm (Fig. 2c). All samples were gradually ground and polished to a 1 μm surface finish. A blunt notch was cut into each sample with a diamond blade and razor micro-notched as described above. Seventeen samples were pre-cracked in cyclic tension using the same parameters as for the C(T) samples. Twelve samples resulted in a/W between 0.45 and 0.55, as required by both standards for valid K_{Ic} testing; five samples had slightly longer cracks of $a/W \approx 0.55–0.7$, which comply with ASTM E1820 standard for J_{Ic} testing.

Additionally, two samples were pre-cracked in compression at 25 Hz frequency, with $R = 20$ and $\Delta K = 20–24$ MPa m$^{1/2}$; the achieved crack lengths of these samples were non-standard, with $a/W \approx 0.37–0.39$. Given the small sizes of the SE(B) samples the pre-cracks were shorter than the minimum requirement of 1.3 mm by the standard; however, in all cases their lengths were >1.5 times the notch root radius in order to be outside any stress concentration resulting from the notch [31,32]. In total, 19 pre-cracked SE(B) samples were prepared for testing.

Fracture toughness tests were performed using a Gatan MicroTest 2kN bending stage (Gatan, Abingdon, UK) in three-point bending at a displacement rate of 0.83 μm s^{-1} ($\dot{K} \approx 0.12$ MPa m$^{1/2}$ s^{-1}), using a loading span $S = 16$ mm for the larger samples and $S = 8$ mm for the smaller ones. Note that the displacement rates were different in the C(T) and SE(B) tests; this was done to maintain the $\dot{K}$ constant for both sets of experiments.

While all SE(B) samples failed catastrophically without significant subcritical crack propagation, two different modes of failure were observed: (i) samples that failed in a nearly linear elastic manner and (ii) samples that failed with a large amount of plastic deformation. In cases of (i), a conditional fracture toughness, K_Q, was calculated from the peak load (type III, Fig. 2b) and samples which met the size requirements for a linear-elastic, plane strain fracture toughness (Eq. (1)) along with the other standard requirements were then denoted as K_{Ic}. For the evaluation of the size requirements a yield strength, σ_{YS} of 1700 MPa was used [33]. If K_{Ic} size requirements were not achieved, the data were reanalyzed using ASTM standard E1820 for J_{Ic} testing with a fracture instability toughness. In those cases, there was no significant plastic contribution to J_Q and Eq. (2) was used as the size requirement with $M = 100$, as required for unstable fracture. In cases of (ii), K_Q was calculated from the load where the load–displacement curve intersected the 95% secant line; none of these samples qualified as valid K_{Ic} tests. Additionally, J_Q was calculated in accordance with ASTM E1820 to quantify both the elastic and inelastic contributions to the fracture toughness; however, none of them met the J_{Ic} validity requirements.

In order to evaluate the influence of the fatigue pre-crack, three SE(B) samples with respective B, W and S of ∼2 × 4 × 16 mm were prepared using the same procedures as described above but were not pre-cracked; the final notch root radii were ∼10–20 μm.

Finally, six samples with B, W and S of ∼2 × 4 × 16 mm, respectively, were prepared for in situ loading in a Hitachi S-4300SE/N (Hitachi America, Pleasanton, CA, USA) scanning electron microscope (SEM) using the Gatan MicroTest 2kN bending stage to observe shear band behavior at the crack tip. The samples were prepared using the same procedures as described above, with fatigue pre-cracking being carried out in tension. Loading was carried out at a displacement rate of 0.55 μm s^{-1} in steps and frequently arrested to collect images of the deformation

mechanisms at the crack tip. Due to the slower loading rate and the incremental loading, the fracture toughness results of these samples are not presented as results.

After all of the testing had been performed, the fracture surfaces were observed using an ASPEX EXplorer SEM (ASPEX, Delmont, PA, USA) for the C(T) samples and the above-mentioned Hitachi SEM for the SE(B) samples. Finally, statistical analysis of the data was done by performing an analysis of variance and Tukey's post hoc test to determine which means were statistically different from each other, with $p < 0.05$ being considered statistically significant.

4. Results

The results of all valid fracture toughness tests (K_{Ic}, J_{Ic}) are shown in Fig. 3 as a function of the uncracked ligament width, b ($= W - a$); for comparison purposes, J_{Ic} toughness values were represented as a stress intensity, referred to as K_{JIc} toughness values, using the standard mode I, linear-elastic, K–J equivalence relationship:

$$K_{JIc} = \sqrt{\frac{E J_{Ic}}{1 - v^2}} \qquad (3)$$

with a Young's modulus of $E = 85.6$ GPa and a Poisson's ratio of $v = 0.375$ [12].

Additionally, to show the data for all valid and invalid tests, K_{Ic}, K_{JIc} and K_Q values are plotted as a function of the ligament size, b, in Fig. 4a. Finally, a summary of all data is shown in Table 1, together with details such as the sample dimensions, and the critical sample thickness, B_{crit}, and the ligament width, b_{crit}, values needed to achieve a valid test by ASTM E399. Additionally shown is the type of failure based on the load–displacement curve categories of ASTM standard E399 and the pre-cracking method.
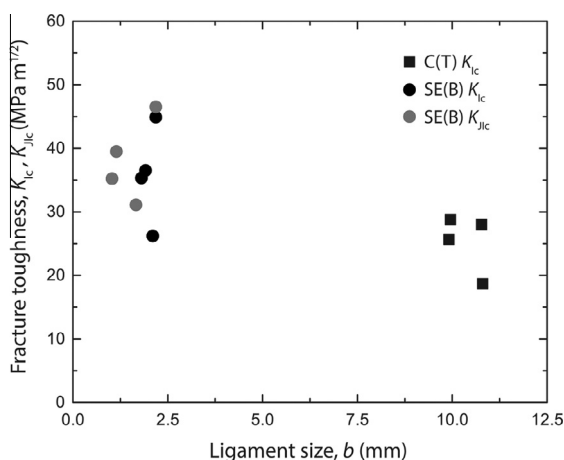

Fig. 3. Fracture toughness, K_{Ic}, K_{JIc}, data of $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ BMG as a function of the uncracked ligament width b. K_{Ic} values were measured in accordance with ASTM standard E399 [23] using both larger sized C(T) samples and SE(B) samples, whereas K_{JIc} values were measured in accordance with ASTM standard E1820 [24] using only using SE(B) samples. All results shown here met the valid sample size requirements of each of their respective standards yet still show a significant trend towards higher toughness values with decreasing uncracked ligament width, b.

Results are presented in this section as mean ± standard deviation. Samples with the largest ligament size, four C(T) samples with $b \sim 10$ mm, showed a fracture toughness K_{Ic} of 25.3 ± 4.6 MPa m$^{1/2}$; these results are valid by ASTM standard E399 and represent the linear elastic, plane-strain fracture toughness for the $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ BMG measured using C(T) samples.

All other tests were performed using SE(B) samples with ligament sizes of $b \approx 1$–2 mm. Twelve samples failed in a nearly linear-elastic manner with very minor plastic deformation, conforming to type III failure by ASTM standard E399. Five of them failed at relatively low loads, leading to K_Q values between 26.2 and 44.9 MPa m$^{1/2}$, so that the sample dimensions are sufficient for the measured numbers to qualify as valid K_{Ic}. One of those samples, however, had an a/W of 0.58 and hence cannot be strictly included as a valid K_{Ic} value (Eq. (1)). The remaining four values led to an average K_{Ic} of 35.7 ± 7.7 MPa m$^{1/2}$.

While the data point with $a/W = 0.58$ is invalid as K_{Ic} by ASTM E399, it is valid in terms of J_{Ic} measurements by ASTM E1820, which allows for a/W ratios between 0.45 and 0.7 and can hence be counted as a valid K_{JIc} result. Three more samples allowed for valid J_{Ic} measurements leading to $K_{JIc} = 39.0 \pm 7.0$ MPa m$^{1/2}$.

The other four samples that showed type III failure, as well as seven samples that failed as type I with significant amounts of plasticity, did not meet the requirements to qualify for either K_{Ic} or J_{Ic}. For type III samples the failure load was used to calculate K_Q, whereas for type I samples the load intersecting with the 95% secant line was used (Fig. 2b); the resulting K_Q for those samples was found to be 58.2 ± 16.1 MPa m$^{1/2}$.

Results of the statistical analysis of all valid data revealed a statistically significant ($p < 0.05$) correlation between the valid fracture toughness (K_{Ic}, K_{JIc}) and the ligament size, b, assuming a linear regression. However, when considering only the valid K_{Ic} data, the correlation just barely missed the criterion for significance ($p = 0.054$). Similarly, although the mean value of the valid K_{Ic} tests for C(T) and SE(B) samples appear different, no statistical difference could be proven by Tukey's test ($p > 0.05$). This is probably because the scatter increases as b decreases, and the two data sets overlap. Conversely, the mean value of the SE(B) tests that gave valid K_{JIc} values by ASTM standard E1820 was found to be even higher, and was statistically different ($p < 0.05$) from the mean K_{Ic} value for the C(T) samples.

The three SE(B) samples that were only notched but not pre-cracked had $b \approx 2$ mm and failed catastrophically in a nearly linear-elastic manner, showing type III failure by ASTM standard E399; their (apparent) toughness values were far higher, at $K_Q = 93.9 \pm 2.2$ MPa m$^{1/2}$.

Both samples with cracks smaller than $a/W = 0.45$ and larger than $a/W = 0.55$ did not seem to show a clear trend towards larger or smaller fracture toughness values. Additionally, no obvious difference between pre-cracking in tension or compression could be discerned.
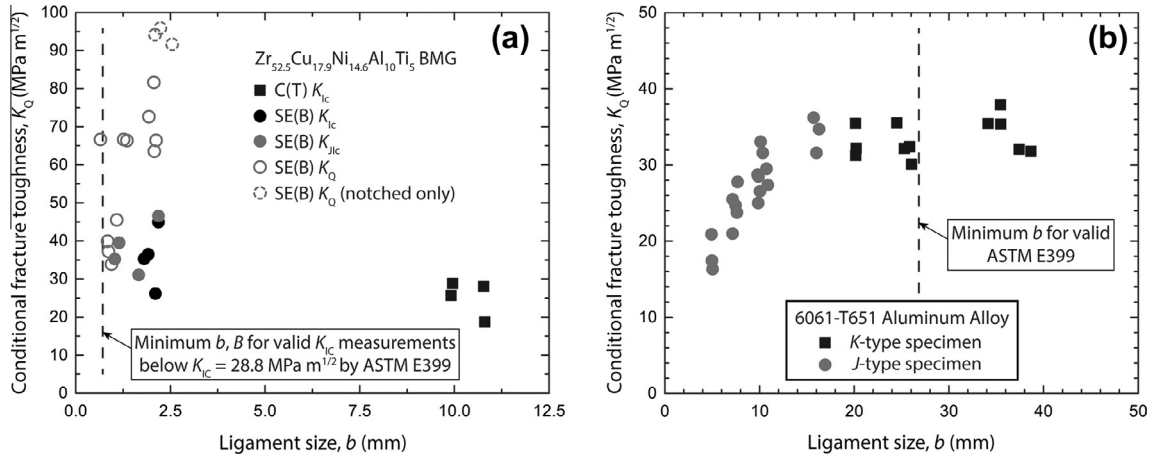
Fig. 4. Conditional fracture toughness, K_Q, data for (a) $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ BMG and (b) 6061-T651 (polycrystalline) aluminum alloy as a function of the uncracked ligament width, b. The metallic glass shows a clear trend towards increasing K_Q results with decreasing ligament size. This trend is contrary to the behavior of polycrystalline metals (e.g. 6061-T651 aluminum), which generally show decreasing K_Q data with decreasing b. The polycrystalline 6061-T651 aluminum alloy data were taken from Ref. [34].

Scanning electron microscopic examinations of the fracture surfaces showed no obvious defects associated with the different locations in the castings for the various samples. Both the C(T) and the SE(B) samples which yielded valid toughness values showed a relatively even crack propagation front and just minor shear banding ahead of the pre-crack (Fig. 5a). In comparison, the SE(B) samples which did not meet the requirements for either K_{Ic} or K_{JIc} failed, and showed significant plastic deformation, multiple shear band formation and blunting at the tip of the pre-crack (Fig. 5b). These samples also showed a clear trend to rougher fracture surfaces, bifurcations and significant deviations from a mode I crack path. Fig. 6 shows an in situ testing result for a sample that demonstrated a large amount of ductility, revealing extensive multiple shear banding from the crack tip and a large crack-tip opening displacement (CTOD) prior to crack extension and bifurcation.

5. Discussion

The mean fracture toughness of the $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ BMG was measured to be 25.3 MPa m$^{1/2}$ using four C(T) specimens. The crack length, a, the ligament size, b, and the sample thickness, B, of these samples are well above the minimum size requirement of Eq. (1) for both K-validity and plane strain conditions according to ASTM standard E399 [23]; the result can therefore be considered as the sample-size independent, linear-elastic, plane-strain fracture toughness of this Zr-based glass.

Testing the same material from exactly the same samples, but with the different SE(B) sample geometry, should therefore allow valid K_{Ic} measurements by ASTM standard E399 as long as a, b and B are > 0.72 mm based on Eq. (1a), with $K_{Ic} = 28.8$ MPa m$^{1/2}$, the highest result of all tested C(T) samples. Despite the fact that most SE(B) samples met this requirement (Table 1), only four SE(B)

samples with a, b and $B \sim 2$ mm showed valid fracture toughness tests according to ASTM E399 with $K_{Ic} = 35.7$ MPa m$^{1/2}$, a 41% increase as compared to the C(T) sample result. Another four samples, however, showed valid results using the J-based ASTM E1820 standard with a back-calculated fracture toughness of $K_{JIc} = 39$ MPa m$^{1/2}$; this represents an increase of more than 54% compared to the results of the valid K_{Ic} values measured with the larger C(T) samples, despite the fact that the K_{JIc} values met the J_{Ic} sample size criteria for validity.

Statistical analysis of these results demonstrated a statistically significant correlation between valid K_{Ic}/K_{JIc} values and ligament size. Furthermore, a statistically significant difference was found between the K_{Ic} for the C(T) samples and the K_{JIc} for the SE(B) samples. These results are in stark contrast to the results of Joyce and Tregoning on sample-size effects in polycrystalline steel, aluminum and titanium samples, which showed no significant effect of ligament size [34]. Indeed, in that study K_{Ic} and K_{JIc} values were found to be equal and consistent over the entire range of samples sizes valid in terms of both the linear-elastic ASTM E399 and J-integral-based ASTM E1820 standards. Furthermore, it is interesting to note that the entire set of valid K_{JIc} samples failed without significant plastic deformation so that the differences in the values would appear to result from changes in the elastic energy portion of the J-integral.

Although the 41% increase between the mean K_{Ic} values of the C(T) and SE(B) samples could not be proven to be statistically significant, this should not be interpreted as definitive proof of no sample-size effect. In fact, the correlation between valid K_{Ic} and ligament size only barely missed the cutoff for statistical significance ($p = 0.054$). Rather, this is likely related to the small number of valid SE(B) tests combined with the large scatter for the SE(B) toughness tests. It is important to note that it was not the intention of the authors to have a small number of valid

Table 1

Summary of sample dimensions and pre-cracking conditions as well as fracture toughness results, failure types and required sample thickness, B_{crit}, and ligament size, b_{crit}, for plane strain and small scale yielding conditions.

B (mm)	W (mm)	a/W	b (mm)	K_{Ic}, K_{JIc}, K_Q (MPa m$^{1/2}$)	B_{crit}, b_{crit} (mm)	Failure type (ASTM E399)	Pre-cracking
C(T) K_{Ic}							
1.99	21.45	0.50	10.8	18.7	0.3	III	Tension
2.03	21.42	0.50	10.77	28	0.68	III	Tension
2.26	19.85	0.50	9.95	28.8	0.72	III	Tension
2.34	19.83	0.50	9.91	25.7	0.57	III	Tension

Mean: 25.3 MPa m$^{1/2}$; standard deviation: 4.6 MPa m$^{1/2}$

B (mm)	W (mm)	a/W	b (mm)	K_{Ic}, K_{JIc}, K_Q (MPa m$^{1/2}$)	B_{crit}, b_{crit} (mm)	Failure type (ASTM E399)	Pre-cracking
SE(B) K_{Ic}							
2.01	4.23	0.55	1.91	36.5	1.15	III	Tension
2.05	4.15	0.49	2.1	26.2	0.59	III	Tension
2.3	3.94	0.54	1.8	35.3	1.08	III	Tension
2.34	4.08	0.47	2.18	44.9	1.74	III	Tension

Mean: 35.7 MPa m$^{1/2}$; standard deviation: 7.7 MPa m$^{1/2}$

B (mm)	W (mm)	a/W	b (mm)	K_{Ic}, K_{JIc}, K_Q (MPa m$^{1/2}$)	B_{crit}, b_{crit} (mm)	Failure type (ASTM E399)	Pre-cracking
SE(B) K_{JIc}							
2.04	4	0.58[*]	1.66	31.1[***]	0.84	III	Tension
2.33	3.95	0.45	2.18	46.5	1.87	III	Tension
1.28	2.34	0.56[*]	1.03	35.2	1.07	III	Tension
2	2.07	0.45	1.14	43	1.6	III	Tension

Mean: 39 MPa m$^{1/2}$; standard deviation: 7.0 MPa m$^{1/2}$

B (mm)	W (mm)	a/W	b (mm)	K_{Ic}, K_{JIc}, K_Q (MPa m$^{1/2}$)	B_{crit}, b_{crit} (mm)	Failure type (ASTM E399)	Pre-cracking
SE(B) K_Q sub-sized							
1.99	4.22	0.51	2.07	63.5	3.49	III	Tension
2.2	4.1	0.48	2.12	66.4	3.81	III	Tension
1.33	2.25	0.63[*]	0.84	39.9	1.38	III	Tension
1.1	2.13	0.7[*]	0.65	66.6	3.84	III	Tension
1.92	1.93	0.44[**]	1.08	45.5	1.79	I	Tension
1.87	4.03	0.52	1.93	72.6	4.56	I	Tension
2.04	2.03	0.58[*]	0.86	37.2	1.2	I	Tension
2.23	2.03	0.54	0.94	33.9	0.99	I	Tension
2.07	3.97	0.48	2.06	81.6	5.76	I	Tension
1.93	2.06	0.39[**]	1.26	66.6	3.84	I	Compression
2.02	2.14	0.37[**]	1.35	66.3	3.8	I	Compression

Mean: 58.2 MPa m$^{1/2}$; standard deviation: 16.1 MPa m$^{1/2}$

B (mm)	W (mm)	a/W	b (mm)	K_{Ic}, K_{JIc}, K_Q (MPa m$^{1/2}$)	B_{crit}, b_{crit} (mm)	Failure type (ASTM E399)	Pre-cracking
SE(B) K_Q notched only							
2.16	4.61	0.45	2.54	91.6	7.26	III	–
2.15	4.08	0.45	2.23	95.9	7.96	III	–
2.72	4.14	0.49	2.09	94.1	7.66	III	–

Mean: 93.9 MPa m$^{1/2}$; standard deviation: 2.2 MPa m$^{1/2}$

[*] Samples with $a/W \approx 0.55$–0.7.
[**] Samples with $a/W < 0.45$.
[***] Not a valid K_{Ic} value due to $a/W > 0.55$.

SE(B) tests. All SE(B) samples were planned to have a, b and $B > 0.72$ mm and most achieved that goal, which should have given valid K_{Ic} values for the SE(B) samples based on the C(T) test results. The small number of valid tests was due to the large scatter in the K_Q values (Fig. 4a), which is again in stark contrast to the results of Joyce and Tregoning on sample-size effects for crystalline steel, aluminum and titanium samples [34] (e.g. Fig. 4b).

One explanation for the observed behavior of increasing apparent fracture toughness with decreasing ligament size could be the critical bending ductility effect in BMGs, as described by Conner et al. [21,22]. In their experiments

on Zr-based BMG plates, they showed an increased propensity for the formation of shear bands (decreasing shear-band spacing) with decreasing plate thickness. This leads to much smaller fracture bending strains in thicker plates compared to the thinner ones since the presence of fewer shear bands leads to more shear deformation accommodated by each shear band, and thus the critical shear offset needed to crack the shear bands is reached at lower strains. This is similar to the behavior seen here for fracture toughness tests with samples of different sizes. SE(B) samples which did not meet the requirements for either K_{Ic} or K_{JIc} failed along with significant plastic
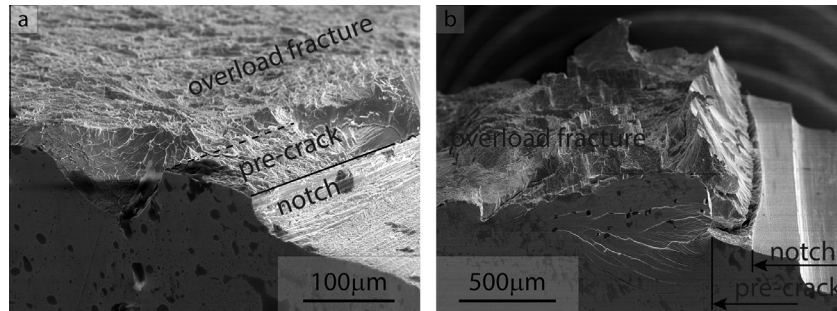
Fig. 5. Micrographs of fracture toughness samples of $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ BMG. (a) Larger samples that failed at lower fracture toughness values with nearly linear load–displacement curves showed just minor shear banding from the pre-crack and a relatively flat fracture surface. (b) Samples with large ductility and high K_Q values showed significant shear banding and blunting at the pre-crack tip and catastrophic failure sometimes occurred via crack bifurcation, leaving a large shear offset step on the fracture surface close to the pre-crack along with an extremely rough fracture surface.
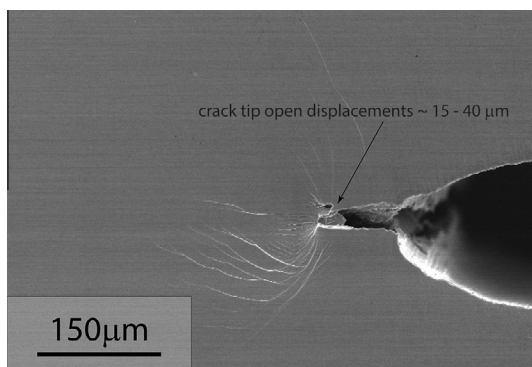
Fig. 6. Micrograph of an in situ loaded $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ SE(B) sample showing large ductility. Loading was performed in steps and frequently arrested to demonstrate the excessive formation and proliferation of shear bands starting from the pre-crack, leading to a large plastic zone and significant CTODs of ~15–40 μm. The measured K_Q values for the highly ductile samples approached those for samples that were only notched but not pre-cracked and show that such large CTODs can cause a relative insensitivity of the apparent fracture toughness to notch root radii on a similar size scale.

deformation and formed multiple shear bands ahead of the pre-crack tip (Fig. 5b), similar to the behavior observed for thin plates in bending [21,22]. This leads to large CTODs, as seen in Fig. 6, and a trend from smooth mode I to rough mixed mode fracture surfaces with increasing apparent toughness, K_Q (Fig. 5). The large CTODs resulted in high apparent fracture toughness values approaching those for the SE(B) samples tested only with notches (Fig. 4a).

The finding of increasing K_Q values with decreasing ligament size (Fig. 4a) is also contrary to the behavior of crystalline metals. Joyce and Tregoning have shown in their work on size criteria for ASTM combined fracture mechanics standards that K_Q values measured on aluminum, titanium and steel samples of different sizes remain constant until the sample ligament size reaches a lower bound, whereupon K_Q starts to decrease [34]. (Their data for aluminum are shown in Fig. 4b for comparison.) This is due to an effect of applying linear-elastic fracture mechanics analyses to undersized samples where small-scale yielding is not achieved, causing K_Q to underestimate the real toughness of the material. Taking into account the

full elastic–plastic response of the individual samples by calculating the fracture toughness in terms of the J-integral, as described in ASTM standard E1820, leads to indistinguishable results for K_{Ic} and K_{JIc}, and no sample-size effect [34]. The BMG examined here behaves entirely differently, with K_Q values showing a sharp increase with decreasing ligament size, implying that, unlike polycrystalline metals, the J-integral approach may be unable to guarantee reproducible size-independent toughness results in small-sized samples of BMGs.

In trying to determine if the ASTM E399 sample-size requirements are stringent enough for BMGs, the results are mixed. On the one hand, the difference between the valid mean K_{Ic} values for C(T) and SE(B) samples was not found to be statistically significant and pooling those data gives $K_{Ic} = 30.5 \pm 8.1$ MPa m$^{1/2}$. On the other hand, though, an almost statistically significant correlation with ligament size was observed, the K_Q values for same-sized SE(B) samples varied widely (Fig. 4a) and many samples showed extensive ductility that did not produce valid fracture toughness data by either ASTM standard. This seems to suggest that the lower sample size limits of the ASTM E399 standard appear to be near a point of transition in material behavior as the uncracked ligament width approaches the critical size to achieve gross plasticity and high ductility. Thus, a portion of the samples lie on each side of the transition point, where extensive crack blunting occurs for some samples but not for others. Furthermore, one can fully bias the tests to the high ductility side of the transition by beginning with a blunt notch, whereby K_Q is elevated to an upper bound for the pre-cracked samples and the scatter in the results is greatly reduced (Fig. 4a). It is also interesting to note that the effect of blunt notches causing an increase in the fracture toughness appears to be mostly independent of the sample size effect reported here; indeed, in recent studies the high toughness of notched samples has been shown to persist to ligament sizes as large as 12 mm [35].

It is argued here that the ASTM standards used to measure the (valid) fracture toughness of materials (E399 and E1820) must be used with care when applied to BMGs.

Results of this study suggest there might be a systematic increase in toughness at small ligament sizes even for samples meeting the most stringent plane strain K_{Ic} sample size requirements of ASTM E399; however, in this study definitive proof could not be obtained since many samples that were expected to give valid tests provided invalid results. Until standards can be more thoroughly examined, the authors suggest cautiously accepting ASTM E399 size requirements as providing size-independent toughness results while also recognizing that the small-sized samples expected to be valid will give a wider scatter in K_Q, a smaller yield of valid tests and possibly somewhat elevated toughness values. This is quite different from the conclusion of Joyce and Tregoning [34], who found the ASTM E399 sample-size requirements to be quite conservative for polycrystalline metals, with K_Q only deviating significantly for highly undersized samples (Fig 4b). Furthermore, it appears that the *J*-integral approaches used in ASTM E1820 may not be fully capable of giving sample-size-independent toughness results for samples undersized relative to ASTM E399 size requirements.

The origin of this difference between crystalline and glassy metals is most likely related to the fact that the size criteria of both ASTM E399 and E1820 were developed for polycrystalline metals while BMGs have quite different deformation behavior. In addition to the above-mentioned size-dependent ductility, BMGs also demonstrate distinctly different strain hardening/softening behavior from crystalline metals. Metallic glasses typically show local strain softening behavior in tension and compression with strain localization often on a single shear band [20,25–27], although they can sometimes demonstrate strain hardening but only as a geometrical effect in bending, as observed, for example, in Ref. [36]. Conversely, any sub-sized samples that require the usage of *J*-integral calculations of plastic contributions by E1820 must assume some local strain hardening behavior akin to crystalline metals. As McMeeking and Parks point out [37], a degree of strain hardening is required for the *J*-integral to characterize a unique Hutchinson–Rice–Rosengren (HRR) field for the stresses and displacements at the crack tip. Without the presence of a unique HRR crack-tip field, the *J*-integral approach is expected to give non-unique crack-tip stress and displacement distributions; as such, toughness measurements would become dependent on both sample size and geometry, akin to the crack-tip fields defined by slip-line field analysis with non-hardening plasticity, which are completely different for cracks loaded in tension vs. bending. Accordingly, the authors believe that the local strain softening behavior of most BMGs may be the root cause of the reported variation and size/geometry dependence in the measured fracture toughness of metallic glasses.

Finally, we should point out that a size-independent fracture toughness value may not be relevant for many target BMG applications. If BMGs are used in components with thin section thickness, designers will likely want to utilize the extra toughness and ductility achieved by the component geometry. In those cases, it will be the toughness at the component size that matters. However, when comparing published fracture toughness results, it is concluded that results based on samples sub-sized relative to the ASTM E399 standard will give sample size- and geometry-dependent results that cannot be compared fairly across studies. When only small-sized samples are permitted by processing limitations or other factors, it would be advisable to compare new BMG compositions to other compositions using samples of exactly the same size and testing configuration to allow fair comparisons. Furthermore, notched samples gave artificially high toughness values compared to all fatigue pre-cracked samples in this study and it is recommended that notched samples should be avoided. Lastly, *R*-curves are well known to be sample size and geometry dependent for all materials, and it is probable, in light of the present work, that the size and geometry dependence of BMG *R*-curves (e.g. such as those in Refs. [7,8]) may be even more significant than for crystalline metals demonstrating *R*-curves.

6. Conclusions

Our experimental studies of the fracture toughness of $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ bulk metallic glass (Vitreloy 105) call into question the precise size requirements that may be needed to measure reproducible size- and geometry-independent toughness values in bulk metallic glasses. Based on this study, the following specific conclusions can be made:

- It is concluded that the ASTM standard E399 sample size requirements should be cautiously accepted as providing size-independent K_{Ic} results while also recognizing that, compared to polycrystalline materials, the small-sized samples expected to be valid will give a wider scatter in K_Q values, a smaller yield of valid tests and possibly somewhat elevated toughness values. Such behavior is distinct from polycrystalline metals, where the size requirements of ASTM standard E399 have been shown to be quite conservative.

- Measured conditional K_Q toughness values are found to increase, with increased scatter, with progressively decreasing uncracked ligament width, *b*. The greater scatter is likely related to the size-dependent bending ductility of BMGs, as samples below a certain critical size are able to prevent catastrophic failure in bending by the formation of multiple shear bands throughout the extent of the uncracked ligament. As the critical dimension is approached, a portion of the samples lie on each side of a transition point, where extensive shear banding and crack blunting occurs for some samples but not for others. Such behavior is again distinct from crystalline metals.

- Samples smaller than required by ASTM standard E399 for valid tests are allowed by the *J*-integral-based analysis of ASTM standard E1820; however, in this study

such tests were found to give either significantly higher K_{JIc} toughness values relative to K_{Ic} or invalid results according to the standard. Such behavior is quite different from crystalline metals and is most likely related to the distinct local strain softening behavior found in metallic glasses, which gives rise to non-unique crack-tip stress and strain fields.

- Toughness values measured using samples smaller than required by ASTM E399 for valid fracture toughness values should be considered as size dependent, even when considered valid by J_{Ic} measurements. When only small-sized BMG samples are permitted by processing limitations or other factors, it would be advisable to compare new BMG compositions to other compositions using exactly the same sample geometry and testing configuration to allow fair comparisons.

Acknowledgements

References

[1] Byrne CJ, Eldrup M. Science 2008;321:502.
[2] Inoue A, Zhang T, Masumoto T. Mater Trans JIM 1989;30:965.
[3] Inoue A, Yamaguchi H, Zhang T, Masumoto T. Mater Trans JIM 1990;31:104.
[4] Schroers J. JOM J Miner Metals Mater Soc 2005;57:35.
[5] Ashby MF, Greer AL. Scr Mater 2006;54:321.
[6] Shek CH, Lin GM, Lee KL, Lai JKL. J Non-Cryst Solids 1998;224:244.
[7] Demetriou MD, Launey ME, Garrett G, Schramm JP, Hofmann DC, Johnson WL, et al. Nat Mater 2011;10:123.
[8] He Q, Shang JK, Ma E, Xu J. Acta Mater 2012;60:4940.
[9] Gilbert CJ, Ritchie RO, Johnson WL. Appl Phys Lett 1997;71:476.
[10] Gilbert CJ, Schroeder V, Ritchie RO. Metall Mater Trans A 1999;30:1739.
[11] Flores KM, Dauskardt RH. Scr Mater 1999;41:937.
[12] Schneibel J, Horton J, Munroe P. Metall Mater Trans A 2001;32:2819.
[13] Flores KM, Johnson WL, Dauskardt RH. Scr Mater 2003;49:1181.
[14] Lewandowski JJ, Wang WH, Greer AL. Philos Mag Lett 2005;85:77.
[15] Launey ME, Busch R, Kruzic JJ. Acta Mater 2008;56:500.
[16] Gu XJ, Poon SJ, Shiflet GJ, Lewandowski JJ. Acta Mater 2010;58:1708.
[17] Philo SL, Heinrich J, Gallino I, Busch R, Kruzic JJ. Scr Mater 2011;64:359.
[18] Ritchie RO, Francis B, Server WL. Metall Trans A 1976;7:831.
[19] Lowhaphandu P, Lewandowski JJ. Scr Mater 1998;38:1811.
[20] Lewandowski JJ, Shazly M, Shamimi Nouri A. Scr Mater 2006;54:337.
[21] Conner RD, Johnson WL, Paton NE, Nix WD. J Appl Phys 2003;94:904.
[22] Conner RD, Li Y, Nix WD, Johnson WL. Acta Mater 2004;52:2429.
[23] E08 Committee. E399–12e1 standard test method for linear-elastic plane-strain fracture toughness KIc of metallic materials. West Conshohocken, PA: ASTM International; 2012.
[24] E08 Committee. E1820–11e2 standard test method for measurement of fracture toughness. West Conshohocken, PA: ASTM International; 2011.
[25] Spaepen F. Acta Metall 1977;25:407.
[26] Steif PS, Spaepen F, Hutchinson JW. Acta Metall 1982;30:447.
[27] Pan J, Chen Q, Liu L, Li Y. Acta Mater 2011;59:5146.
[28] Hassan HA, Kecskes L, Lewandowski JJ. Metall Mater Trans A 2008;39:2077.
[29] Lin XH, Johnson WL, Rhim WK. Mater Trans 1997;38:473.
[30] Maxwell DC. Strain based compliance method for determining crack length for a C(T) compact(type) specimen. Air Force Wright Aeronautical Laboratories. Report no. AFWAL-TR-87-4046; 1987.
[31] Dowling NE, Wilson WK. Eng Fract Mech 1984;20:569.
[32] Fett T, Munz D. Arch Appl Mech 2006;76:667.
[33] Morrison ML, Buchanan RA, Liaw PK, Green BA, Wang GY, Liu CT, et al. Mater Sci Eng A 2007;467:190.
[34] Joyce J, Tregoning R. In: Jerina K, Paris P, editors, Fatigue and fracture mechanics, vol. 30. West Conshohocken, PA: ASTM International; 2000. ASTM STP 1360.
[35] Sunny G, Prakash V, Lewandowski JJ. Metall Mater Trans A 2013;44:4644.
[36] He Q, Xu J. J Mater Sci Technol 2012;28:1109.
[37] McMeeking R, Parks D. In: Landes J, Begley J, Clarke G, editors. Elastic plastic fracture. West Conshohocken, PA: ASTM International; 1979. ASTM STP 668.