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Flaw tolerance of metallic glasses

Wen Chen [a], Ze Liu [a, b, *], Jittisa Ketkaew [a], Rodrigo Miguel Ojeda Mota [a], Sung-Hyun Kim [a], Michael Power [c], William Samela [d], Jan Schroers [a, e, **]

[a] Department of Mechanical Engineering & Materials Science, Yale University, New Haven, CT 06511, USA
[b] Department of Engineering Mechanics, School of Civil Engineering, Wuhan University, Wuhan 430000, China
[c] School of Engineering and Applied Science, Yale University, USA
[d] Department of Geology and Geophysics, Yale University, USA
[e] Center for Research on Interface Structures and Phenomena (CRISP), Yale University, USA

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ABSTRACT

The flaw tolerance of bulk metallic glasses (BMGs) is evaluated using a thermoplastic synthesis approach. We found that flaw tolerance quantified by the notch toughness decreases apparently with decreasing radius until a critical value. Below this critical value, measured notch toughness is independent of its radius, revealing a flaw tolerance behavior of BMGs. We explain such flaw tolerance by a critical plastic zone originating from the BMGs' inherent crack tip blunting capability. This zone defines a characteristic distance over which stable shear banding plastic process develops prior to fracture instability. The specific characteristic distance and crack blunting capability vary widely among BMGs, which rationalizes the vast variety in their fracture behavior and suggest specific flaw tolerance. Our finding is encouraging for BMGs' structural applications since flaws smaller than the critical value are increasingly difficult to avoid but are "indistinguishable" in their influence to fracture toughness.

1. Introduction

Flaws are almost inevitably present in engineering structural materials. They originate from fabrication, mechanical damage, corrosion, or structural features such as microcracks, corners, holes, and welded joints. In general, structural flaws behave as stress risers and can exert a drastic effect on the materials' mechanical performance. For example, in brittle materials such as ceramics and oxide glasses, flaws in the form of pores and microcracks can dramatically degrade strength and toughness, and often cause premature brittle fracture at a stress well below their estimated yield strength [1]. As a consequence, most structural materials, particularly brittle ones are not limited by their strength but rather by their flaw resistance (or fracture toughness). Therefore, fracture toughness is often the limiting design consideration [2], and a fundamental understanding of the material's response to flaws is of great significance.

Most structural flaws can be represented as sphere-like defects with finite radii. Through introducing a notch crack into a bulk sample, notch toughness testing has been widely utilized to assess the flaw resistance in a wide range of materials [3–5]. In quantifying the notch toughness, K_Q, the notch radius, ρ, is a key parameter that defines the sharpness of the notch and hence simulates the stress concentration factor of a flaw. Variation of ρ may affect stress field and crack propagation behavior. Influence of ρ on notch toughness measurements has been widely studied in various materials classes such as crystalline metals, ceramics, and composites [6–11]. It has been found for some materials that K_Q decreases with decreasing ρ due to the elevation of stress intensification until a critical notch radius, ρ_c, is reached. Below ρ_c, K_Q has been found to be independent of ρ, which means flaws smaller than ρ_c are indistinguishable from ρ_c [7–11]. This critical radius, which is essential for the use of corresponding materials in structural applications, has been associated with the material's specific microstructural features. For example, in crystalline metals or ceramics where grain boundaries often act as an effective barrier to crack propagation, ρ_c is controlled by the grain size [7,9]. In composites, the value of ρ_c has been associated with the spacing of secondary phases or inclusions [10,11]. More generally, microstructural

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* Corresponding author. Department of Mechanical Engineering & Materials Science, Yale University, New Haven, CT, 06511, USA.
** Corresponding author. Department of Mechanical Engineering & Materials Science, Yale University, New Haven, CT, 06511, USA.
E-mail addresses: ze.liu@whu.edu.cn (Z. Liu), jan.schroers@yale.edu (J. Schroers).

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features of a material have been associated with the fracture toughness or the flaw tolerance.

Lacking microstructural features, bulk metallic glasses (BMGs) are a class of amorphous structural alloys for which crystallization can be readily avoided during solidification [12]. Due to an isotropic and homogeneous structure, BMGs generally exhibit high strength and high elasticity [12–14]. Fracture toughness, on the other hand, ranges from ideally brittle to extraordinary tough within the material class of BMGs [5,15]. Precise measurements of BMGs' toughness have been proven challenging [16]. For example, toughness values ranging from ~16 to ~130 MPa $\sqrt{m}$ have been measured for $Zr_{41.2}Ti_{12.5}Cu_{10}Ni_{10}Be_{22.5}$ BMG [17–21]. It has been speculated that observed large scatter originates from the variety of sample geometries or loading modes [22–26], as well as difficulties in the sample preparation such as difficulties in fabricating precise notches or pre-cracks, different cooling rates, casting defects, and compositional fluctuations during vitrification [16,27]. The featureless structure of BMGs lacks an obvious length scale to set ρ_c and has suggested that the BMGs' notch toughness often decreases with decreasing notch radius [17,28,29]. This behavior is concerning for their structural applications since it would suggest that BMGs become increasingly “brittle” with decreasing flaw size. Practically, small flaws are significantly more difficult to avoid than larger flaws during the alloying and vitrification process.

To systematically investigate the effect of notch sharpness on the toughness of BMGs, we introduce a method to fabricate toughness test samples, where we can minimize the previously suggested potential sources of error and precisely manipulate the notch radius within 1 μm to measure toughness of BMGs in a highly reproducible manner. Our method is based on a combination of thermoplastic forming (TPF) of BMGs in their supercooled liquid region and Si photolithography. Through this method, we varied the notch radius continuously from pre-cracked to 380 μm. The effect of the notch radius on the crack fracture behavior was examined for three well studied and representative BMGs, $Zr_{44}Ti_{11}Ni_{10}Cu_{10}Be_{25}$ [30], $Pd_{43}Cu_{27}Ni_{10}P_{20}$ [31], and $Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5}$ [32]. Surprisingly, and in contrast to expectations and most previous findings [17,28,29], we found that considered BMGs exhibit a specific critical notch radius, ρ_c. Above ρ_c, K_Q decreases apparently with decreasing ρ while below ρ_c, K_Q is independent of ρ, suggesting a flaw tolerance behavior. We argue the intrinsic length scale setting ρ_c is the BMG's specific shear banding plastic zone size which originates from the BMGs' specific capability in blunting the crack tip.

2. Experiments

2.1. Sample preparation

In this work, we consider $Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$, $Pd_{43}Cu_{27}Ni_{10}P_{20}$, and $Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5}$ BMGs. The three alloys represent the wide range of toughness behaviors of BMGs [33]. Master alloy ingots were prepared by arc-melting a mixture of the pure elements in an argon atmosphere with a low oxygen level of 350 ppm. The amorphous state was achieved by rapid quenching of alloy melts into a metal mold. The amorphous nature of the BMGs was confirmed by X-ray diffraction (XRD-6000 Shimadzu) and differential scanning calorimeter (Perkin Elmer Diamond DSC).

We employ TPF-based molding to fabricate various single edge notched tension (SENT) samples by replicating BMGs from Si molds. Within our fabrication method, we first design the geometry of SENT BMG samples with different notch radii in AutoCAD software. This design is used to construct a photomask with a Heidelberg DWL-66 laser beam writer. Silicon lithography and deep reactive ion etching were subsequently performed to fabricate Si molds with an etching depth of approximately 350 μm. Replication of the SENT BMG samples was achieved by TPF of BMGs into the Si molds within ~100 seconds at 430 °C under a pressure of 20 MPa for $Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$, at 365 °C under a pressure of 5 MPa for $Pd_{43}Cu_{27}Ni_{10}P_{20}$, or at 270 °C under a pressure of 1 MPa for $Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5}$. After molding, the extra BMG was removed and BMG SENT samples with varying notch radii of pre-cracked, 3 μm, 10 μm, 25 μm, 50 μm, 100 μm, 150 μm, 230 μm, and 380 μm were released out of Si molds by etching in 20% (mass fraction) KOH solution (Fig. 1). The thickness of the BMG SENT samples is approximately 300 μm. Overall, we fabricated ~100 test samples of $Zr_{44}Ti_{11}Ni_{10}Cu_{10}Be_{25}$, $Pd_{43}Cu_{27}Ni_{10}P_{20}$, and $Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5}$ with various notch radii (Fig. 1). For the pre-cracked samples, a 2 Hz cyclic load with a load range of $\Delta K \approx 10$ MPa $\sqrt{m}$, and $K_{min}/K_{max} \approx 0.2$ was applied to initiate and grow pre-crack ahead of the 3 μm notch using the Instron 5543 tensile machine.

2.2. Mechanical characterization and analysis

The notch toughness of all considered SENT samples was evaluated via uniaxial tensile tests which were quasi-static displacement-controlled (strain rate: 10^{-4} s^{-1}) on an Instron 5543 tensile testing machine. The precision in fabricating, in particular the notch, is better than 1 μm (Fig. 1). This high precision combined with the unique TPF fabrication mitigates most of the extrinsic influences such as casting defects, cooling rate, residual stress, thermal history, and sample fabrication precision. As a consequence, the experimental scatter in K_Q measurements using our preparation method is drastically smaller than previously reported [16]. To evaluate reproducibility, 20 $Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$ SENT samples with $\rho = 150$ μm were fabricated and tested. For the 20 samples, we measured an average notch toughness of $\overline{K_Q} = 109$ MPa $\sqrt{m}$ with a standard deviation of only ~3 MPa $\sqrt{m}$ [34]. Seven samples were prepared and tested for each other notch radius for the considered BMGs to quantify scatter. During deformation, in-situ images were recorded by a home-built Qimaging microscope to capture the plastic zone development in front of the notch (Fig. 2). After fracture, the fractured surface morphology was examined by scanning electron microscopy (SEM). The stress intensity factor K for the SENT geometry is calculated by $K = \sigma\sqrt{\pi a}\,F\left(\frac{a}{W}\right)$, where $a = 4$ mm, the notch length, σ is the applied far-filed stress, $W = 8$ mm, the width of the SENT sample, $F\left(\frac{a}{W}\right) = \sqrt{\frac{2W}{\pi a}\tan\frac{\pi a}{2W}}\cdot\frac{0.752+2.02\left(\frac{a}{W}\right)+0.37\left(1-\sin\frac{\pi a}{2W}\right)^3}{\cos\frac{\pi a}{2W}}$ is a configuration correction factor [35]. To investigate the validity of the assumption of linear elastic K-field dominance, the critical energy release rate G_c was also measured and a back-calculated K_Q was derived in order to examine the validity of K-field approximation in notch toughness characterization of considered BMGs (see Appendix).

3. Results

3.1. Notch radius dependence of K_Q

For $Zr_{44}Ti_{11}Ni_{10}Cu_{10}Be_{25}$, we found that K_Q increases linearly with $\rho^{1/2}$ for $\rho > 100$ μm. Specifically, K_Q increases from 97 MPa $\sqrt{m}$ for $\rho \le 100$ μm up to 143 MPa $\sqrt{m}$at $\rho = 380$ μm (Fig. 3a). Below ρ_c, $K_Q \approx 97$ MPa $\sqrt{m}$ is independent of ρ, denoted as K_c (Fig. 3a). Hence, this suggests a critical notch radius of $\rho_c \approx 100$ μm. For $Pd_{43}Cu_{27}Ni_{10}P_{20}$, K_Q exhibits a similar qualitative trend but a different $\rho_c \approx 30$ μm. Below ρ_c, K_Q remains approximately constant ($K_Q \approx 46$ MPa $\sqrt{m}$) and above ρ_c, K_Q increases linearly with $\rho^{1/2}$ (Fig. 3b). A similar phenomenon of a critical notch radius has also