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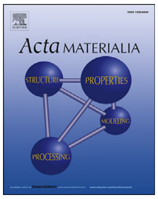

Full length article

On the understanding of the effects of sample size on the variability in fracture toughness of bulk metallic glasses

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ABSTRACT

High strength in combination with improvements in failure characteristics and associated gains in fracture toughness have placed bulk metallic glasses (BMGs) among the most damage-tolerant materials to date. Recent studies show, however, that there can be large variabilities in the mechanical performance of these alloys, particularly in their toughness, which are likely associated with sample-size effects or structural variations from differences in processing. Here, we examine the variation in fracture toughness of the Pd-based metallic glass $Pd_{77.5}Cu_6Si_{16.5}$, using single-edge notched bend specimens but in two different sizes. Although all toughness results on this glass were "valid" in terms of the nonlinear-elastic fracture mechanics *J*-standard, *i.e.*, one would expect a single value of the fracture toughness for this alloy, marked differences were apparent in the toughness values and failure characteristics of the differently-sized samples. Specifically, significantly larger variations in toughness values were measured in larger-sized samples, which all essentially failed catastrophically, whereas none of the smaller-sized samples failed catastrophically yet displayed far less scatter in their measured toughness. Additional *in situ* tests on the smaller-sized samples in a scanning electron microscope revealed stable crack growth and progressive resistance to crack extension, *i.e.*, rising crack-resistance (*R*-curve) behavior. Overall, this marked transition from brittle catastrophic failure in large samples, where a size-independent fracture toughness can be measured, to non-catastrophic, more ductile (*R*-curve), behavior in smaller samples, the latter associated with higher toughness, is related to the distinct size-dependent bending ductility and strain-softening behavior in BMGs.

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1. Introduction

Since their introduction in the early 1960s [1], metallic glasses have gained significant attention owing to their exceptional combination of properties such as near-theoretical strength, low stiffness and large elastic strain limits [2–7]. Due to the development of bulk glass-forming alloys [8–10] together with the constantly increasing processing dimensions, bulk metallic glasses (BMGs) are now considered as structural materials [11]. Despite their generally acceptable combination of strength and toughness (*i.e.*, damage tolerance) – most glasses exhibit strength levels well above 1.5 GPa with fracture toughness values that are mainly reported to lie between 10 and 100 MPa.m$^{1/2}$ [12–20] – the often large variability in mechanical performance, particularly in their fracture and toughness behavior, has been one factor that has compromised their potential use for many structural applications to date. Although some of this variability can be traced to the poor quality of some early BMGs, *e.g.*, there is evidence that certain glasses failed in a highly brittle manner at fracture toughness K_{Ic} values as low as ~2 MPa.m$^{1/2}$ [21], there are reports of recently developed monolithic and composite BMGs, specifically Pd-based and Zr-based glasses, with fracture toughnesses as high as ~200 MPa.m$^{1/2}$, that have been achieved by promoting ductility through the formation of multiple shear bands, leading to subcritical crack growth and increasing fracture resistance with crack extension, *i.e.*, rising crack-resistance curve (*R*-curve) behavior [22–25].

To discern the origins of this variability in properties, significant experimental and theoretical efforts are currently underway to

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improve our understanding of the links between processing, structure and properties in BMGs [5,6,11,26–40]. Aside from these structure-property relationships, which invariably dictate the characteristics and hence performance of a material, there also appears to be a strong influence of testing conditions on the mechanical behavior of (certain) metallic glasses. It is known in the glass community, for example, that in strength tests, BMGs typically show local strain-softening behavior in tension and compression with strain localization often occurring on a single shear band [41–46], whereas in bending, they can conversely demonstrate strain hardening as a result of the formation and multiplication of shear bands (which can be considered as a geometrical effect of this loading condition) [47]. Another influence of testing conditions in BMGs is apparent in their fatigue properties which can display a particularly high susceptibility to the testing environment affecting their fatigue strengths and crack-growth behavior [48–56]. Although environmental effects on a material's fracture toughness are less pronounced, it remains unclear how loading conditions can influence metallic glass samples that contain a crack. While Lewandowski and co-workers have shown that, akin to crystalline materials [57], some of the fracture toughness variability found in the literature can be explained by factors such as the use of notched *vs*. pre-cracked samples [43,58], we were recently able to attribute some of the variations in the properties of BMGs to a fracture mechanics-based sample-size effect [59]. Specifically, we found that fracture toughness tests on differently sized compact-tension and single-edge notched bend samples of the Zr-based glass $Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$ (Vitreloy 105) gave a definitive trend of progressively increasing toughness values with decreasing sample size coupled with a wider scatter in the results. We attributed this to the distinct size-dependent bending ductility known from bend tests on plates of various thickness [60,61], and to the strain-softening behavior found for metallic glasses [41–44] which acts to severely limit the extent of the unique Hutchinson-Rice-Rosengren (HRR) crack-tip field that relies on power-law hardening [62,63]. In light of this, it appears that although the use of the linear-elastic-based *K*-approach, *i.e*., ASTM standard E399 [64], may be perfectly appropriate for evaluating the toughness of brittle glasses, the corresponding use of the nonlinear-elastic *J*-integral-based ASTM standard, E1820 [65], may be questionable for high-toughness glasses, because of the highly restricted range of crack-tip *J*-field validity compared to that for strain-hardening crystalline materials.

To provide further insight into the complex effect of sample size on the variability of fracture behavior of metallic glasses, we have examined here one specific Pd-based glass in a single test geometry, that of the highly constrained single-edge notched bend geometry, but in two different sizes. Compared to both geometries of the Zr-glass samples in Ref. [59], where dimensions were either comparable or well above the glass' critical bending thickness, here we specifically focus on samples that in terms of size range from the Pd-based glass' critical bending thickness to that significantly below this dimension, and compare both the material's fracture toughness and fracture behavior using ASTM recommended sample geometries and size configurations.

2. Background

Our analysis of the variability in toughness values in metallic glasses relies on the concept of bending ductility in these materials. After numerous indications of thin wires and foils of amorphous metals showing good ductility in bending [66–73], Katuya et al. [74] and Inoue et al. [75] were first to report that such significant bending ductility in metallic glasses could only be achieved if the thickness of the bent sample was below a critical value; this implied that the bending ductility of glasses is size-dependent. Based on these observation, Conner et al. associated the high ductility in plates of a Zr-based metallic glass subjected to bending to the formation of a large number of shear bands [21,22]. Specifically, when they tested plates with a thickness, *t*, less than 1.5 mm that were bent around dies of different radii, *r*, these plates showed an increased propensity for the formation of shear bands, *i.e*., decreasing shear-band spacing, λ, and increasing ductility prior to fracture with decreasing plate thickness; the relevant dimensions are shown in Fig. 1. They argued that the much smaller fracture bending strains in thicker plates compared to those measured in thinner ones result from pronounced local strain relaxation in the vicinity of each shear band, thus preventing other shear bands to form close to the existing ones; this results in a larger shear-band spacing in thicker plates. Since the presence of fewer shear bands leads to more shear deformation accommodated by each individual shear band, the critical shear offset needed to open a shear band and form a crack is reached at lower strains. This ultimately results in a more brittle behavior of thicker metallic glass samples and leads to the conclusion that BMG plates below a certain critical thickness can achieve the relevant number of shear bands to demonstrate significant bending ductility. Whereas the critical bending thickness is a well-known parameter in the metallic glass community and can be readily measured by bending plates of diminishing thickness, to date there is only limited analysis available to predict what this thickness should be.

3. Experimental procedures and data analysis

3.1. Fabrication and characterization of the Pd-based bulk metallic glass

The Pd-based master alloys were prepared by arc melting high-purity raw materials (Pd 99.95%, Si 99.9997%, Cu 99.995%) according to their atomic ratios in a 6N argon atmosphere using an arc melter (Edmund Bühler Labortechnik, Germany). These master alloys were used for the preparation of $Pd_{77.5}Cu_6Si_{16.5}$ bulk metallic glasses by flux treatment. The ingots were fluxed with dehydrated boron oxide, B_2O_3 (99.98%), in quartz tubes and subsequently water-quenched from 1150 °C to obtain amorphous rods of 8 mm diameter. The cyclic heating-cooling treatment associated with fluxing was performed for eight fluxing cycles between 300 °C and 1150 °C with an overall fluxing time of 24 h. A detailed description of the fluxing procedure is provided in our preceding publications [76,77].

The amorphous structure of the glass was verified by x-ray diffraction, XRD, using a Stoe STADI x-ray diffractometer in Bragg–Brentano geometry. Similarly, all thermal characteristics of

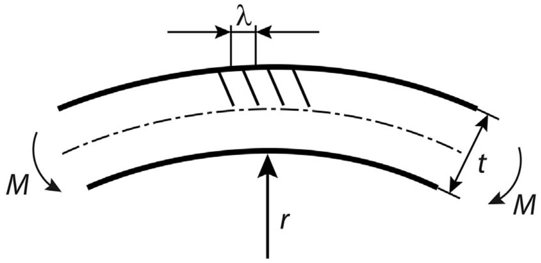

Fig. 1. Bending ductility of bulk metallic glasses. Conner et al. [21,22] have shown that below a certain critical thickness, *t*, the spacing of shear bands, *λ*, in bulk metallic glass (BMG) samples decreases with increasing bending moment, *M*, and decreasing radius, *r*. This enables BMGs to prevent catastrophic failure through the formation of multiple shear bands resulting in increasing ductility in bending with decreasing thickness. (Figure taken from Ref. [59].)