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## Effect of sample size on ductility of metallic glass

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### Effect of sample size on ductility of metallic glass

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With decreasing sample dimension, the compressive plastic strain of a Zr-based metallic glass increases from near zero to as high as 80% without failure. This indicates that macroscopically brittle or ductile deformation behaviour can occur in chemically and structurally identical metallic glass. A concept of critical shear offset is proposed to explain the strong size effect on the enhanced plasticity of metallic glass by taking the shear fracture energy density into account. This finding provides new understanding on the principle that for metallic glass 'smaller is more ductile', even on the macroscopic scale.

Keywords: metallic glass; sample size; ductility; deformation

#### 1. Introduction

For a number of face-centred-cubic single crystals, their strength is found to be highly dependent on the sample size in the micron or submicron range [1-3]. For example, the strength of Ni or Au micrometre-sized single crystals is several times higher than that of large-size bulk samples, and the strength can be even further increased by more than one order of magnitude in the case of submicrometre-size pillars [1-3]. This dramatic size effect often occurs when the dimension of the sample is smaller than the characteristic length for multiplication of dislocations [1-3].

In contrast to crystalline metallic materials, for metallic glasses without slip systems or a dislocation-free structure, shear banding becomes the important plastic deformation mechanism at room temperature [4]. Once yielding starts, shear bands propagate rapidly, leading to catastrophic failure [4]. Since the yield strength of metallic glasses is considered to approach the theoretical strength limit [5], there should be no significant size effect on the strength values. Some recent reports indicate that the strength of micrometre-size metallic glass samples is only slightly higher than that of the corresponding bulk samples [4,5]. However, the size effect on the plastic deformation capability of metallic glass is still not well understood. Recently, it was found that when the sample size is decreased to

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a dimension close to the characteristic length-scale for the nucleation and propagation of shear bands, homogeneous plastic deformation appears in metallic glasses [6,7]. In this letter, we show that macroscopically brittle and ductile deformation behaviour can occur in chemically and structurally identical metallic glass, depending on the sample size. Based on these findings, we propose a new concept of critical shear offset to explain the transition from macroscopically brittle to macroscopically ductile behaviour in metallic glass.

#### 2. Experimental procedure

An ingot with a composition of Zr<sub>52.5</sub>Ni<sub>14.6</sub>Al<sub>10</sub>Cu<sub>17.9</sub>Ti<sub>5</sub> was prepared by arc melting a mixture of pure elements in a Ti-gettered argon atmosphere on a water-cooled copper plate. The ingot was re-melted several times for homogenisation. The final ingot had the shape of rectangular bar with a dimension of  $60 \times 30 \times 3 \text{ mm}^3$  (length, height and width). The microstructure and the phase of the prepared ingot were characterised by using a Leo Supra 35 scanning electron microscope (SEM), respectively, as well as by X-ray diffraction using a Rigaku diffractometer with Cu-K $\alpha$  radiation. The final ingot showed only broad diffraction maxima and no peaks corresponding to crystalline phases were detected, revealing the amorphous structure of the sample. The samples for the compression tests with an aspect ratio of 2:1 and dimensions of  $3.0 \times 3.0 \times 6.0 \text{ mm}^3$  (sample S1),  $1.5 \times 1.5 \times 3.0$  mm<sup>3</sup> (sample S2), and  $1.0 \times 1.0 \times 2.0$  mm<sup>3</sup> (sample S3) were machined from the same cast bar. All the lateral surfaces of each sample were polished by 1.5 µm diamond paste. Uniaxial compression tests were performed with a MTS810 testing machine at room temperature using a constant strain rate of  $1 \times 10^{-4} \,\mathrm{s}^{-1}$  (same initial strain rate for all samples as evidenced by the fixed displacement rate). Each experiment was repeated more than three times. The deformed samples were investigated by SEM to reveal the deformation features.

#### 3. Results and discussion

Figure 1 shows the engineering stress–strain curves of samples S1, S2 and S3. It can be seen that the plasticity recorded for the different specimens changes significantly, although the yield strength is almost the same. Converting the engineering stress–strain curves into true stress–strain curves yields compressive plasticities of 1.1 and 6.4% for samples S1 and S2, respectively. Sample S1 failed by shear fracture, and there are only two major shear bands and several weak secondary shear bands visible on the surface, as shown in Figures 2a and b. However, there are a few major shear bands on the surface of sample S2, as indicated in Figures 2c and d. Both samples S1 and S2 failed with a fracture angle smaller than 45°, which is consistent with previous reports [8]. It is interesting to note that the sample S3 could finally be compressed to a plastic strain as high as 80% without obvious fracture, exhibiting super plastic deformation capability [9,10], as shown in Figures 3a and b. Dense, multiple, and intersecting shear bands were formed in sample S3, as indicated by the arrows in Figures 3c and d. To confirm this new phenomenon, the experiments were repeated for more than three times, it was found that all the repeated experiments have the similar tendency as mentioned above.

As reported recently, there is a strong size effect on the mechanical properties of metallic glasses for micrometre-size specimens [4,5]. However, from the present results,



Figure 1. (Color online). Engineering stress–strain curves of samples S1, S2 and S3 with the same aspect ratio of 2:1 under unaxial compression, showing that the plasticity increases with decreasing sample size.



Figure 2. SEM images showing the deformation features of (a) and (b) sample S1, and (c) and (d) sample S2. Sample S1 failed by the formation of one or two major shear bands (including the fracture plane) and a few tiny secondary shear bands. However, a number of major shear bands were produced in sample S2.



Figure 3. SEM images showing the super plastic deformation features of sample S3. (a) and (b) the sample was compressed into a disk; (c) and (d) multiple interacting shear bands were formed in the sample.  $\sigma_c$  denotes the loading direction.

we find that a strong size effect on the shear deformation behaviour of metallic glass already occurs on a macroscopic scale, which is different from the results reported by Shen's group [11,12]. In their work, the plasticity was influenced by both the size of the casting and the specimen size. However, the current samples with different sizes were cut from an identical metallic glass bar and, therefore, our present results were purely related to the effect of the sample size [13]. To understand such size effect, it is necessary to know the whole process of the transformation from a shear band to a crack and the final fracture. As is well known, the room-temperature plastic strain of metallic glass is produced by the shear offset of two undeveloped parts separated by the localised shear band [14]. With the shear band propagating, the shear offset increases, so does the plastic strain. When a shear band propagates to a critical length, it develops maturely with a low bonding strength [15], leading to the final catastrophic fracture along shear band. Thus, there should be a 'critical shear offset  $\lambda_c$ , above which, the shear band starts to be unstable, leading to the final shear fracture [16]. The critical shear offset is a direct parameter phenomenally reflecting the stable shear capability. The length of critical shear offset is equal to that of the smooth region at the initial fracture surface of metallic glass sample after deformation [16,17]. Therefore, it is suggested that the shear deformation capability of metallic glass is related to the critical shear offset: the plastic strain of metallic glass at fracture increases linearly with increasing critical shear offset. Furthermore, based on the concept of critical shear offset, the overall deformation behaviour of metallic glass can be categorised into two regimes with regard to the sample size. At first, if the sample size w is significantly larger than the critical shear offset  $\lambda_c$ , the shear offset produced by propagation of shear band will reach the critical one  $\lambda_c$ , and catastrophic failure will occur. Therefore, the plastic strain at failure  $\varepsilon_p$  can be calculated by the sample size w (the aspect ratio is 2) and the critical shear offset  $\lambda_c$ , which is expressed as

$$\varepsilon_{\rm p} = \frac{\lambda_{\rm c} \cos\theta}{2w},\tag{1}$$

where  $\theta$  is the shear angle between the shear plane and the loading direction (42° for the present metallic glass), and w is the sample size (diameter or width of the sample). For a given metallic glass, the critical shear offset  $\lambda_c$  is constant. Therefore, according to Equation (1), a decreasing sample size w will obviously increase the plasticity of metallic glass sample, as schematically shown in Figure 4 (region I).



Specimen size (w)

Figure 4. Illustration of the sample size effect on the shear deformation ability of metallic glasses with stable and unstable regions. When the sample size is larger than  $w_c$ , the sample exhibits limited plastic deformation; however, when the sample size is smaller than  $w_c$ , the sample exhibits super plastic deformation. The inset is a SEM image showing the critical shear offset  $\lambda_c$  measured on the fracture surface of sample S1.

When the sample size w is equal to or smaller than the critical sample size  $w_c$ , the critical plastic strain  $\varepsilon_c$  (it is the minimum plastic strain resulting in the two parts separated by shear band that touch the upper and lower platens) is estimated to be about 22.2% from Equation (1). In this case, the two parts of the sample separated by the major shear band will touch the upper and lower platens of the compression clip. Thus, the shearing process should be stable due to the constraint by the two platens, so that no catastrophic failure occurs for a sample with a size smaller than  $w_c$ , as shown schematically in Figure 4 (region II). This is consistent with the observations from Figures 1 and 3. According to Equation (1), the critical size  $w_c$  can be calculated as

$$w_{\rm c} = \frac{\lambda_{\rm c} \cos \theta}{2\varepsilon_{\rm c}}.$$
(2)

Based on the SEM observations on the fracture surfaces of samples S1 and S2, the critical shear offset  $\lambda_c$  for the present Zr-based metallic glass is about 50 µm (see the inset SEM image in Figure 4), and the shear angle  $\theta$  is about 42°. Inserting these values together with the critical plastic strain  $\varepsilon_c = 22.2\%$  into Equation (2) gives a critical sample size  $w_c = 83.7 \mu m$ , which is only one-tenth of the experimentally observed sample size exhibiting large plastic strain. However, the calculated critical sample size is based on the ideal single shear band model. When taking two or more shear bands into account (Figures 2b and d), the critical sample size is possibly several times larger than for the case of a single shear band, e.g. the critical sample size of the present Zr-based metallic glass is near 1.0 mm according to the results for samples S1, S2 and S3.

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On the other hand, owing to the highly localised shear deformation in metallic glass, the elastic energy stored before fracture is mostly dissipated on the fracture surface as heat [18]. Some results showed that heat plays an important role in the softening of a shear band and the catastrophic fracture of metallic glass [19]. The energy density of the shear fracture surface caused by the elastic energy release during the fracture process can be approximately expressed as

$$\delta = \frac{1}{2}\sigma_{\rm e}\varepsilon_{\rm e}V/A = w\sigma_{\rm e}\varepsilon_{\rm e}\sin\theta,\tag{3}$$

where  $\sigma_e$  is the maximum elastic stress (elastic limit),  $\varepsilon_e$  is the maximum elastic strain, V is the volume of the sample, A is the area of the shear plane, w is the sample size (diameter or width) and  $\theta$  is the shear angle between the shear plane and the loading direction. According to Equation (3), it is clear that the energy density dissipated on the shear fracture surface decreases linearly with decreasing sample size (w). Thus, a decreasing sample size will enhance the stability of the shear band, i.e. it becomes difficult to render the shear band to propagate maturely to form a crack. Therefore, with decreasing sample size, multiplication of shear bands becomes more likely, thus enhancing the plasticity of the metallic glass.

Critically assessing the recent reports on ductile metallic glasses with excellent plasticity [4,9,10,20,21] reveals that most of them were tested with a sample size between 1 and 2 mm. The most important problem is that a comparison of the plasticity of different metallic glasses does not fit to a unified condition; especially the data that were not recorded for the same sample size. However, the present findings indicate that the sample size effect should be taken into account. Thus, the results reported in literature are often not strictly comparable.

#### 4. Conclusions

In summary, the compressive plasticity of chemically and structurally identical metallic glassy specimens can display a large difference when their dimensions are decreased from  $3.0 \times 3.0 \times 6.0$  to  $1.0 \times 1.0 \times 2.0$  mm<sup>3</sup>. The size effect on the mechanical properties of metallic glass can be explained from two aspects: the critical shear offset and the energy density of the shear fracture surface caused by the elastic energy release. With decreasing sample size, the energy density dissipated on the shear fracture surface decreases, thus stabilising the shear bands. Moreover, with decreasing sample size down to near the critical shear offset, the shear process will be stable causing intersection and multiplication of shear bands, which improves the compressive plasticity of metallic glass. The prospect of bulk metallic glasses being used as engineering materials is still not fully clear. However, the present results imply that metallic glass exhibits a new feature, i.e. 'smaller is safer'. Therefore, metallic glasses can be potential candidates as materials in microelectromechanical systems (MEMS).

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