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## Transition of Failure Mode and Enhanced Plastic Deformation of Metallic Glass by Multiaxial Confinement\*\*

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Metallic glasses possess unique properties, such as high strength and high hardness; thus, much attention has been paid to the development of new alloy systems with good mechanical properties.<sup>[1–4]</sup> Meanwhile, the mechanical properties of the numerous metallic glasses available have been investigated from different aspects. With the development of the focus-ion-beam (FIB) technique, the size effect on the mechanical properties of metallic glasses on the micrometer scale has also been determined.<sup>[5–10]</sup> These studies found that there is little size effect on the strength, but great effect on the plastic-deformation ability of metallic glasses.<sup>[5–8]</sup> Moreover, multiple shear bands were found to be operating in the samples and the plastic strain was in the form of a shear-band burst.<sup>[5-8]</sup> In addition, the effect of the geometry on the mechanical response in metallic glasses has also been studied. For example, Lewandowski et al.<sup>[11]</sup> and Lu et al.<sup>[12]</sup> investigated the effect of pressure on the yielding and fracture of metallic glasses under tension or compression. In contrast to the catastrophic shear failure in uniaxial compression, the metallic glasses exhibited a large plastic deformation of more than 10% under confinement.<sup>[12]</sup> In addition, rolling has been applied, to induce profuse shear bands and large deformation and study the evolution of the free volume,<sup>[13–15]</sup> and also to reveal the effect of pre-existing shear bands on the mechanical properties of metallic glasses.<sup>[16]</sup> Meanwhile, it has been found that the residual stress on the surface of metallic glasses is also useful for improving their mechanical properties.<sup>[17]</sup>

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Recently, the mechanical properties of metallic glasses under biaxial tensile loading were studied by the small punch test (SPT).<sup>[18-20]</sup> It was interesting to find that the metallic glasses could be controlled to create regularly arrayed, fine, multiple shear bands under the SPT, indicating that metallic glasses essentially have a good plastic-deformation capability and thus high ductility under suitable loading conditions. The findings showed that the initiation and propagation of shear bands in metallic glasses strongly depend on the stress state. However, under the SPT, some metallic glasses show a faint shear-deformation capability, such as Ti-based metallic glass, which exhibits a significantly unstable shear deformation and a different failure mode to that of Zr-based metallic glass.<sup>[20]</sup> Therefore, in this communication, the shear-deformation behavior of a Ti-based metallic glass under multiaxial confinement is evaluated and analyzed using the SPT. We confirm that under multiaxial confinement, the sheardeformation capability of a metallic glass can be greatly improved, and that the failure modes can also be changed under this confinement.

Figure 1 shows the load–deflection curves of samples S1 and S2, subjected to the SPT. The maximum deflection of sample S1 was 0.63 mm, and the initial stiffness was 2.98 kN mm<sup>-1</sup>. However, with confinement by a Cu/11.6 at % Al alloy, the maximum deflection for sample S2 increased to 1.12 mm, which was nearly twice of that of sample S1. It is obvious that the load capability of sample S2 was increased with the confinement. According to the load–deflection curves, the mean plastic strain,  $\varepsilon_p$  (global plasticity), of samples S1 and S2 can be approximately calculated using Equation (1):<sup>[20]</sup>

$$\varepsilon_p = \sqrt{\left(\left(\frac{f-f_0}{r}\right)^2 + 1\right)} - 1,\tag{1}$$

In Equation (1),  $f_0$  is the initial elastic deflection of the load–deflection curve, f is the total deflection of the sample before fracture, and r is the effective radius of the sample, that is to say, the radius of the lower die of the small punch test. Substituting the deflection data of samples S1 and S2 into Equation (1), the equivalent plastic strains for samples S1 and S2 are obtained as 1.8% and 7.9%, respectively. This indicates that the mean plastic strain of sample S2 was increased to be three-times larger than that of sample S1 due to the additional confinement. However, it is noted that the compressive plasticity of the Ti-based metallic glasses is near zero.<sup>[20]</sup> These

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Fig. 1. Load–deflection curves for samples S1 and S2 subjected to the small punch test. Sample S1 was free of confinement on the lower surface, whereas sample S2 was confined on the lower surface by a disc of CuAl alloy.

results indicate that the brittle Ti-based metallic glass can also display a macroscopically high plasticity under the current SPT method with additional confinement, which must be



Fig. 2. a) Low-resolution SEM image of sample S1 after deformation. b) Multiple shear bands formed in sample S1. c) Catastrophic fracture occurred in sample S1. d–f) SEM images showing the deformation and fracture pattern of sample S2. Areas A–D in Fig. 2a and 2d correspond to Fig 2b, 2c, 2f and 2e, respectively.  $A \rightarrow (b)$ ,  $B \rightarrow (c)$ ,  $C \rightarrow (f)$ ,  $D \rightarrow (e)$ 

highly related to the microscopically dense shear bands, and is discussed below.

The deformation features of samples S1 and S2 are shown in Figure 2. Samples S1 and S2 both underwent a mixeddeformation process, forming radial and circumferential shear bands, respectively.<sup>[18-20]</sup> As summarized in Table 1, for sample S1, the radial-shear region was 2.69 mm in diameter, and the circumferential-shear region was 1.13 mm in diameter. The densities of the shear bands were  $35.7 \text{ rad}^{-1}$  in the circumferential direction and  $2.1 \times 10^5 \, \text{m}^{-1}$  in the radial direction, respectively. However, under confinement, the radial- and circumferential-shear regions of sample S2 were increased to 3.25 mm and 1.93 mm, respectively, and the densities of the shear bands were increased to  $62.1 \text{ rad}^{-1}$  in the circumferential direction and  $3.8 \times 10^5 \,\text{m}^{-1}$  in the radial direction. Though the radial-shear offset changed slightly, the circumferential-shear offset increased from 7.4 µm to 17.4 µm, implying that the improved shear-deformation capability of sample S2 was mostly attributed to the circumferential-shear offsets.<sup>[18-20]</sup> Therefore, with additional confinement, more multiple shear bands with larger shear offsets can be formed, which contributes to the improved shear-deformation capability of the brittle Ti-based metallic glass, leading to the significant global plasticity, as shown in Figure 1 and Table 1.

Furthermore, the failure mode of sample S2 was completely different to that of sample S1, as shown in Figure 2c and 2f. With the free lower surface, the failure of sample S1 occurred along the radial direction, and clear vein patterns along with a few molten droplets were observed on the fracture surface,<sup>[21,22]</sup> as shown in Figure 2c. However, under confinement, the fracture mode of sample S2 changed to the circumferential direction, and one of the typical features is that no molten droplets appear on the fracture surface, as displayed in Figure 2f, indicating that its final failure is not catastrophic.

High-resolution scanning electron microscopy (SEM) images show more details of the deformation features on sample S2 under confinement, as indicated in Figure 3. A few fine shear bands were observed on the shear offset caused by the radial-shear deformation,

Table 1. Mechanical properties and analysis data of shear bands for samples S1 and S2 under biaxial-tension load by the small punch test.

Samples	Maximum deflection [mm]	Range of shear bands [mm]		Density of shear bands		Shear offset [µm]	
		Radial direction	Circumferential direction	Radial direction [m <sup>-1</sup> ]	Circumferential direction [rad <sup>-1</sup> ]	Radial direction	Circumferential direction
S1 S2	0.63 1.16	3.03 3.25	1.20 1.93	$\begin{array}{c} 2.1\times10^5\\ 3.8\times10^5\end{array}$	35.7 62.1	11.6 8.7	7.4 17.4





Fig. 3. a) Secondary shear bands formed in a preformed shear band of small-punch sample S2. b) Multiple interacted shear bands on the outskirts of small-punch sample S2.

and, in the intersection region, a lot of irregular shear bands were formed (Fig. 3a). On the outskirts of sample S2, arc-like shear bands were intersected with each other, and a lot of finer and fainter shear bands were formed in the region separated by the arc-like shear bands (Fig. 3b). All of the fine shear bands also contributed to the large shear-deformation capability of sample S2 under confinement. However, this phenomenon was not observed in sample S1 without confinement, which implies that the fine shear bands formed in sample S2 are attributed to the confinement.

The plasticity of a metallic glass is attributed to the formation of shear bands; thus, its plasticity can be expressed by the relation given in Equation (2):

$$\varepsilon_{\rm p} = \sum_{i=1}^{N} \left( \frac{\lambda_i \cos \theta_i}{L} \right) \tag{2}$$

In Equation (2),  $\lambda_i$  is the shear offset of each shear band,  $\theta_i$  is the shear angle between the shear plane and the load direction, *L* is the gauge length of the sample in the load

direction, and N is the total number of shear bands.<sup>[23]</sup> Therefore, there are two premises for enhancing the plasticity of metallic glasses: one is to improve the shear-deformation capability of a single shear band, and the other is to increase the density of the shear bands.<sup>[23]</sup> The shear-deformation capability of a single shear band is characterized by the critical shear offset,  $\lambda_c$ .<sup>[23]</sup> When the active shear offset is smaller than the critical shear offset, the free volume in the shear band multiplies and the shear banding can proceed continuously and stably.<sup>[23,24]</sup> However, when the active shear offset is equal to or large than the critical value, the free volume will coalesce to form voids and then further to form shear cracks.<sup>[23,24]</sup> Once the shear cracks reach the critical shearcrack length, the stable shear banding will be transformed into a catastrophic propagation of the shear cracks.<sup>[23]</sup> For a given metallic glass, the shear deformation capability of a single shear band is governed by its chemical composition and atomic-scale structure. In order to improve the plastic deformation of the metallic glass, one should control the size of the shear offset to be smaller than the critical one, or improve the density of the shear bands to form more shear offsets with suitable values, smaller than the critical value, that is to say, to produce multiple shear bands with more homogeneous shear offsets. As far as the present sample is concerned, sample S2 was more-severely constrained with the increasing deformation amount, with the result that the radial shear band was smaller than the critical shear offset. However, the density of the radial shear bands was improved compared to that in the sample S1 (see Table 1). In this case, the radial-shear offset was well decreased and formed homogeneously, resulting in a postponement of the radial fracture of sample S2. Furthermore, the circumferential-shear bands will have the chance to further improve the plastic deformation of sample S2. Therefore, confinement is beneficial for activating more shear bands with smaller shear offsets forming in the meantime.

These results provide significant evidence that the surface stress state is very important for controlling the deformation behavior of metallic glasses.<sup>[12,17,25]</sup> The confinement stress becomes larger and larger with the going-head deformation in place of the shear offset. Therefore, the confining stress can promote the generation and interaction of multiple shear bands throughout the whole metallic-glass sample, leading to a significantly improved global plasticity. Furthermore, this means that metallic glass can be used as a safe structural material by changing its surface stress state, either by external confinement or through its residual stress on the surface:<sup>[12,25]</sup> in other words, making the surface closed rather than open can improve the shear-deformation properties of metallic glasses.<sup>[26]</sup>

To conclude, the mechanical performance, especially the plastic deformation capability, is significantly enhanced under multiaxial confinement. Through the multiaxial confinement, the density of the shear bands was greatly improved, even in the brittle Ti-based metallic glass. The fracture mode was also changed from catastrophic fracture to stable shear fracture under this confinement. The results present a simple step toward toughening brittle metallic glasses and potentially broadening their applications.

## Experimental

The Ti-based metallic glass, with nominal chemical composition of Ti<sub>40</sub>Zr<sub>25</sub>Ni<sub>3</sub>Cu<sub>12</sub>Be<sub>20</sub>, was prepared by arcmelting and was cast into a copper mold.<sup>[27,28]</sup> The final ingots had a rectangular shape with dimensions of  $60 \times 30 \times 3$  mm. The microstructure was characterized using a Rigaku X-ray diffractometer (XRD) with Cu  $K_{\alpha}$  radiation. The XRD patterns showed that the as-cast alloy had a fully glassy structure. The mechanical properties of the metallic glass were characterized using uniaxial compression and the SPT method. The nominal dimensions of the rectangular specimens for uniaxial compression were  $3.0 \times 3.0 \times 6.0$  mm. The SPT specimens, with a thickness of 0.7 mm and diameter of 10 mm, were cut from the Ti-based metallic glass ingots using an electric-spark cutting machine, and were then ground and polished to a thickness of 0.3-0.5 mm using  $1.5 \,\mu$ m diamond abrasive paste. The confinement discs were made of a cold-rolled Cu/11.6 at % Al alloy with a yield strength of 575 MPa and a total elongation of more than 10%, and the disc size was identical to that of the Ti-based metallic samples for SPT. The surfaces of the confinement discs were polished using 1.5 µm diamond abrasive paste. The samples without confinement (sample S1) and with confinement (sample S2) were tested to observe their difference in shear deformation and fracture. All of the mechanical tests were performed using an MTS810 testing machine at room temperature. After tests, all of the specimens were observed using a Leo Supra 35 scanning electron microscope (SEM) to reveal the deformation and fracture features.

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