A statistical physics consideration about the strength of small size metallic glass pillars

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Abstract. We have fabricated micro-/nano- pillars of a Zr-based metallic glass, Zr$_{50}$Ti$_{16.5}$Cu$_{15}$Ni$_{18.5}$, with pillar tip diameters ranging from ~750 nm to ~110 nm. These pillars were mechanically tested quantitatively in-situ in a Transmission Electron Microscope (TEM). Due to a slight tapering of ~3°, the deformation accommodated with shear bands is driven downwards from the top. However, the real time monitoring of shear bands evolution in-situ in TEM makes it possible to measure the effective load-bearing diameter, i.e., the diameter ahead of the most forefront shear band, which enables the precise measurement of yield strength of these pillars. Consequently, it turns out that the yield strength is essentially size independent and close to the bulk value. Statistical physics description of strength of small size metallic glasses is discussed, and it is concluded that Weibull Statistics is inappropriate in describing strength of metallic glasses at micro and nano length scales.

1. Introduction

Size effect on strength of metallic glasses receives recently increasing attention, due to their unique mechanical properties and potential application in microelectromechanical systems. Compared to single crystals which exhibit strengths times, or even orders, higher than their bulk counter part at micro- or nano- scale due to a dislocation source limited deformation [1-3], MGs obviously lack a dislocation involved mechanism due to the intrinsically disordered structure. Nevertheless, a couple of recent publications reported more or less size effects on strength of micrometer scaled specimens [4-8]. Lee et al [4] and Lai et al [5] reported up to 100 and 86 per cent increment of yield strength, respectively, of micrometer-diameter pillars of a Mg-based and a Zr-based MG over the values of bulk glasses, and interpreted the size effect according to Weibull statistics. Cheng et al [6] measured a yield stress of 2.25 GPa of micropillars of another Zr-base glass whose bulk value is 1.9 GPa. Schuster et al [7] reported modest increase (~9%) of the yield strength of Pd$_{40}$Ni$_{40}$P$_{20}$ micropillars over the bulk one, and later [8] attributed this effect to measurement artifacts. In contrast, experiments by Volkert et al [9] on micropillars of PdSi showed a slight decrease of strength with decreasing size, but it is still hard to conclude about a possible size effect because of experimental errors [9,10].

Apparently, there is at present no general consensus on how size effects influence the strength of metallic glasses. Accurate measurement of individual small metallic specimens is still a challenge mainly because of the difficulties in achieving uniform gauge lengths of pillars with diameters smaller than 1 µm. This is in fact one of the origins of the discrepancy in size effects reported in literature. Also the positioning of the specimen and alignment of the test system is a critical issue for extremely...
small specimens. In this letter we present quantitative in situ TEM compression of micro-/nano- MG pillars, which enables measuring pillars of diameters down to tens nm scale. Quantitative load response is correlated with dynamic observation of yielding evolution, which allows real time measurement of effective diameter bearing the externally applied load.

2. Experimental

Amorphous micropillars were fabricated by FIB (focused ion beam) from metallic glass ribbon which is Zr-based Zr$_{50}$Ti$_{16.5}$Cu$_{15}$Ni$_{18.5}$, by using An FEI Strata DB235 dual beam FIB-SEM system operating at 30 KV and 20 PA for final shaping. More than 40 pillars with tip diameter ranging from 110 nm to 750 nm and having well defined gauge lengths ranging from 1.2-2.0 µm, aspect ratios from 3 to 8, and small taper angles between 2.0-3.5º, are fabricated and studied.

In situ TEM compression experiments were performed using a recently developed Hysitron picoindenter TEM holder (Hysitron Inc., Minneapolis, MN) equipped on JEOL 2010F TEM, with a diamond flat punch of 2 µm diameter. It is integrated with a miniature capacitive load–displacement transducer that permits high resolution load and displacement measurements (resolution of ~0.3µN in load, ~1 nm in displacement) [1113]. A nominal strain rate of $-2\times10^{-3}$/s is used.

3. Results

The deformation of all specimens is dominated by intermittent shear banding. With the small tapering, the deformation is driven downwards from the top, leaving the lower part purely elastically deformed. The effective diameter was measured just ahead of the most forefront shear band, which is the minimum diameter at the purely elastically deformed region. Although dynamic measurement was continuously performed, to ensure accurate measuring of effective diameter which is the main source of error, a compression test was normally interrupted at a certain stage to acquire images still after unloading. The recorded peak load before unloading divided by measured diameter was taken as yield strength. A typical example is illustrated in Figures. 1. It can be seen that the engineering stress taking into account the diameter change (taper angle) during compression still overestimates the results. However, the yield stresses calculated by measuring the effective diameter in two successive test cycles (indicated in the curve) are essentially consistent (1.73GPa), indicating an accurate measurement.

The values of the yield strength measured from a large number of Zr-based micropillars are plotted in Figure. 2 as a function of effective pillar diameter. With the accurate measurement of load-bearing diameter, the error bars indicate very small uncertainties in the measurement of the yield strength. It turns out that there is no dependence of the yield stress on pillar diameter over the current size scale. The result is understandable. As well known the high yield strength of metallic glass is due to the lack of an ‘easy’ flow mechanism like ‘dislocation sliding’ in crystalline materials. As a consequence, in metallic glass one would not expect a “smaller is stronger” phenomenon observed in nanocrystalline materials due to dislocation starvation mechanism. Strength of BMGs is essentially controlled by interatomic bonding and has a roughly linear relationship with elastic modulus as $\sigma_y = 0.02E$ or $\tau_y = 0.027G$ [14], where $\sigma_y$ and $\tau_y$ are normal and shear fracture strength, E and G, Young’s modulus and shear modulus, respectively.

4. Discussion

This finding is different from the refs [4-7], but is consistent with refs [8,9]. When inspecting literature about the yield strength of pillars it should be kept in mind that, by assuming a tip diameter or an average diameter as the pillar size without taking into account the dynamical evolution of the effective diameter during compression, an artificial size effect will always be attained, since a
constant tapering angle has larger influence on thinner pillars. This kind of error however is circumvented here by in-situ dynamically measuring the effective diameter as mentioned above.

**Figure 1.** TEM micrographs showing the deformation morphology of a pillar of 270 nm tip diameter subjected to two loading-unloading cycles: (a) before compression and (b) and (c) after unloading of each compression cycle with the yield stress indicated; the effective load bearing diameter is measured just ahead of the most forefront shear band as illustrated in (b) and (c); (d) engineering stress vs displacement curve, with the yield stress measured in-situ in TEM is also indicated.

**Figure 2.** Yield stress versus pillar diameter for both metallic glasses. The error bars reflect the uncertainties in the stress due to actual pillar diameter, which are minimized by measuring the effective diameter in the in situ TEM images.

It would be also interesting to make a connection to a statistical physics description of strength by investigating the dependence of the failure stress on the effective size. The reported small size induced increase in yield stress has been interpreted according to Weibull statistics (WS). In WS [15] the probability of failure at a stress $\sigma$ is expressed by $P_f(\sigma)=1-\exp[-(\sigma/\sigma_0)^m]$, where $\sigma_0$ is a scale parameter, $m$ Weibull modulus. This equation is widely used in fitting of $\ln[-\ln(1-P_f)]$ versus $\ln \sigma$, with $m$ being the slope. A size dependence of the mean value of $\sigma$ can be derived, which is described by $\sigma_1/\sigma_2=(V_2/V_1)^{1/m}$ or generally $\sigma \propto V^{-1/m}$. If WS would be continuously extrapolated from macro- to micro-/nano- scale specimens, the strength of submicrometer MGs would be exceptionally high. The reported size independent strength in this work indicates that the Weibull statistics cannot describe the nature of strength data of submicrometer scale MGs.

WS does not apply due to the fact that it does not make a distinction between nucleation, propagation and overlap of stress fields. WS is derived based on the weakest link hypothesis that fracture occurs once the fracture of the weakest link occurs and there is no interaction among flaws. The size effect described is due to the fact that a WS material has high probability of having a larger destructive defect in a larger volume [15]. Together with a Griffith failure criterion, sizes of the destructive flaws $a_{c_1}$ and $a_{c_2}$ in $V_1$ and $V_2$ respectively are correlated by $a_{c_1}/a_{c_2}=(V_1/V_2)^{1/m}$. Clearly, very small flaws become fracture origins in small specimens. In fact, an inverse-power-law
distribution of flaw size (with the exponent being \(-m/2\)) is a necessary condition of WS [16]. This tendency of a large amount of smaller flaws acting as fracture (or yield) origin in smaller specimens may put the applicability of WS in small specimens questionable. It has been recognized that materials become insensitive to flaws when the flaw size is smaller than 30 nm [17], since the widely used engineering concept of stress concentration is no longer valid for such small flaws.

Although WS may be useful in describing the strength of BMGs [18,19] when the strength is limited by large flaws (>micrometer), like as-casting pores, inclusions, surface irregularities, which act effectively as stress-concentrators, it is not applicable to sub-micrometer MG pillars as shown above.

5. Conclusions
We have fabricated micro-/nano- pillars of a Zr-based metallic glass, Zr_{50}Ti_{16.5}Cu_{15}Ni_{18}, with pillar tip diameters ranging from ~650 nm to ~90 nm. These pillars were mechanically tested in-situ in TEM as a function of pillar diameters. For other details see [20]. It turns out that the yield strength of the metallic glass is close to those of the bulk glass and is essentially size independent. Statistical physics description of strength for small size metallic glass is discussed, and Weibull statistics is found inappropriate in describing the strength of metallic glasses at micro and nano length scales.

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