



Contents lists available at ScienceDirect

## Journal of Non-Crystalline Solids

journal homepage: [www.elsevier.com/locate/jnoncrysol](http://www.elsevier.com/locate/jnoncrysol)

## The correlation between weakest configurations and yield strength of Zr-based metallic glasses

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## ARTICLE INFO

## Keywords:

Metallic glasses  
Yield strength  
Solvent atoms  
Free volume

## ABSTRACT

A direct relationship between the yield strength and the atomic ratio of solvent (Zr) atoms in the Zr-Cu-Al-Ni metallic glasses system is firstly uncovered. It is found that either shear modulus or yield strength decreases almost nearly with the increase in atomic ratio of Zr. The origin of this relationship is ascribed to the preferential straining of the weakest configurations, which consist of the solvent-solvent bonds and the free volume concentrated in them. It is suggested that a higher atomic ratio of Zr corresponds to a larger amount of weakest configurations, which will facilitate the activation and the accumulation of the shear transformations and finally results in the lower yield strength. This finding may provide an effective strategy for designing high-strength metallic glasses by modifying the chemical composition.

## 1. Introduction

The structural picture of metallic glasses becomes clearer and clearer with durative efforts [1–5]. The solute-centered clusters and their efficient packing construct the short- and/or medium-range order. The long-range disorder can be achieved by linking these ordered clusters through excessive solvent atoms. Therefore, it is accepted that these structures on different length scales will determine the physical properties from relaxation (fast or slow) to deformation (elastic or plastic) [6–9]. For instance, Ma et al. [8] found that metallic glasses inherit shear/Young's modulus from their solvent atoms. This surprising finding substantiates the idea that the bonds of solvents could determine the shear modulus on the long-range scale [7,8]. Moreover, these loosely packed solvent-solvent junctions usually lead to additional scattering of transverse phonons in the THz region, which is quite common for various glasses [10,11]. Very recently, Ding et al. [12] introduced the concept of “flexibility volume”, a volume-scaled vibrational mean square displacement of THz phonons, which can predict the shear modulus of metallic glasses quantitatively. This further confirms that solvent-solvent junctions can directly dominate the shear modulus of the metallic glasses. Meanwhile, it is well known that the shear modulus controls the energy barrier for relaxation and shear flow, as indicated in both the shoving model of Dyre [13] and the cooperative shear model of Johnson and Samwer [14]. Specifically, the yield strength in shear mode shows a linear relationship to the shear modulus

with respect to the atomic ratio of solvent atoms for a variety of metallic glasses [15,16]. It is therefore expected that the solvent-solvent junctions will determine the plastic yielding of metallic glasses beyond the elastic range. However, this needs further experimental evidence.

In the present work, a series of metallic glasses with identical chemical constituents of Zr-Cu-Al-Ni but different atomic ratio were selected as the model system. It is clearly demonstrated that the yield strength (under compression) as well as shear modulus of these glasses strongly depend on the contents of their solvent (Zr) atoms. Based on structural observations on parts of these materials, the dependence of shear modulus and yield strength on the atomic ratio of Zr can be understood from the weak links that locate in the solvent-rich configurations with relatively high excessive free volume.

## 2. Experiments

Ingots with the compositions of  $\text{Zr}_{70}\text{Cu}_{13.5}\text{Al}_8\text{Ni}_{8.5}$  ( $\text{Zr}_{70}$ ),  $\text{Zr}_{64.13}\text{Cu}_{15.75}\text{Al}_{10}\text{Ni}_{10.12}$  ( $\text{Zr}_{64}$ ),  $\text{Zr}_{55}\text{Cu}_{30}\text{Al}_{10}\text{Ni}_5$  ( $\text{Zr}_{55}$ ) and  $\text{Zr}_{50.7}\text{Cu}_{28}\text{Al}_{12.3}\text{Ni}_9$  ( $\text{Zr}_{50}$ ) were prepared by arc-melting high purity Zr, Cu, Al and Ni in a argon atmosphere. Ti-getter was used to further reduce the oxygen partial pressure in the melting chamber. Cylindrical rods with a dimension of  $\Phi 3 \times 30$  mm were prepared via the copper-mold suction-casting method under the same condition. The glassy state of all rods was verified by both X-ray diffraction and high resolution transmission electron microscopy (HRTEM, JEOL JEM-2100F). The

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**Table 1**

Summary of shear modulus ( $G$ ), yield strength ( $\sigma_y$ ), glass transition temperature ( $T_g$ ) and molar volume ( $V_m$ ) of 11 kinds of Zr-Cu-Al-Ni metallic glasses.

| Alloys.                                                                      | $G \pm 5\%$ (GPa) | $\sigma_y$ (GPa) | $T_g \pm 5$ (K) | $V_m$ (cm <sup>3</sup> /mol) |
|------------------------------------------------------------------------------|-------------------|------------------|-----------------|------------------------------|
| Zr <sub>72</sub> Cu <sub>13</sub> Al <sub>7.5</sub> Ni <sub>7.5</sub>        | 27                | 1.54 [17]        | 625 [17]        | 12.24                        |
| Zr <sub>70</sub> Cu <sub>13.5</sub> Al <sub>8</sub> Ni <sub>8.5</sub>        | 27.3              | 1.57 [18]        | 625 [18]        | 12.11                        |
| Zr <sub>65.5</sub> Cu <sub>22.4</sub> Al <sub>5.6</sub> Ni <sub>6.5</sub>    | 28.9              | 1.60 [20]        | 636 [19]        | 11.74                        |
| Zr <sub>65</sub> Cu <sub>15</sub> Al <sub>10</sub> Ni <sub>10</sub>          | 30 [15]           | 1.65 [21]        | 652 [21]        | 11.74                        |
| Zr <sub>64.13</sub> Cu <sub>15.75</sub> Al <sub>10</sub> Ni <sub>10.12</sub> | 28.5 [22]         | 1.69 [22]        | 646 [22]        | 11.75                        |
| Zr <sub>62</sub> Cu <sub>15.4</sub> Al <sub>10</sub> Ni <sub>12.6</sub>      | 29 [16]           | 1.81 [16]        | 652 [16]        | 11.67                        |
| Zr <sub>61.88</sub> Cu <sub>18</sub> Al <sub>10</sub> Ni <sub>10.12</sub>    | 29.1 [23]         | 1.73 [23]        | 653 [23]        | 11.60                        |
| Zr <sub>57</sub> Cu <sub>27</sub> Al <sub>11</sub> Ni <sub>5</sub>           | 32.7              | 1.84 [24]        | 682 [24]        | 11.32                        |
| Zr <sub>55</sub> Cu <sub>30</sub> Al <sub>10</sub> Ni <sub>5</sub>           | 32.5              | 1.82 [25]        | 685 [25]        | 11.15                        |
| Zr <sub>53</sub> Cu <sub>18.7</sub> Al <sub>16.3</sub> Ni <sub>12</sub>      | 33.4              | 1.88 [26]        | 709 [28]        | 11.16                        |
| Zr <sub>50.7</sub> Cu <sub>28</sub> Al <sub>9</sub> Ni <sub>12.3</sub>       | 34.8              | 1.90 [27]        | 719 [28]        | 10.79                        |

thermal properties of these specimens were characterized by differential scanning calorimetry (DSC). The DSC tests were conducted in a TA Q2000 thermal-analysis instrument with a heating rate of 0.33 K/s in a flow of argon. The base line for each DSC measurement was obtained by a second run under identical conditions. With the purpose of ensuring the reliability of the experimental data, at least three specimens for each of the metallic glass have been tested on the same instrument under identical conditions. Meanwhile, qualitative distribution of free volume in each specimen was investigated using the weak-phase object HRTEM imaging method. The specimens for both the DSC measurements and the HRTEM analyses were cut from the same position of the as-cast rods and then prepared in standard mechanical grinding to the same thickness. Furthermore, the grinded cross-sectional HRTEM specimens were milled by low-energy ion beam thinner under the same conditions.

### 3. Results

Table 1 presents the shear moduli ( $G$ ), yield strengths ( $\sigma_y$ ), glass transition temperatures ( $T_g$ ) and molar volumes ( $V_m$ ) of a series of Zr-Cu-Al-Ni metallic glasses with different solvent-Zr content ( $C_{Zr}$ , at.%) [15–28]. Here, all of the yield strengths are summarized from literatures, which were carried out under uniaxial compressions at quasi-static strain rates. Under such conditions, the yield strength is an inherent material property and should be almost constant for a given metallic glass [29,30]. The shear moduli in both the literatures and present work were measured by ultrasonic method, which has an accuracy better than 5% [31–33]. The maximum error value of 5% is adopted in the current work. The glass transition temperatures are also summarized from literatures, which were measured by DSC. The random errors of the measured  $T_g$  is usually less than 5 K [34–36]. Here, an experimental error of 5 K for the glass transition temperature is taken. The values of molar volumes are calculated according to the rule of mixtures [37]. As pointed out previously [15,16,38], both shear modulus  $G$  and yield strength  $\sigma_y$  showed an approximately linear dependence on the value of  $T_g/V_m$ . For the current studied materials, these relationships are quite similar as presented in Fig. 1a. Such correlations imply a similarity between plastic yielding and glass transition in metallic glasses [16,22]. Furthermore, the variations of shear modulus as well as yield strength with the solvent-Zr content are plotted in Fig. 1b. It is interesting to find that either shear modulus or yield strength has a nearly linear reduction with increasing the atomic ratio of Zr in these Zr-Cu-Al-Ni metallic glasses. In addition, the shear moduli of these glasses are less than that of pure Zr, indicating a deviation from modulus' inheritance [8,39,40]. Moreover, the deviation becomes larger with increasing the atomic ratio of Zr.

The DSC curves of the Zr<sub>70</sub>, Zr<sub>64</sub>, Zr<sub>55</sub> and Zr<sub>50</sub> glasses are shown in Fig. 2. All curves show the distinct feature of a glass transition (marked by the arrows) before crystallization. The glass transition temperatures

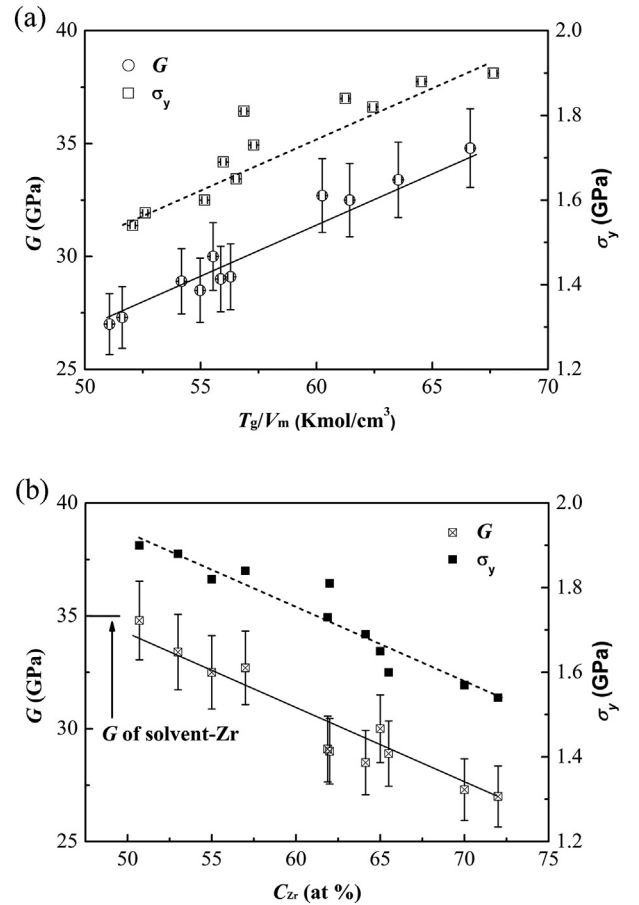


Fig. 1. The variation of shear modulus ( $G$ ) and yield strength ( $\sigma_y$ ) with (a) the parameter combination ( $T_g/V_m$ ) and (b) the atomic ratio of Zr ( $C_{Zr}$ , at.%).

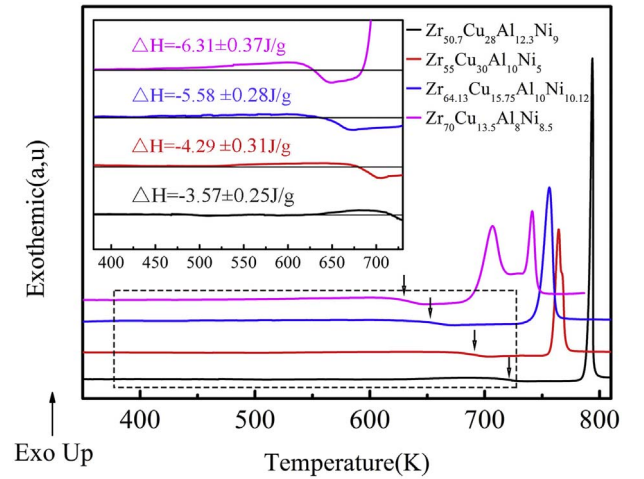


Fig. 2. DSC curves of the four Zr-Cu-Al-Ni metallic glass samples. The inset shows the different exothermic signals before the glass transition among these samples.

of the four metallic glasses were measured to be about 627 K, 651 K, 690 K and 719 K, respectively. This indicates that the decrease of Zr content leads to an increase of  $T_g$ . Importantly, the exothermic signals before the glass transition display an apparent difference among these samples, which is presented in the inset of Fig. 2. It is well known that the exothermic enthalpy ( $\Delta H$ ) prior to glass transition can be directly related to the excessive free volume ( $v_f$ ) by an empirical relationship of  $\Delta H = \beta' v_f$  [41,42] with a constant  $\beta'$ . The values of  $\Delta H$ , shown in the inset of Fig. 2, indicate that the free volume concentration decreases

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