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The influence of structural relaxation on the mechanical properties of $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ bulk metallic glass

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Abstract

$Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ bulk metallic glass (BMG) was annealed at 573 K under a high pressure of 3 GPa. The effects of the structural relaxations of BMG on its mechanical and acoustic properties are investigated by using x-ray diffraction, ultrasonic study, density measurements, compression testing, as well as microhardness. It is found that high-pressure relaxation results in a microstructural transformation in the BMG. A sample relaxed under high pressure exhibits 3% plastic strain compared with the crystallized form, and the compression strength achieved, 1800 MPa, is improved by nearly 20% over that for the as-prepared sample; moreover, a BMG with relaxed structure exhibits markedly different acoustic properties.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Subtle rearrangement of atoms in a material upon annealing result in a decrease of the free energy, which is a well known structural relaxation phenomenon. Many physical properties sensitive to local atomic structure—for example, atomic diffusivity, viscosity, ductility, magnetic anisotropy—are affected the structural relaxation [1, 2]. Therefore, we should be able to relate changes in physical properties to the variations in the structure. Recent innovations in processing metallic glasses in bulk form permit important mechanical properties to be tested and acoustic study of them [3–6]. Besides conventional methods, such as energy dispersive x-ray diffractometry (EDXD) [7] and differential scanning calorimetry (DSC) [8], both mechanical property testing and ultrasonic measurement could be used for the study of structural relaxation in bulk metallic glasses (BMGs). Ultrasonic measurement provides a powerful tool

for studying the structure of matter, and thus important information about the structural as well as vibrational characteristics can be obtained [9]. Moreover, the more open geometry of the BMGs makes them more amenable to measurements of elastic wave propagation compared to conventional metallic glasses [10]. The structural relaxation under high pressure is investigated by contrasting differences in mechanical as well as acoustic properties between a relaxed sample and an as-prepared one. It is found that structural relaxation leading to an increase in both ductility and strength.

2. Experimental details

Ingots were made by arc-melting a mixture of elements in a Ti-gettered argon atmosphere, then remelted in a vacuum-sealed quartz tube and quenched in water to produce an amorphous rod with diameter 5 mm. The amorphous nature as well as the homogeneity of the BMG were ascertained by x-ray diffractometry (XRD), DSC, and transmission electron microscopy (TEM) [11]. The $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ alloy samples were cut from rods with a length of 5 mm, for mechanical and ultrasonic measurements.

The BMG was relaxed under high pressure. The samples were embedded into sodium chloride. The sample was annealed at a temperature of 573 K under 3 GPa pressure for 2 h. The temperature was determined by NiCr–NiAl thermocouples placed inside the high-pressure cell. The details of the HP experiment were described at length in [12].

The crystallization experiment was performed in a furnace with a vacuum of 10^{-5} T for 2 h at 673 K. The relaxed and crystallized samples were inspected by XRD.

Compression tests were performed on an MTS 810 axial load frame. The compressive displacement rates were 0.2 mm min^{-1}. The Vickers microhardness was measured at a testing load of 200 gf and the hold time was 15 s following microscopy. The acoustic velocities were measured by using a pulse-echo overlap method [13]. The measuring sensitivity was of the order of 0.5 ns. The density was measured by the Archimedean technique and the accuracy was within 0.1%.

3. Results and discussion

The XRD pattern for the BMG annealed under high pressure exhibits no difference as compared to that of the as prepared alloy—this indicates that the crystallization does not occur while that for the one annealed at 673 K in vacuum shows crystallization peaks. The compressive stress–strain curves are shown in figure 1. Alloys A and B show 1 and 3% strain plastic deformation, respectively; however, no distinct plastic deformation is observed in alloy C. In contrast to these three curves, high values of both ductility and strength were found for alloy B.

Macroscopic information about the natures of the deformations of alloys B and C were obtained by scanning electron microscopy (SEM). Figures 2(a) and (b) show the appearance of the surface of alloy B, and alloy C, after fracture, respectively. The fracture of alloy B is at an angle of about 45° while that of alloy C is at nearly 0°. Alloy B exhibits similar XRD patterns to alloy A; however, the marked difference in mechanical properties indicates distinctly different structures of the three alloys. In order to further verify this difference, parameters sensitive to microstructural change such as the acoustic velocities, microhardness, and density were measured for the three alloys; the results are listed in table 1. For alloy B, in the structural relaxation state under high pressure, the acoustic velocities v, elastic constant E, Debye temperature θ_D, microhardness H_V, and stress σ are close to those for the crystallized sample (alloy C); however, the relative density change is 0.36%, which is much larger than that

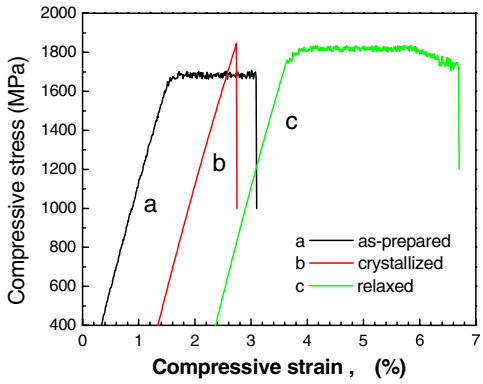

Figure 1. The compressive stress–strain curves: (a) for alloy A, (b) for alloy C, and (c) for alloy B.

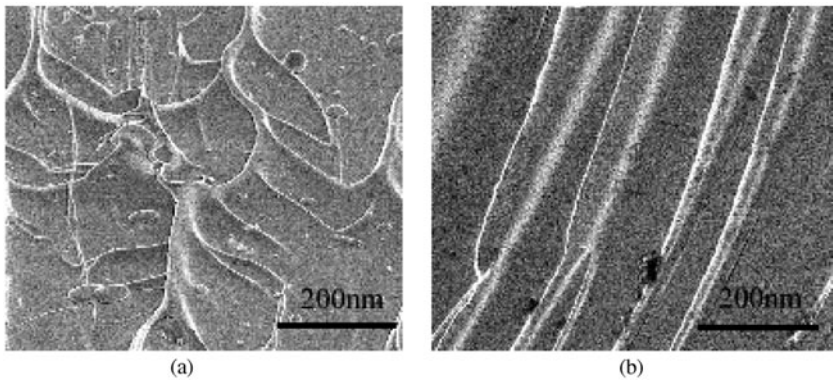

Figure 2. SEM micrographs of fracture surface: (a) alloy B; (b) alloy C.

Table 1. The properties of the ZrTiCuNiBe samples in the as-prepared state (alloy A), the relaxed state held at 573 K under 3 GPa for 2 h (alloy B), and the crystallized state held under ambient pressure at 673 K for 2 h (alloy C).

Sample	H_V (GPa)	E (GPa)	ρ (g cm^{-3})	$\Delta\rho/\rho$	v_1 (km s^{-1})	v_s (km s^{-1})	θ_D (K)	σ (MPa)	ε (%)
Alloy A	5.94	101.2	6.125		5.17	2.47	326.8	1658	1
Alloy B	6.24	118.7	6.341	0.36	5.30	2.65	350.7	1800	3
Alloy C	7.03	124.2	6.201	0.12	5.33	2.75	364.2	1846	0

of alloy C (0.12%), though it has a similar XRD pattern to the as-prepared sample (alloy A). The mechanical and acoustic results confirm that a microstructural transformation in ZrTiCuNiBe BMG is induced by high pressure.

It is well known that the crystalline phases in the ZrTiCuNiBe BMG are usually intermetallic compounds [14], which are commonly supposed to exhibit large hardness and to be brittle. Upon loading, they would not deform and would act as obstacles preventing shear bands from moving; the yield stress is raised while at the same time ductility decreases, because the shear bands, in which plastic deformation takes place, cannot move freely through the material [15]. Thus, the increase in strength and concurrent decrease in ductility in the crystallized ZrTiCuNiBe specimen compared with the as-prepared one is attributed to the supposed hardness and brittleness of the compounds, mentioned above. However, the increase

in both strength and ductility in the relaxed specimen which is still in glassy state we assume to be attributable to a special medium-range order (MRO) microstructure induced by high-pressure annealing [16]. In contrast to the short-ranged order (SRO) in the as-prepared BMG, the so-called MRO is considered to be an ordered configuration in amorphous phases of 0.5–2 nm—which is larger than the critical nuclei—but which does not yield a characteristic crystalline reflection in XRD or other diffraction patterns [10, 17]. The HRTEM picture confirmed the MRO structure in the BMG after HP annealing. Small nuclei of 1–2 nm can be clearly seen [18]. As the structural scale reduces to the nanometre range, the interfacial effects, which are caused by the intrinsically high interface-to-volume ratio of these small-dimension nanoparticles, can dominate the microstructure and properties, leading to unusually large strains [19]. In the ordered configuration in the amorphous phase, the particles, 2 nm in diameter, have tremendous interface-to-volume ratios of around 3 mm^2 mm^{-3} [20], which probably leads to mechanical properties different from those of the crystallized material with the usual grain size; increases in both strength and ductility were observed in the structurally relaxed specimen with MRO.

4. Conclusions

In conclusion, high-pressure-induced relaxation results in a microstructural transformation in the BMGs. A BMG with relaxed structure exhibits markedly different acoustic and mechanical properties to the as-prepared and crystallized ones. The structural relaxation leads to increases in both ductility and strength. High-pressure annealing of BMGs below the glass transition temperature may provide us with a new channel for improving the mechanical properties of BMGs and further enhancing their practical applicability as engineering materials.

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