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Atomic configuration evaluation of $Zr_{60}Ni_{21}Al_{19}$ bulk metallic glass under high pressure

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ABSTRACT

The atomic configuration evaluation in $Zr_{60}Ni_{21}Al_{19}$ bulk metallic glass at high pressures has been revealed by using *in situ* synchrotron X-ray diffraction. The radial distribution function is gained by Fourier transformation. The investigation shows that the amorphous structure is retained and the coordination number keeps 12.0 within the experimental pressures (0–24.5 GPa). The quantitative determination of the neighbor atomic distance suggests that high pressure alters topological but not chemical short range ordering through shortening the second nearest neighbor atomic distance. The atomic coordination is analyzed by the inherent chemical parameters of the ternary $Zr_{60}Ni_{21}Al_{19}$ amorphous alloy.

1. Introduction

Because of high strengths, ductilities in compression, low coefficients of friction, high wear resistance, high corrosion resistances [1–5], multicomponent Zr-based glass bulk metallic glasses (BMGs) with a larger geometry by a conventional casting process with a low cooling rate are being widely studied [6–8]. The structural evaluation of metallic glass contains a series of continuous change on the atomic level which is linked to the atomic short range ordering (SRO) or middle range ordering (MRO) [9,10]. Many physical properties sensitive to local atomic structure, for example, atomic diffusivity, viscosity, ductility, magnetic anisotropy, are affected by the structural change. Energy dispersion X-ray diffraction is one of the simplest and most effective ways to investigate this kind of change in atomic level. Since the diffraction data of the material under high pressure can be gained by high-strength synchrotron radiation source and radial distribution function (RDF) can be obtained by these data, the change of the amorphous structure can be analyzed on the atomic level. High pressure, which is the simplest way to vary the interatomic distance of substance and thus change its physical properties [11,12], has been employed as an important means to investigate the structural evolvement of multicomponent Zr-based BMGs under high pressure [8,13,14]. However, since ternary Zr-based BMGs are relatively new materials, little is known about some aspects of the materials under high pressure. In particular, the measurements of compression properties are of great importance to understand the relationship between unique mechanical properties and configuration changes under pressure.

In this paper, the compression behavior of the structure associated with changes in the atomic rearrangement of a new $Zr_{60}Ni_{21}Al_{19}$ BMG under high pressure at room temperature has been unraveled using energy dispersive X-ray diffraction with a synchrotron radiation source, and the change of amorphous structure and atomic configuration are discussed using atomic size difference and heat of mixing.

2. Experimental

$Zr_{60}Ni_{21}Al_{19}$ ingots were prepared by arc-melting the mixture of pure metals in Ti-gettered argon atmosphere. $Zr_{60}Ni_{21}Al_{19}$ BMG was produced by suction of the melt into a copper mold to get cylindrical rods 2 mm in diameter. The composition was quantified to be $Zr_{60}Ni_{21}Al_{19}$ by chemical analyses. The amorphous nature as well as the homogeneity of the $Zr_{60}Ni_{21}Al_{19}$ metallic glass was ascertained by X-ray diffraction.

Some powder was prepared from the amorphous rod for pressure experiments. The pressure was generated by using a diamond anvil cell (DAC) driven by an accu-

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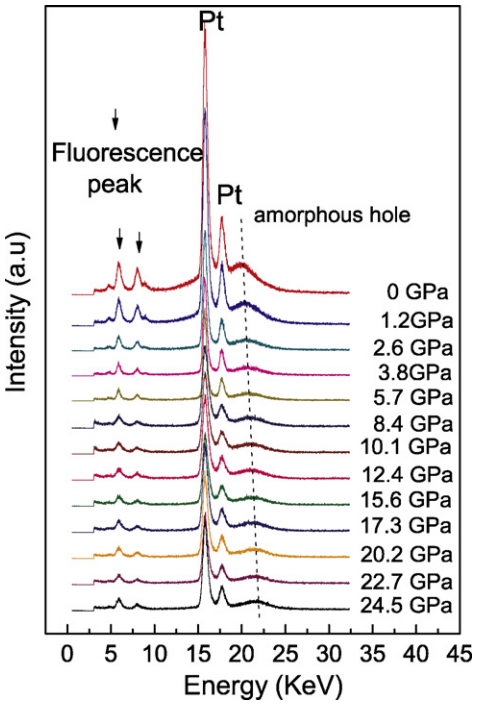

Fig. 1. Energy dispersion X-ray diffraction patterns of $Zr_{60}Ni_{21}Al_{19}$ bulk metallic glass under various pressures.

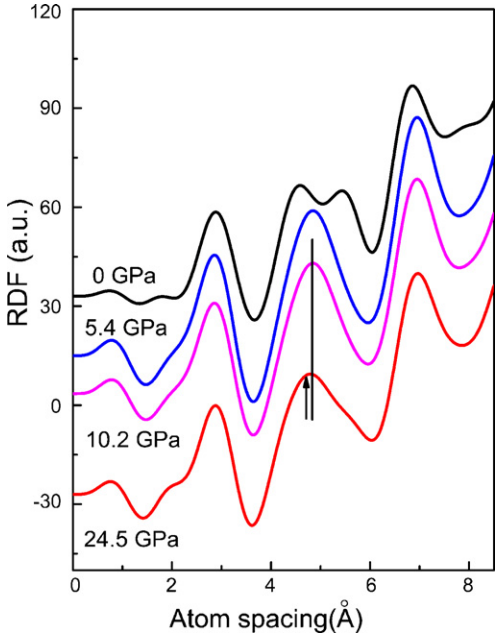

Fig. 2. Radial distribution functions of $Zr_{60}Ni_{21}Al_{19}$ bulk metallic glass derived from *in situ* X-ray measurements at various pressures.

rately adjustable gear-worm-level system. Pt is used to be pressure calibrators. The amorphous powder sample together with the selected pressure-calibrator powder was loaded into a 200-μm diameter hole of a L301 stainless steel gasket, which was preindented to a thickness of about 20 μm. A mixture of methanol, ethanol and water was used as the pressure-transmitting media. The *in situ* high-pressure energy dispersive X-ray diffraction measurements were carried out in Beijing Synchrotron Radiation Laboratory (BSRL). The size of X-ray spot was 55 μm × 55 μm with the storage ring operating at 2.8 GeV and 40–70 mA. A Si (Li) detector was used to collect the diffraction signal under various pressures. The experimental pressure was determined from the position of (1 1 1) diffraction peak of Pt, as its equation of state (EOS) was well established. The basic relation in the energy dispersion technique follows:

$$E_{hkl}d_{hkl}\,(\mathrm{keV\,nm}) = \frac{hc}{2\sin\theta} = \frac{0.61993}{\sin\theta}, \quad (1)$$

where E_{hkl} is the energy at which the diffraction line (hkl) located, d is the interplane spacing, h and c are the Planck constant and the speed of light in vacuum. The diffraction angle θ was determined to be 6.87° in the experiment.

3. Results and discussion

Fig. 1 shows synchrotron radiation X-ray diffraction spectrum for the $Zr_{60}Ni_{21}Al_{19}$ BMG at selected pressures. The sharp peaks are fluorescence peaks coming from Pt. With the pressure increasing, the position of fluorescence peaks remains unaltered, but the broad diffusive amorphous hole obviously shifts to the higher energy (shown by the dot line). No any new diffraction peaks is detected at pressures ranging from 0 to 24.5 GPa. This means that the structure of the BMG is quite stable at room temperature. Although the synchrotron X-ray diffraction patterns recorded were still that of an amorphous alloy, fine structural variations may occur through investigation of structure factor $S(q)$. The total structural factor obtained by using Faber and Ziman formula [15], $S(q)$ is related to the radial distribution function $4\pi r^2\rho(r)$ by the Fourier transform:

$$S(q) - 1 = \int_{r=0}^{\infty} \frac{4\pi r^2[\rho(r) - \rho_0]\sin(qr)}{(qr)\ \mathrm{d}r}, \quad (2)$$

where $\rho_0 = \lim_{r\to\infty}\rho(r)$ is the average density of atoms, r the interatomic distance from the nearest neighbor atom. The RDF is derived from the synchrotron X-ray diffraction data by Fourier transformation using a computer program (the detail of the calculation can be found in Ref. [13]). RDF curves under different pressures are shown in Fig. 2.

It can be seen that only at ambient pressure, the second peak split in the RDF curve of the BMG, which is the character of the amorphous alloy. With the increase of pressure, the split disappears, and the maximum of peak shifts to left at higher pressures (see the curve of 24.5 GPa), but the first and the third peaks keep original positions within the experimental range.

The atomic size [16], heat of mixing (ΔH^{mix}) [17] and the atomic percentage of constituent elements are important to understand the local atomic configuration for the Zr-based BMG. Fig. 3 shows characteristics of the $Zr_{60}Ni_{21}Al_{19}$ ternary BMG: Zr is illustrated with closed circle in black, Ni in darkness gray and Al in white. The mixing enthalpy values of $\Delta H^{\mathrm{mix}}_{\mathrm{ZrAl}}$, $\Delta H^{\mathrm{mix}}_{\mathrm{ZrNi}}$, and $\Delta H^{\mathrm{mix}}_{\mathrm{NiAl}}$ are also shown. As we can see, Zr is the main constituent element with the highest composition in the $Zr_{60}Ni_{21}Al_{19}$ ternary BMG system (with 60 at.%). The largest negative mixing enthalpy in the system is $\Delta H^{\mathrm{mix}}_{\mathrm{ZrNi}}$ ($-49\,\mathrm{kJ\,mol^{-1}}$), and then is $\Delta H^{\mathrm{mix}}_{\mathrm{ZrAl}}$ value of $-44\,\mathrm{kJ\,mol^{-1}}$.

Three peaks of RDF, respectively, correspond to the first, second and third nearest neighbor atomic distance. The atomic radii of Zr, Al, and Ni are 1.60, 1.43 and 1.24 Å, respectively [16]. The first peak indicates the nearest neighbor interatomic distance, and the value is of 2.86 Å, which is closed to the pair of Zr–Ni (2.84 Å). Moreover, among Zr, Ni and Al, Zr–Ni exhibits the largest negative

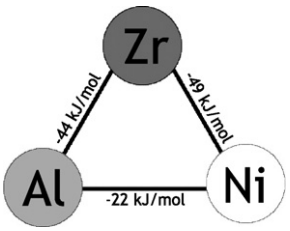

Fig. 3. Atomic radii of Zr, Ni, and Al, and mixing enthalpies of $Zr_{60}Ni_{21}Al_{19}$ bulk metallic glass.