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Elastic behaviour and microstructural characteristics of Nd₆₀Al₁₀Fe₂₀Co₁₀ bulk metallic glass investigated by ultrasonic measurement under pressure

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Abstract

An ultrasonic pulse–echo method was used to measure the transit time of longitudinal and transverse (10 MHz) elastic waves in a Nd₆₀Al₁₀Fe₂₀Co₁₀ bulk metallic glass (BMG). The measurements were carried out under hydrostatic pressure up to 0.5 GPa at room temperature. On the basis of experimental data for the sound velocities and density, the elastic moduli and Debye temperature of the BMG were derived as a function of pressure. Murnaghan's equation of state is obtained. The normal behaviour of the positive pressure dependence of the ultrasonic velocities was observed for this glass. Moreover, the compression curve, the elastic constants, and the Debye temperature of the BMG are calculated on the basis of the similarity between their physical properties in the glassy state and those in corresponding crystalline state. These results confirm qualitatively the theoretical predictions concerning the features of the microstructure and interatomic bonding in the Nd₆₀Al₁₀Fe₂₀Co₁₀ BMG.

Since amorphous solids do not have long-range order, a determination of the positions of all the atoms becomes impractical. Therefore, the structure of amorphous solids is normally described in terms of short-range atomic order existing in amorphous solids [1]. Previous studies showed that the bulk metallic glass (BMG) consisting of a mixture of atoms with different atomic sizes had a highly dense randomly packed structure as compared to conventional metallic glasses (MGs) [2]. The structural characteristic for glasses was found to be related to the Poisson ratio σ , which characterizes the relative values of the compressive and shear deformations

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of a solid. For example, the oxide glasses, with relatively small σ (0.15–0.25) [3, 4], are brittle, since atoms or molecules can hardly rearrange themselves in response to shear strains without drastic disturbance in the bonding configurations. In contrast, the conventional MGs (e.g. $\text{Pt}_{60}\text{Ni}_{15}\text{P}_{25}$, $\sigma = 0.421$) with poor glass formation ability have higher values of σ [5], indicating ease of atomic rearrangement and consequently ductile plastic deformation of these materials. For the BMGs with excellent glass formation ability, such as Zr-based ($\sigma = 0.34$) and Pd-based ($\sigma = 0.397$) [6, 7] ones, the value of σ is between those of the conventional MGs and oxide glasses. The nature of the chemical bond determines the microstructure in a solid; thus a difference in microstructure will influence the mechanical properties of a solid, resulting in variation of the elastic properties. The comparison indicates that σ has a correlation with the atomic mobility in these glasses. It is also known that the ratios between the bulk and shear moduli K/G are about 1.7 and 5.2 for amorphous carbon [3, 8] and for conventional MGs [5], respectively, while the values of K/G are 2.71 and 4.5 for the Zr-based and Pd-based BMGs, respectively [6, 7]; the marked difference indicates the difference in microstructural characteristics between the three types of glass, and the metallic bond is retained in BMGs even though they lack long-range order. The previous work demonstrates that the atomic close-packed configurations have a close correlation with the elastic behaviour of the glasses, and the elastic constants are intimately related to physical, mechanical, and thermodynamic properties of materials. The results provide important information about the structural and vibrational characteristics of metallic glasses [9, 10]. In recent years, some experimental effort has been expended on BMGs leading to a considerable advancement in the understanding of their elastic behaviour under pressure [6, 7, 11, 12]. It has been shown that the compression of $\text{Pd}_{40}\text{Ni}_{20}\text{P}_{20}$ metallic glass is similar to those of crystalline nickel and palladium [12], and that the compression curves of Zr-based and Pd-based BMGs have a correlation with those for their metallic components and represent a rough average of those for these elements, indicating that the short-range order structure of the BMGs has a close correlation with the atomic configurations of their metallic components [7, 11]. In this study, the hydrostatic pressure dependence of the sound velocities of a $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG, which is a hard magnetic material and possesses highly ordered microstructure [13, 14], was measured by the ultrasonic pulse–echo technique. Based on the compressibility and elastic constants of its constituents, quantitative estimates of the compression curve and elastic constants of the BMG are given. The calculated results agree well with experimental data obtained from ultrasonic measurement.

$\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG was prepared by a die casting method; the details of the experimental procedure are given in [13]. The structure of the samples was characterized by means of x-ray diffraction (XRD) using a MAC M03 XHF diffractometer with $\text{Cu K}\alpha$ radiation. Differential scanning calorimetry (DSC) measurements were carried out under a purified argon atmosphere in a Perkin-Elmer DSC-7. A BMG rod 5 mm wide was cut to a length of about 10 mm and its ends were polished flat and parallel. The acoustic velocities and the pressure dependences of the elastic constants of the BMG were measured at room temperature by using the pulse–echo overlap method [15]. The travel time of ultrasonic waves propagating through the sample with a 10 MHz frequency was measured using a MATEC 6600 ultrasonic system with a measuring sensitivity of 0.5 ns. The accuracy for the acoustic velocities is about 0.5%. This system is capable of resolution of the velocity changes to 1 part in 10^5 and is particularly well suited to determination of pressure-induced changes in velocities. The high-pressure investigation was performed using a piston–cylinder high-pressure apparatus, and electric insulation oil was used as the pressure-transmitting medium (for which hydrostaticity had already been established). The temperature for the sample during the high-pressure acoustic testing was kept as room temperature. The measurements

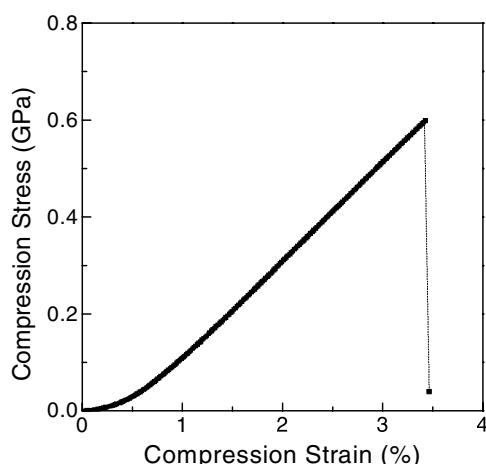


Figure 1. A uniaxial stress–strain curve in compression for $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG.

were performed for several pressure load–unload cycle times to examine the reproducibility. Upon pressure loading, the density and the length of the rod were modified with by Richard Cook method [16]. Density ρ was measured by the Archimedean technique and the accuracy is 0.1%. Elastic constants (e.g., Young’s modulus E , shear modulus G , bulk modulus K , and Poisson’s ratio σ) and the Debye temperature Θ_D of the BMG were derived from the acoustic velocities and the densities [11]. The compression test was conducted using INSTRON 5567, a testing machine (USA); the specimen dimensions were $\varnothing 3 \times 6 \text{ mm}^3$.

A mechanical test was conducted on the $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG. Figure 1 shows uniaxial compressive stress–strain curves typical for the BMG. The monolithic BMG material shows a linear elastic behaviour up to fracture stresses of 0.6 GPa. It fails, being brittle without any macroscopic plasticity, at fracture strains below 3.4% in compression. The stress–strain curve shows purely elastic behaviour before fracture. Figure 2 shows the pressure dependence of the reduced longitudinal and transverse velocities at the first pressure load and unload cycle, $\Delta v(P)/v(P_0) = (v(P) - v(P_0))/v(P_0)$, for the $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG at room temperature, where P_0 is the ambient pressure. Neither observable permanent changes in sound velocities with pressure (P) up to 0.5 GPa nor density increase in the sample was found within experimental limits after the testing. These results indicate that the measurements are within the elastic region of the BMG. Upon pressure loading, a normal behaviour of positive pressure dependence of the ultrasonic velocities was observed for this glass; v_l and v_s increase roughly linearly, and the relative changes of v_l and v_s are ~ 1.32 and 0.75% , respectively. Therefore, the longitudinal velocity is slightly more sensitive to the pressure variation than the transverse one. Similar P -dependences of the sound velocities are also found for other BMGs [6, 7].

It should be noted that, in the first pressure load run, the v_s exhibits a small kink at about 0.2 GPa; the slope of the relative variation for the transverse wave velocity is found to change from 1.17 to 1.70. The kink disappears in the subsequent pressure load and unload cycles. This phenomenon was also observed for other MGs, and low-temperature annealing could remove the sharp break and extend the behaviour in the annealed state [17]. The phase transition can induce an abrupt K -change in a MG [18]; however, no phase transitions have been observed in the BMG in the applied pressure range by means of XRD and TEM. This would suggest a pressure-induced short-range rearrangement; i.e. the structural relaxation results in an abrupt

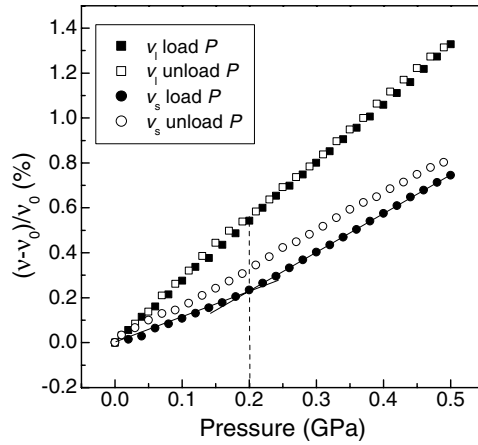


Figure 2. Variations of the longitudinal velocity, v_l , and the transverse velocity, v_s , of the $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG with pressure P . v_0 is the velocity at ambient pressure P_0 .

change at 0.2 GPa. The pressure, like the temperature, can induce structural relaxation which has been observed in a lot of MGs [19, 20]. From figure 2 it can be clearly seen that the transverse velocity v_s shows a obvious elastic retardation during pressure unloading, while the longitudinal velocity does not exhibit any retardation; this result may suggest that v_s is more sensitive to microstructure change, and that the small kink results from the structural relaxation induced by pressure.

Cook's method [16], by which the elastic constants and sample dimensions can be calculated simultaneously and self-consistently, was used for dimensional or density correction with compression. Figure 3 shows the relative variations of $(Y - Y_0)/Y_0$ with pressure, where Y_0 is the normal modulus at P_0 . As shown in figure 3, $\partial K/\partial p$, $\partial E/\partial p$, and $\partial G/\partial p$ for the BMG are positive, and the values are 3.42, 2.31, and 0.79, respectively. The elastic constants increase with pressure, indicating the increase in atomic force of the BMG under higher hydrostatic pressure; the increase can be attributed to the denser packing of the BMG [5]. It can be seen from figure 3 that a small kink at about 0.2 GPa is also found in the variation of elastic constants upon application of pressure.

Considering the BMG as a monatomic lattice with an average cellular volume, the Debye temperature Θ_D can be calculated from the acoustic velocities using the following equation:

$$\Theta_D = \frac{h}{k_B} \left(\frac{9}{4\pi\Omega_0} \right)^{1/3} \left(\frac{1}{v_l^3} + \frac{2}{v_s^3} \right)^{-1/3}, \quad (1)$$

where h and k_B are the Planck constant and the Boltzmann constant, respectively. Ω_0 is the atomic volume. As shown in figure 4, Θ_D increases almost linearly with the pressure up to 0.5 GPa, indicating an increase in the rigidity of the BMG with P [21].

On the basis of the bulk modulus and its pressure dependence, an isothermal equation of state (EOS) can be established in the Murnaghan form [22]:

$$P = \frac{K_0}{K'_0} \left[\left(\frac{V_0}{V(P)} \right)^{K'_0} - 1 \right], \quad (2)$$

where K_0 and K'_0 are the bulk modulus and its pressure derivative at zero pressure, respectively, and V_0 is the volume at zero pressure. From figure 3, K_0 and K'_0 are derived as 46.5 and

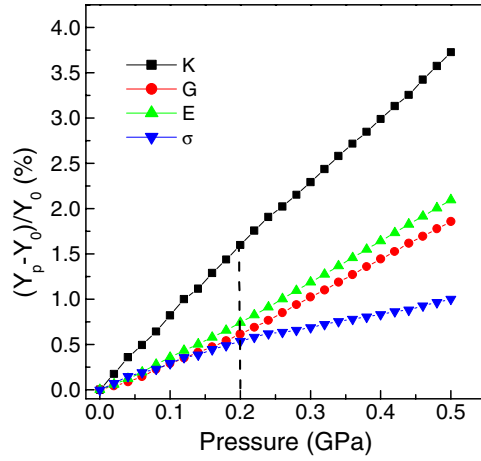


Figure 3. Variations of elastic constants Y ($Y = E, G, K, \sigma$) of $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG with pressure P . Y is normalized using $(Y - Y_0)/Y_0$ where Y_0 is the normal modulus at ambient P_0 .

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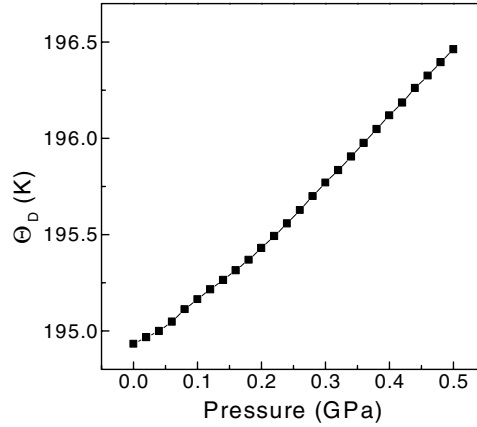


Figure 4. The pressure dependence of Debye temperature for $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG.

3.42 GPa, respectively. Accordingly, the isothermal EOS of the BMG in the elastic region is described as

$$P = 13.61 \left[\left(\frac{V_0}{V(P)} \right)^{3.42} - 1 \right]. \quad (3)$$

The volume compressibility of metallic elements can be expressed as [23]

$$\Delta V/V_0 = -aP + bP^2, \quad (4)$$

where a and b are coefficients, $\Delta V = V(P) - V_0$ is the volume compression. Using equation (4) and available data for Al, Fe, Co, and Nd [23, 24], the compression curves of Nd, Al, Fe, and Co as well as the EOS of the BMG given by equation (3) were obtained; these are plotted in figure 5. For metallic glass, it is known that the persistence of the metallic bonding and short-range order can be described by a dense-random-close-packing (DRP) model [1]. However, for NdAlFeCo BMG, whose constituents are all metallic elements, has a highly ordered microstructure [13, 14]. Since the compressibility of a solid is determined by the

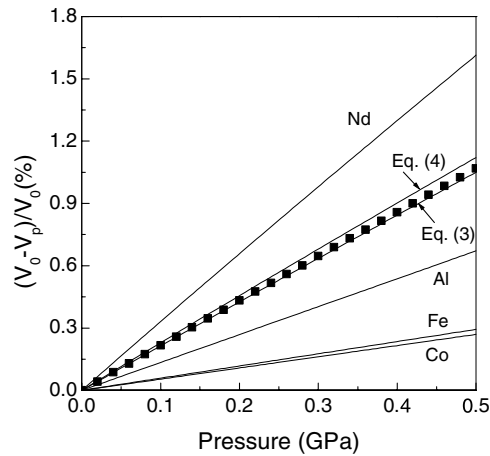


Figure 5. A comparison of the volume compression curves for the $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG calculated using equations (3) and (4); the experimental results are denoted by the symbols ■. The figure also gives the compression curves of the constituent elements.

nature of the interatomic potential and the atomic configurations [23], it seems reasonable that the volume compression of the BMG is calculated as a mean of the values for all elements based on the atomic percentages of the constituent elements. The total compression of BMG is ascribed to the attributes of individual metallic elements; thus equation (4) can be used to calculate the volume compression of the BMG. The calculated result is also shown in figure 5. It is seen from figure 5 that the compression curve for the $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG lies among those of its metallic components Nd, Al, Fe, and Co, indicating that the compression of the BMG depends on its metallic components, and is an average of those of its elements; it is very likely that the same atomic close-packed configurations dominate the short-range structure of the $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ BMG. The values calculated using equation (4) are consistent with the EOS of equation (3) derived from the Murnaghan form and experimental data.

According to the relationship between compressibility of a solid and its moduli, the elastic constants E , G , and K as well as σ can also be calculated in the form

$$M^{-1} = \sum f_i M_i^{-1}, \quad (5)$$

where M denotes any elastic constant and f_i is the atomic percentage of the constituent element. And the Debye temperature can be estimated from [25]

$$\Theta_D^{-2} = \sum f_i \Theta_i^{-2}, \quad (6)$$

where Θ_i is the Debye temperature of the constituent element. The values of the elastic constants and Debye temperature of the BMG at ambient conditions calculated using equations (5) and (6) are listed in table 1. For comparison, the density, longitudinal and transverse sound velocities, elastic constants, and Debye temperature for the Nd-based BMG and available data for the metallic components of the BMG are given in table 1. The elastic constants and Debye temperature of the BMG calculated using equations (5) and (6) are in good agreement with those obtained by using the ultrasonic pulse–echo method. This further confirms that the BMG can be considered as exhibiting dense packing ascribable to its short-range order.

In conclusion, the elastic constants and Debye temperature of the bulk $\text{Nd}_{60}\text{Al}_{10}\text{Fe}_{20}\text{Co}_{10}$ metallic glass as well as their pressure dependence, and the equation of states of the BMG, have been determined by ultrasonic measurements. It is found that the volume compression and

Table 1. Elastic properties of Nd₆₀Al₁₀Fe₂₀Co₁₀ BMG and its constituent metals at ambient conditions.

Materials		ρ (g cm ³)	v_l (km s ⁻¹)	v_s (km s ⁻¹)	E (GPa)	G (GPa)	K (GPa)	σ	Θ_D (K)
Nd ^a		7.007	2.718	1.438	37.9	14.5	32.4	0.31	147.5
Al ^b		2.699	6.794	3.235	77.4	28.6	88	0.35	426.6
Fe ^b		7.874	6.064	3.325	223.2	86.9	173.1	0.28	479
Co ^b		8.9	5.827	3.049	215.6	82.2	190.4	0.31	453
Nd ₆₀ Al ₁₀ Fe ₂₀ Co ₁₀	Experiment	7.0	3.242	1.714	54.1	20.7	46.5	0.31	194.9
	Calculated	6.929			54.1	20.7	46.9	0.31	184.2

^a Reference [26].^b Reference [27].

elastic constants of the BMG lie among those for the metallic components, being averages of those for the elements, indicating that the same atomic close-packed configurations dominate the elastic behaviour of the metallic glass. The calculated results for the volume compression and elastic constants based on short-range order are in agreement with experimental data. Our results further confirm that the BMG configuration has a close correlation with the atomic configurations of the metallic components and can be considered as having highly densely packed microstructure.

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