



Elasticity, thermal expansion and compressive behavior of $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ bulk metallic glass

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ABSTRACT

The existence of special covalently bonded short-range ordering structures in a $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ bulk metallic glass (BMG) is confirmed by thermal expansion and compression behavior. Under ambient conditions the linear thermal expansion coefficient obtained is almost constant in the glassy state with a value of $4.0 \times 10^{-5} \text{ K}^{-1}$. By fitting the static equation of state at room temperature under ambient conditions we find the value for bulk modulus B of 48.7 GPa, which is in excellent agreement with the experimental study by pulse-echo techniques of 44.7 GPa. Unlike many bulk metallic glasses, such as Zr- and Pd-based, which bulk modulus is much larger than 100 GPa, the value B of $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG falls into the range of SiO_2 and fluorozirconate glass ZBLAN. Moreover, the elastic constant of the $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG is almost the same as those of ZBLAN. No evidence for the high-pressure phase transitions of the $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG has been found up to 31.19 GPa at room temperature.

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1. Introduction

Mg-based bulk metallic glasses (BMGs) with 4–7 mm in diameter prepared by copper mold casting under vacuum or argon conditions have superior specific strength, plasticity and good corrosion resistance [1–4]. Recently, a new highly processable $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ alloy with high glass-forming ability (GFA), high ignition and oxygen resistances were successfully fabricated in air atmosphere [5,6]. Bulk metallic glasses containing rare earth ions or nano-regions of ferroelectric crystals have received much interest, because such materials have potential for laser host, tunable waveguide and tunable fiber grating. The mechanical and/or elastic properties of materials are always influenced by the change of environment, such as temperature and pressure. Since the strength of materials increases with their elastic module, it is therefore possible to assess strength indirectly from their elastic property [7]. In an amorphous solid, such as glass, the elastic strain produced by a small stress can be described by two independent elastic constants, c_{11} and c_{44} . The Cauchy relation $c_{12} = c_{11} - 2c_{44}$

allows one to determine c_{12} . For pure longitudinal waves $c_{11} = \rho V_L^2$, and for pure transverse waves $c_{44} = \rho V_T^2 = G$, where V_L and V_T are the longitudinal and transverse velocities, respectively. The sound velocities also allow the determination of Young's modulus, E , bulk modulus, B , and Poisson's ratio, σ , by the following equations [8]:

$$E = \rho V_T^2 \frac{3V_L^2 - 4V_T^2}{V_L^2 - V_T^2}, \quad (1)$$

$$B = \rho \frac{3V_L^2 - 4V_T^2}{3}, \quad (2)$$

$$\sigma = \frac{V_L^2 - 2V_T^2}{2(V_L^2 - V_T^2)}. \quad (3)$$

Elastic properties of glasses show a very interesting correlation with the glass structure, the knowledge of elastic characteristics would allow the analysis of the thermal expansion, specific heat and would provide considerable information about the structure of non-crystalline solids since they are directly related to the inter-atomic forces and potentials [9]. In the meanwhile, the measurements of compression properties (true compressibility) are of great importance to understand the relationship between unique

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mechanical properties and configurational changes under pressure [10]. Moreover, according to the relationship between isothermal compressibility χ_T (theory compressibility) and elastic constants: $\chi_T = 3/(c_{11} + 2c_{12})$ [11], we can compare the true compressibility with theory one to confirm the structure of Mg-based BMG.

In this work, ultrasonic velocities and several physical properties, including density and thermal expansion of a $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ ternary BMG were measured. The experimental results are used to obtain elastic constants. Information about the structure of glasses can be deduced from the calculation of the elastic moduli, Debye temperature and Poisson's ratio. The equation of state (EOS) of the BMG has been obtained. From these studies we can extract valuable information about the structure of the prepared glasses.

2. Experimental

The preparation of a $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG can be found in Ref. [5]. The amorphous nature as well as the homogeneity of the BMG was ascertained by X-ray diffraction, transmission electron microscopy and differential scanning calorimetry (DSC). 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 accurately adjustable gear-worm-level system. The culet of the diamond anvil is 500 μm in diameter. The amorphous powder sample together with the pressure-calibrator ruby was loaded into a 200 μm -diam hole of a T301 stainless steel gasket, which was preindented to a thickness of about 61 μm . Silicone oil was used as the pressure-transmitting media. The in situ angle dispersive X-ray diffraction (ADXRD) experiment under high pressure was performed at 4W2 High-Pressure Station of Beijing Synchrotron Radiation Facility (BSRF). The Debye rings were recorded using an image plate in transmission mode, and the XRD patterns were integrated from the images using the FIT2d software [12]. The size of X-ray spot was $130 \times 130 \mu\text{m}^2$. The experimental pressure was determined from the position of diffraction peak of ruby. More experimental details can be found in Ref. [13].

Room temperature ultrasonic measurements were performed by the pulse-echo method using a flaw detector (USM3-Krautkrämer). X-cut transducers were employed for longitudinal modes and Y-cut for transverse modes. The pulse transiting time was measured using a Hewlett-Packard model 54502A oscilloscope. The velocity was therefore obtained by dividing the round trip distance by the elapsed time. Ultrasonic travel time was measured at a frequency of 10 MHz and at room temperature. Several effects such as multiple internal reflections within the transducer, sample thickness, and the acoustic impedance mismatch between the glass sample and the transducer influence the accuracy of ultrasonic velocity measurements. The uncertainty is estimated to be about $\pm 1\%$.

The linear thermal expansion coefficient (α) of the metallic glasses was measured using a thermo-mechanical analyzer (DIL402C, NETZSCH Instrument Co., Ltd.) at a heating rate of 0.083 K^{-1} . The load applied on the specimen was 3 g force, corresponding to a compressive stress of 0.002 MPa.

The density was determined by an Archimedes technique. The accuracy of the measurement was about $\pm 0.001 \text{ g/cm}^3$.

3. Results

3.1. Compressive behavior

The stability of MgCuTb BMG under high pressure was investigated by using ADXRD. Fig. 1(a) shows selected synchrotron radiation X-ray diffraction spectrum under different pressures in $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG. As can be seen, with increase of pressure the

broad diffusive amorphous halo obviously shift to the higher angle, which shows the compression behavior of bulk amorphous alloys. No new diffraction peaks were detected from the curves at the range from 0.7 to 31.19 GPa, which means that the structure of the bulk metallic glass is quite stable at room temperature. The result can also be confirmed in the collected Debye rings on the image plate for $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG at pressures of 0.7 GPa, 14.33 GPa and 31.19 GPa shown in Fig. 1(b). As shown in Fig. 1(b), only a broadened diffraction ring corresponding to the amorphous nature can be detected among the whole pressure experiment.

3.2. Obtaining the equation of state and bulk modulus of $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ bulk metallic glass

In order to have a better understanding its behavior at high pressures for $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG, it is necessary to obtain the equation of state. Bridgman presented the EOS as follow [14]:

$$-\Delta V/V_0 = a_0 + aP + bP^2 + cP^3 + \dots, \quad (4)$$

where V_0 is the volume at zero pressure, coefficients a_0 , a , b , and c can be determined by using the least squares method. One can estimate the relative volume change $\Delta V/V_0$ ($\Delta V = V_P - V_0$) at a given pressure (V_P) to that at zero pressure (V_0). The experimental $\Delta V/V_0 - P$ data were shown in Fig. 2. When fitted by the Bridgman equation, EOS can be expressed as

$$-\Delta V/V_0 = -0.01162 + 0.02052P + 8.425 \times 10^{-4}P^2 + 1.425 \times 10^{-5}P^3. \quad (5)$$

The bulk modulus B can be obtained according to the relationship, $B = \frac{1}{\alpha}$. The bulk modulus B of the BMG is 48.7 GPa, which less than half of Zr- and Pd-based BMGs' [10,15]. The smaller bulk modulus indicates that the material is relatively softer. Therefore, it is easy to understand the larger compressibility about 21%. The result indicates that the BMG has much looser atomic configuration compared with Zr- and Pd-based BMGs [10,15], which B -value is much larger than 100 GPa and compressibility is around 16%.

3.3. Thermal expansion

The thermal expansion is due to the anharmonicity of the atomic potentials. Therefore, investigation of thermal expansion in a multi-component metallic glass is of great interest and importance for understanding chemical/configurational structure further. Typical linear thermal expansion $\Delta L/L$ data versus temperature for the $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG is shown in Fig. 3, inset is the thermal expansion coefficient (α) of $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG according to data $\Delta L/L$. It is clear that the linear thermal expansion increased with increasing temperature before crystallization temperature (487 K). If the thermal expansion coefficient (α) of $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG is considered from 350 to 487 K, we will have the average thermal expansion coefficient of $4.4 \times 10^{-5} \text{ K}^{-1}$, just similar to that of oxyfluorovanadate glasses $2.5 \times 10^{-5} \text{ K}^{-1}$ [16], but is much higher than that of SiO_2 glass $5.4 \times 10^{-7} \text{ K}^{-1}$ (140–1000 K) [17].

3.4. Elastic constant

Elastic constant measurements are of central importance in investigations of the vibrational properties of a metallic glass, and second-order elastic constants (SOECs) give the slope of dispersion curves at long wavelength limit [18], therefore, density, longitudinal and transverse sound velocities, the calculated SOECs (c_{11} and c_{44}), Young's modulus, bulk modulus and the Poisson's ratio for $\text{Mg}_{65}\text{Cu}_{25}\text{Tb}_{10}$ BMG, is given in Table 1. For comparison, other three glasses $\text{Pd}_{39}\text{Ni}_{10}\text{Cu}_{30}\text{P}_{21}$ BMG, 53ZrF4–20BaF2–4LaF3–3AlF3–20NaF (ZBLAN) [19] and a pure SiO_2 glass are also listed

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