Journal of Non-Crystalline Solids 406 (2014) 127–132

Contents lists available at ScienceDirect

Journal of Non-Crystalline Solids

journal homepage: www.elsevier.com/locate/jnoncrysol

Study of the mechanism of the internal friction peak in a $Cu_{36}Zr_{48}Al_8Ag_8$ bulk metallic glass

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ARTICLE INFO

Article history:
Received 29 July 2014
Received in revised form 27 September 2014
Accepted 30 September 2014
Available online 11 October 2014

Keywords:
Bulk metallic glasses;
Internal friction;
Peak;
Frequency;
Relaxation behavior

ABSTRACT

The nature of internal friction (IF) peak in the internal friction versus temperature curve (Q^{-1}–T curve) remains a controversial problem in the field of bulk metallic glasses (BMGs). In order to study the mechanism of the internal friction peak, the evolutions of internal friction at fourteen frequencies and electrical resistivity with temperature were simultaneously measured in a $Cu_{36}Zr_{48}Al_8Ag_8$ BMG. The frequency dependence of internal friction (Q^{-1}–f curves) was acquired from the Q^{-1}–T curves by the linear interpolation method. The results show that there are two different mechanisms for the internal friction peaks in the Q^{-1}–T curves. The high-frequency internal friction peak is a first-order phase transformation peak and appears at the crystallization process. However, the low-frequency internal friction peak is a relaxation peak, which is also observed in the Q^{-1}–f curve at the corresponding temperature, and the relaxation process is controlled by a cooperative hopping movement of atoms.

1. Introduction

Internal friction (IF), which is defined as Q^{-1} and expressed as $Q^{-1} = \tan \phi$ (ϕ is the angle by which strain lags behind stress), is very sensitive and effective in detecting atomic rearrangements and the kinetics of atomic movements involved in microstructural evolution [1–3]. For amorphous alloys, IF has been extensively used to study the structural relaxation, glass transition and crystallization behaviors [4–10]. One of the important IF characteristics is that there usually appears a peak in the IF versus temperature curve (Q^{-1}–T curve) when an amorphous sample is heated to a higher temperature than its onset crystallization temperature. However, the origin of the peak is still a controversial problem. Some investigators proposed that the peak originates from both the glass transition and crystallization [5], while some researchers inferred that the peak temperature corresponds to that of a phase transition which occurs the fastest [11], but He et al. [12,1] suggested that the position of peak is in accordance with the onset temperature of phase transformation rather than the point of the fastest phase transformation rate.

Given the aforementioned inconsistent viewpoints, it is essential to unravel the nature of the IF peak in the Q^{-1}–T curve in metallic glasses. In this work, the temperature dependence of IF was measured at fourteen frequencies in a Cu–Zr–Al–Ag BMG. Based on the Q^{-1}–T curves, the frequency dependence of IF (Q^{-1}–f curves) was obtained by the linear interpolation method. Electrical resistivity, as a sensitive probe of structure rearrangements [13], was widely used to investigate the structural transformations of amorphous alloys. In order to acquire the information of structural transformations of the Cu–Zr–Al–Ag BMG during the IF measurement process as accurately as possible, the temperature dependence of electrical resistivity was simultaneously measured. The experimental results show that there exist two different mechanisms for the low-frequency and high-frequency IF peaks in the Q^{-1}–T curves.

2. Experimental procedures

An ingot with a nominal composition of $Cu_{36}Zr_{48}Al_8Ag_8$ (at.%) was prepared by arc melting a mixture of Cu, Zr, Ag, and Al metals with a purity of over 99.5 wt.% in a Ti-gettered high purity argon atmosphere. The ingot was remelted four times to ensure a chemical homogeneity followed by sucking the melt into a copper mold with an internal cavity of $0.002 \times 0.01 \times 0.08$ m^3. The amorphous nature of the as-cast sample was verified by X-ray diffractometry on a D/MAX-2500V diffractometer using Cu K_α radiation. The thermal behavior of the as-cast amorphous alloy was examined by differential scanning calorimetry (DSC) on a Perkin-Elmer DSC-8000 at a heating rate of 0.0167 K/s. The DSC system was calibrated for temperature and enthalpy by using Zn and In standards, giving an accuracy of $\pm$0.2 K and $\pm 2 \times 10^{-5}$ mW, respectively. Rectangular sheet samples with dimensions of $0.002 \times 0.0015 \times 0.06$ m^3 were cut from the as-cast amorphous alloy by spark machining for the IF and electrical resistivity measurements. The studied amorphous alloy is well known due to its exceptional glass forming ability and thermal stability [14].

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http://dx.doi.org/10.1016/j.jnoncrysol.2014.09.053
0022-3093/

The temperature dependences of IF Q^{-1} and relative modulus M were performed using a conventional inverted torsion pendulum apparatus by a forced vibration mode from room temperature to 873 K at a heating rate of 0.0167 K/s in low vacuum with a resolution in Q^{-1} better than 10^{-4} and a resolution in M lower than 10^{-3}. Fourteen frequencies of 0.1, 0.2, 0.3, 0.4, 0.6, 0.8, 1.0, 1.2, 1.7, 2.5, 3.5, 5, 7, and 10.0 Hz were employed with a strain amplitude of 2.0×10^{-5}. Some modifications were made on the IF apparatus for the electrical resistivity measurement and four molybdenum probes were inserted to enable current flow and detect the voltage. The electrical resistivity specimen was placed next to the IF specimen. Accompanying the IF measurement, the electrical resistivity of the $Cu_{36}Zr_{48}Al_8Ag_8$ BMG as a function of temperature was measured by the direct current four-probe method, of which more details can be found in Ref. [15]. The precision of the electrical resistivity measurement is $\pm 1 \times 10^{-9}$ Ωm. The temperature of the samples was measured using a chromel–alumel thermocouple with a precision of ±0.5 K. To warrant the reproducibility of experimental data, the experiments of internal friction, electrical resistivity and DSC were conducted three times. The average values would be available. The random errors are the differences between measured values and mean values. Based on the Q^{-1}–T curves, the Q^{-1}–f curves were obtained by the linear interpolation method.

3. Results

Fig. 1 shows the XRD pattern of the as-cast $Cu_{36}Zr_{48}Al_8Ag_8$ sample. The broad diffraction peak is a typical characteristic of an amorphous structure.

Fig. 2 displays the DSC and normalized electrical resistivity ρ/ρ_0 curves for the $Cu_{36}Zr_{48}Al_8Ag_8$ BMG at a heating rate of 0.0167 K/s, where ρ and ρ_0 represent the electrical resistivities at temperature T and room temperature, respectively. Unfortunately, it is difficult to determine the glass transition temperature T_g from the DSC curve because the endothermic step peculiar to the glass transition cannot be obviously observed at the low scanning rate. However, in the ρ/ρ_0–T curve one can see a distinct change in the temperature coefficient of resistivity at 652.3 K, which is attributed to the glass transition, and a sudden drop of resistivity at 736.2 K, which is considered to correspond to the onset crystallization temperature [16–18]. The crystallization temperature T_x obtained from the DSC thermogram is in good agreement with that from the electrical resistivity curve.

Fig. 3 demonstrates the temperature dependences of Q^{-1} (Q^{-1}–T curves) and M (M–T curves) for the $Cu_{36}Zr_{48}Al_8Ag_8$ BMG with fourteen frequencies from room temperature to 873 K at a heating rate of 0.0167 K/s. For each frequency, at low temperature Q^{-1} is very low. With elevating temperature Q^{-1} gradually increases. Q^{-1} and M change in opposite trends. Above a certain temperature Q^{-1} increases quickly and M decreases rapidly. No peak is observed at or near the glass

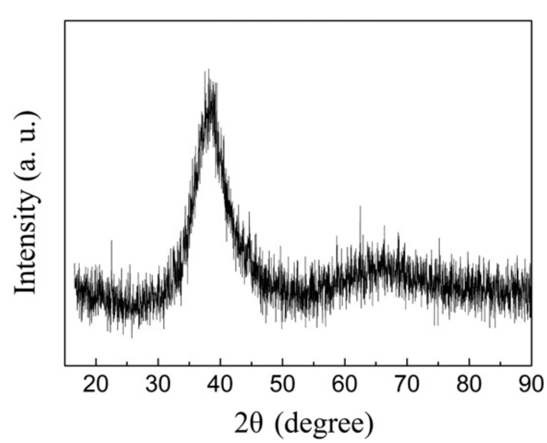

Fig. 1. XRD pattern of the as-cast $Cu_{36}Zr_{48}Al_8Ag_8$ sample.

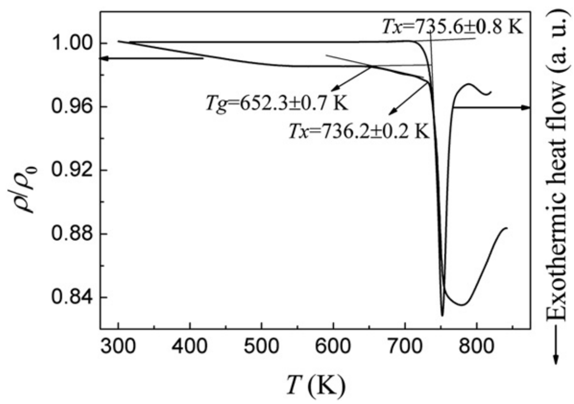

Fig. 2. DSC and normalized electrical resistivity ρ/ρ_0 curves for the $Cu_xZr_{84-x}Al_8Ag_8$ BMG at a heating rate of 0.0167 K/s.

transition temperature T_g in the Q^{-1}–T curves. Subsequently, an internal friction peak corresponds approximately to a relative modulus dip. The temperature of modulus dip has a little hysteresis behind that of internal friction peak, which was also observed in Zr–Cu–Al–Ni–Nb and La–Ce–Al–Co amorphous systems and may reflect the difference in the structure sensitivity of between Q^{-1} and M [5,19]. The essence of IF peaks was the focus of this study and would be discussed as follows.

The IF peak temperature T_p as a function of frequency is displayed in Fig. 4. It is surprising that T_p shows a crossover with increasing frequency. At the low frequency side (from 0.1 to 0.8 Hz), the peak shifts to a higher temperature (from 718 to 737 K) with increasing the measured frequency, which is one of noteworthy characteristics for a relaxation IF peak, implying that the low-frequency IF peak may be related to a thermally activated relaxation process. Above a certain frequency (0.8 Hz), the position of peak is almost independent of the frequency, which is one of striking features for a first-order phase transformation (FOPT) IF peak, indicating the high-frequency IF peak is closely associated with FOPT. It should be pointed out that the high-frequency IF peak temperature is slightly larger than the onset crystallization temperature T_x.

Fig. 5 exhibits the relationship between the magnitude of IF peak Q_p^{-1} and the reciprocal of frequency $1/f$ for the $Cu_{36}Zr_{48}Al_8Ag_8$ BMG. Fig. 5(a) reveals that there is a linear relationship between Q_p^{-1} and $1/f$ at the high frequency side. But the linear relation of Q_p^{-1} and $1/f$ does not exist at the low frequency side, as shown in Fig. 5(b). Delorme and Gobin [20] proposed that a linear relation of Q_p^{-1} and $1/f$ exists for this kind of IF peaks during FOPT. The result in Fig. 5(a) is in agreement

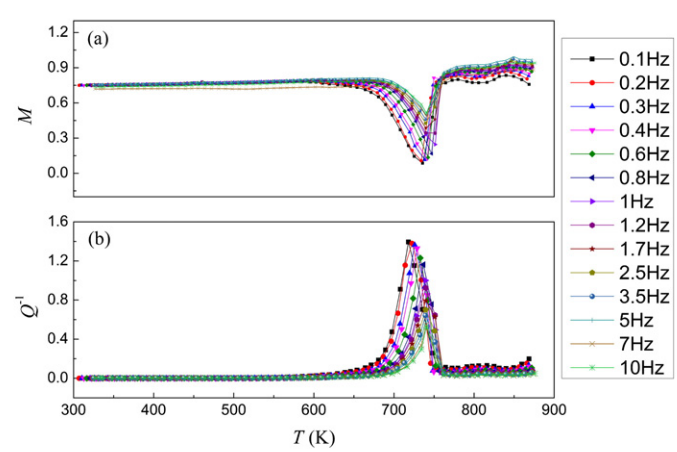

Fig. 3. Temperature dependences of (a) M and (b) Q^{-1} for the $Cu_{36}Zr_{48}Al_8Ag_8$ BMG with fourteen frequencies at a heating rate of 0.0167 K/s. The solid lines are guides to the eyes.