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Deformation behavior and dilatometric measurements of Nd–Fe based bulk metallic glass

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Abstract

Mechanical deformation and thermal expansion behavior of $Nd_{60}Fe_{20}Co_{10}Al_{10}$ bulk metallic glass (BMG) were studied. A stress overshoot followed by a viscous flow approaching a steady state can be seen in the stress–strain curves within a wide temperature range. Moreover, the flow behavior of the BMG is strongly dependent on the strain rate. The dilatometric measurements of the alloys during continuous heating suggest that the glass transition begins above 470 K, which is accompanied by several weak crystallization processes. The rather unique thermal stability of the Nd–Fe based BMGs is responsible for the difference in homogeneous deformation behavior between the present BMG and other BMGs.

Keywords: Bulk metallic glass; Mechanical properties; Hardness; Glass transition

1. Introduction

In contrast to binary and ternary amorphous alloys produced in the form of ribbons, bulk metallic glasses (BMGs) are characterized by a strong glass forming ability together with a wide supercooled liquid region defined by the temperature range between glass transition temperature (T_g) and crystallization temperature (T_x) [1–3]. The homogeneous deformation of BMGs, especially in the supercooled liquid region, has attracted a growing interest due to the fact that BMGs have excellent formability in this region, promising applications in the field of near-net shape fabrication [4,5]. It is reported that the deformation mode is strongly dependent on the alloy system, as well as testing temperature and strain rate [6–10].

Recently, bulk metallic glasses (BMGs) obtained in multicomponent (Nd, Pr)–Fe based systems with hard magnetic properties at room temperature have been reported [11–20]. These systems do not exhibit a distinct glass transition prior to crystallization according to constant-rate heating DSC measurements, indicating a low thermal stability of the supercooled liquid. However, an extremely high value of the ratio between the T_x and T_l (liquidus temperature) around 0.9 was observed for such systems, which indicates a rather high stability of the amorphous phase against crystallization. The contradiction between the absence of a marked glass transition and the extremely high value of T_x/T_l shows that these systems are rather unique within the different families of BMGs.

In this study, the deformation and thermal expansion behaviors of the $Nd_{60}Fe_{20}Co_{10}Al_{10}$ BMG have been systematically investigated. The results will be helpful to clarify the thermal stability of the Nd–Fe based BMGs.

2. Experimental procedure

Pre-alloyed $Nd_{60}Fe_{20}Co_{10}Al_{10}$ ingots were prepared by arc-melting a mixture of Nd, Fe, Co, and Al elements with a purity of at least 99.9% in titanium-gettered argon atmosphere. Cylindrical specimens of 3 mm in diameter and 70 mm in length were prepared from the pre-alloyed ingots by suction casting into a copper mold. The structure of the cast cylinders was characterized by X-ray diffraction (XRD) in a Philips PW 1050 diffractometer using Co Kα radiation. Thermal analysis was performed with a Perkin–Elmer DSC 7 differential scanning calorimeter under argon atmosphere. A constant heating rate of 0.17 K/s was employed.

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The thermal expansion was measured on cylinder samples with 3 mm in diameter under a compressive load of 0.3 N. The experiments were performed using a NETZSCH DIL 402C dilatometer with a resolution of $\Delta L = 1.25$ nm. A low heating rate of 0.017 K/s was used to ensure the temperature homogeneity in the sample during heating. The compression tests on cylindrical samples of 3 mm in diameter and 5.8 mm in length were performed in an Instron-type testing machine at various temperatures from room temperature up to 723 K. The cross-head was moved at a constant speed with an initial strain rate of $1.4 \times 10^{-3}\,s^{-1}$.

3. Results and discussion

The as-cast cylinder exhibits an XRD spectrum typical for amorphous phase without obvious crystalline reflection peaks. The temperature dependence of the stress–strain curves at an initial strain rate of $1.4 \times 10^{-3}\,s^{-1}$ is shown in Fig. 1. The samples were equilibrated for about 300 s at each test temperature before testing. The alloy is extremely brittle at room temperature with a fracture stress of about 398 MPa and no plastic deformation. Premature brittle fracture also occurs at 473 K. The deformation mode changes from inhomogeneous deformation to homogeneous deformation around 493 K, and the samples do not fail at temperatures above 503 K. A stress overshoot followed by a viscous flow approaching a steady state can be seen in the curves measured in the temperature range from 503 to 623 K. In addition, the stable flow stress decreases continuously with increasing testing temperature. Fig. 2 exhibits the strain rate dependence of the stress–strain curves at 513 K. At a high strain rate of $1.4 \times 10^{-2}\,s^{-1}$, the peak stress drops suddenly and then the alloy fails at the strain of about 10%. The stress overshoot phenomenon is also observed at lower strain rates, but the level of stress drop after yielding decreases continuously with decreasing strain rate.

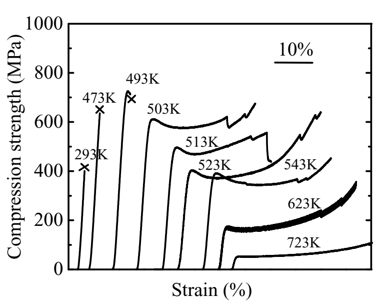

Fig. 1. The temperature dependence of the stress–strain curves for $Nd_{60}Fe_{20}Co_{10}Al_{10}$ BMG.

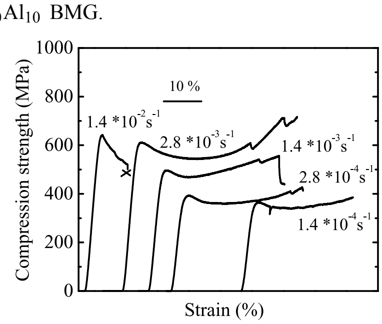

Fig. 2. The strain rate dependence of the stress–strain curves at 513 K for $Nd_{60}Fe_{20}Co_{10}Al_{10}$ BMG.

The stress overshoot phenomenon and a similar temperature dependence of the deformation behavior were also observed for Zr-, Pd-, Cu-, and La-based metallic glasses at temperatures near T_g [6–10]. Though the reasons for the presence of the stress overshoot in stress–strain curves are still a matter of debate in the literature, most authors tend to follow that the yield drop was a result of rapid increase in free volume during high strain rate deformation [6,7,10]. It is noteworthy that homogeneous deformation in metallic glass usually takes place at the temperature T_g, which is determined by constant-rate heating DSC measurements. However, for the present BMG no distinct glass transition can be seen in the DSC curve as shown in Fig. 3. Instead, the thermal flow deviates from the base line as early as about 470 K, and more obvious exothermic signal is observed above 580 K with a peak temperature of about 690 K. It is interesting to note that the homogeneous deformation together with the stress overshoot phenomenon is observed in the stress–strain curves in this temperature range (Fig. 1). Our previous studies on dynamic mechanical properties of this BMG exhibited a rapid decrease of modulus and a distinct internal friction peak starting at about 470 K, which suggests that a glass transition process takes place [17,19]. In the present compressive tests, $Nd_{60}Fe_{20}Co_{10}Al_{10}$ BMG exhibits typical plastic deformation behavior as that of other BMGs near the supercooled liquid region. This confirms that glass transition takes place at the temperature far below the major crystallization temperature of 770 K.

It should be pointed that the non-Newtonian behavior associated with stress overshoot of BMGs testing at a moderate strain rate usually takes place in a narrow temperature range of the order of tens Kelvin [6–10]. However, the present BMG exhibits a stress overshoot in very wide temperature range of more than 150 K at the strain rate of $1.4 \times 10^{-3}\,s^{-1}$ (Fig. 1). Furthermore, the level of stress drop is obviously

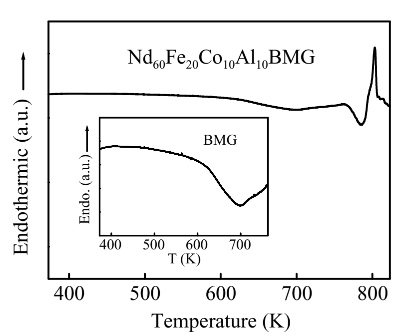

Fig. 3. DSC curve of the as-cast $Nd_{60}Fe_{20}Co_{10}Al_{10}$ BMG. The inset shows the enlarged part of the DSC scan revealing the exothermic heat flows in the low temperature region.

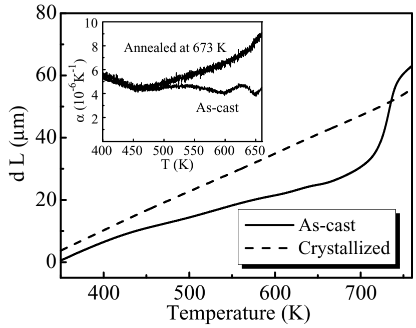

Fig. 4. Typical dilametric measurements of the as-cast and fully crystallized $Nd_{60}Fe_{20}Co_{10}Al_{10}$ BMG. The inset shows the temperature dependence of linear expansion coefficient of the as-cast sample and the sample annealed at 673 K.

lower than that reported in other BMG families at the comparative strain rate [6–10].

Nieh et al. has studied the structures of ZrAlTiCuNi BMG after the plastic deformation and pointed that the non-Newtonian behavior was associated with the concurrent crystallization of the amorphous structure during deformation [8]. The structural stability of Nd–Fe based BMG is still unclear, especially in the temperature range from 470 to 700 K. This obstructs the understanding of the observed flow behavior of the present BMG. Here, we studied the thermal expansion behavior of the alloy, as it is sensitive to the structural change of amorphous alloys. Fig. 4 shows the change of length of the as-cast $Nd_{60}Fe_{20}Co_{10}Al_{10}$ sample, ΔL, during heating at the heating rate of 0.017 K/s. The same measurement was also carried out for the fully crystallized sample, and the result is shown for comparison. During the continuous heating of the as-cast sample, several smooth decreases of the slope (representing the thermal expansion coefficient, α) are observed from 430 to 660 K. While, the length of the crystallized alloy increases linearly with increasing temperature. As supercooled liquid can access additional configuration states of lower density, metallic glass will exhibit an increase in α when it is heated to the temperature above T_g [21,22]. However, this expected increase of α is not observed near the brittle–ductile transition temperature (483 K as shown in Fig. 1). The temperature dependence of α of the sample annealed at 673 K is shown in the inset of Fig. 4. A gradually increase of the thermal expansion coefficient is observed above 470 K. This confirms that the majority of the alloy is already in a supercooled liquid state. Consequently, the increase of α in the curve of the as-cast sample is probably superposed by the decrease of α due to the relaxation and a few primary weak crystallization events. These processes also lead to the smooth exothermic flow in this temperature range as shown in the inset of Fig. 3. Earlier X-ray studies of the structures of the Nd–Fe based BMG also indicate that a very small fraction of crystalline phases precipitated after annealing at temperatures in this range [19]. The homogeneous deformation behavior and the gradual increase of α the temperature above 470 K suggest that the alloy is a mixture of supercooled liquid region and a small fraction of crystalline phases in the temperature interval between about 470 K and the eutectic crystallization temperature of the remaining matrix (770 K as shown in Fig. 3). These weak crystallization processes may partly contribute to the strong dependence of temperature and strain rate dependence of the plastic deformation behavior of the BMG, as crystallization is also a kinetic process, which is temperature and time dependent.

A relative high stress level (>300 MPa) is observed below 623 K in compressive tests (Fig. 1). This is thought to due to that the glass transition of the present BMG is accompanied by a series of primary crystallization processes, Further increasing the testing temperature, the stress decrease significantly due to the decrease of viscosity of the supercooled liquid phases. Previous studies proved that the annealing of the Nd–Fe based BMG at the temperature below 750 K for 3600 s did not decrease the hard magnetic properties obviously [12,19]. This indicates that this $Nd_{60}Fe_{20}Co_{10}Al_{10}$ BMG is easily to deformation in the temperature range from 623 to 750 K without sacrificing the hard magnetic properties.

4. Conclusions

In conclusion, the deformation behavior of $Nd_{60}Fe_{20}Co_{10}Al_{10}$ bulk metallic glass has been studied under uniaxial compression condition. The deformation mode changes from inhomogeneous deformation to homogeneous deformation around 493 K. A stress overshoot followed by a viscous flow approaching a steady state can be seen in the curves measured in the temperature range from 503 to 623 K. This mechanical behavior is similar to that of other BMGs deformed near supercooled liquid region. The results of thermal expansion measurements of as-case sample indicate that several weak primary crystallization processes take place at the temperatures far below the eutectic crystallization temperature of the remaining glassy matrix. This remaining matrix forms a large fraction of supercooled liquid during heating, indicated by the continuous increase of the thermal expansion exponent above 450 K in the sample annealed at 673 K. The presence of the supercooled liquid in the alloy sheds light on the good mechanical formability of the alloy at elevated temperatures.

Acknowledgements

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