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Stress effects on magnetic property of Fe-based metallic glasses

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ABSTRACT

The investigation on the stress relief of magnetic materials is important both for the industrial process and the academic research. In this work, the stress effects on the magnetic characteristics in Fe-based metallic glasses (MGs) are investigated under the applied bending stress and various annealing treatments. It was found that both the magnetic induction intensity $\boldsymbol{B}$ and effective permeability $\boldsymbol{\mu'}$ decrease while coercivity $\boldsymbol{H_c}$ increases with the applied compressive stress, indicating that the compressive effect is dominant on the magnetic characteristics in $Fe_{78}Si_9B_{13}$ MG ribbons. We also found that the wide domain wall movement and the rotation of the narrow domains are seriously affected by internal stress during the magnetization process. Our results reveal that the stress relief plays a critical role in improving magnetic properties of Fe-based MGs and are helpful for understanding the correlations between the stress, microstructure change and magnetic softness in magnetic MGs.

1. Introduction

Ferromagnetic metallic glasses (MGs) can be widely used in various electronic devices such as transformers, reactors, motors and mutual inductors. Ferromagnetic MGs with higher saturated magnetic flux density ($\boldsymbol{B_s}$), lower coercivity ($\boldsymbol{H_c}$), higher permeability ($\boldsymbol{\mu}$), better mechanical properties and lower materials cost [1–4], are promising candidates to replace traditional soft magnetic crystalline materials, and therefore attracted considerable research interests over past decades. In general, the amorphous alloys obtained by rapid quenching from the melt are often in a metastable nature. As a result, their microstructure and properties can easily be changed by subsequent heat treatment or mechanical deformation [5,6]. The magnetic MGs lack the magentocrystalline anisotropy as usually found in crystalline alloys. However, due to the internal stress developed along the ribbon axis during melt-quenching process and the interaction between elastic stress and spontaneous magnetization, their magnetic properties are also sensitive to the external stress and the thermal history [7,8]. Supplementary treatments were often introduced for removing the internal stress [9–11]. For example, it has been reported that desirable soft magnetic properties in MGs can be achieved by many stress-relieving techniques, such as conventional annealing below Curie temperature [5,6], flash annealing [9,12,13] and cryogenic thermal cycling (CTC) after conventional annealing [3].

However, even after annealing, there are still many factors that can cause the internal stress in the magnetic MG materials. As a result, extensive experimental and theoretical investigations have been dedicated to the relationship between the internal stress and magnetic parameters such as magnetic domain [14–16], magnetization process [17,18], magnetic saturation [19,20], coercively [21,22], effective permeability [20,21], etc. in magnetic MGs of various forms, like wires [19,23], thin films [24,25], ribbons [26,27] and bulk samples [28–31]. Despite of these efforts, there are still a lot of inconsistency in the experimental results and discrepancies in the theoretical analysis among these many kinds of magnetic MGs. Particularly, a quantitative understanding on the contribution of stress to the magnetization process and the influence of stress relief on the magnetic properties in MGs during the post-processing [32] are not yet clear.

In this work, we investigated the magnetization process of disk-shaped Fe-based samples. The effects of structural relaxation on internal intrinsic stress relief and the effect from subsequent external applied stresses on the magnetic properties are systemically studied. These results are helpful for designing stress-sensitive components.

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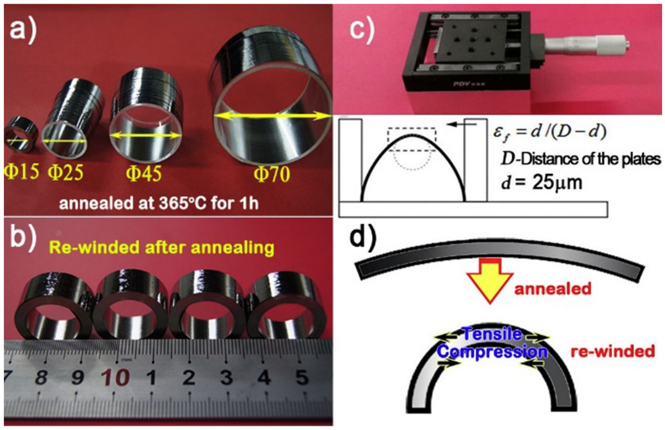

Fig. 1. (a) The mandrel-winded samples with the different radius of curvature after annealing under 365 °C for 1 h time, (b) the re-winded samples with the same internal diameter of 15 mm, (c) the relative strain at fracture (RSF) equipment and experimental method, (d) schematic diagram of magnified region of the ribbon indicating the stress state. During re-winding the bending process causes the external layered stresses-tensile near the surface on the free side and compressive underneath on the wheel side.

2. Experiments

The MG ribbons of $Fe_{78}Si_9B_{13}$ (supplied by An Tai Inc. of *China Iron & Steel Group*) with a uniform thickness of 25 μm and a width of 10 mm were prepared by the melt-spun technique. The amorphous nature of as-cast and annealed specimens was ascertained by X-ray diffraction (XRD, Bruker D8 Advance) with CuK_α radiation and differential scanning calorimetry (DSC, Perkin Elmer DSC8000). The anelastic structural relaxation behavior was explored using DSC [33–35] after annealing for 10 min, 1, 2, 3 h at $T = 365$ °C in a purified argon atmosphere at a heating rate of 20 K/min. The values of density ρ were obtained by the Archimedean technique.

For evaluation of subsequent stress effects after structural relaxation, the MG ribbons were mandrel-winded with various radius of curvature $r_{a,i}$ (*i*-indexed sample's radius) [initial internal diameter $\Phi_{\text{in}} = 2r_{a,i} = 15, 25, 45, 70$ mm, ∞(straight sample)] and then annealed at 365 °C for 1 h (Fig. 1a). All metallic glass ribbons for measurements of magnetic properties have the same length of 7 m. The $Fe_{78}Si_9B_{13}$ ribbons maintain the curvature after annealing because the stress is released completely during the annealing process [36]. In order to evaluate the embrittlement of the ribbons due to the annealing of winded samples on mandrels of different radii, the relative strain at fracture (RSF), which was represented by $\varepsilon_f = d/(D-d)$ (D is the distance between two parallel plate, d is the ribbon thickness) [37,38] was measured (Fig. 1c).

The samples were removed from the mandrel after annealing, then re-winded to a smaller curvature diameter of $\Phi_{\text{in}} = 2r_s = 15$ mm, as shown in Fig. 1b. After being curved into a smaller curvature, stresses were introduced again (tension in outer surface and compression in inner surface), as shown in Fig. 1d. The stress was greater for the cores with a larger initial curvature radius.

The magnetic induction and coercivity were measured using a *B-H* loop tracer (MATS 2010SD) under a maximum applied field of 800 A/m. The exciting and searching coils are 20 turns and 2 turns. The effective permeability at various frequency (0.1, 1, 10, 100 kHz) was measured by using an impedance analyzer (Agilent 4294A) equipped with a test fixture (16047E). The inductance of the samples was measured to characterize the magnetic effective permeability. When the ferromagnetic sample is magnetized by an AC excitation magnetic field, the magnetic effective permeability is expressed as a following form, $\mu' = L_s/L_0 = L_s 2\pi / \left(\mu_0 N^2 h \ln\left(\frac{or}{ir}\right)\right)$, where L_s & L_O are effective inductance (H) of the core with winding and inductance of the coil. N is the number of turns of the coils, h is the height of the toroidal core (mm). *or* & *ir* are the outer and the inner diameter of the toroidal core (mm), μ_0 is the permeability of the vacuum: $4\pi \times 10^{-7}$ [39–41]. To avoid accidental error, every measurement was performed over 3 samples and every data point was the average of 3 to 5 observations, especially 100 times for RSF measurement.

3. Results and discussions

The DSC results show that the value of T_c increases obviously with the annealing temperature and time, indicating that the isothermal annealing induces the structural relaxation due to the free volume reduction [33,42] and the annealing treatment affects the electron energy state of the MGs. Following the mean field theory, T_c depends on the exchange integral [43]. In the molecular field approximation, Curie temperature is expressed as

$$T_c = \frac{2S(S+1)}{3k} \sum_{ij} J_{ij} \tag{1}$$

where S is atomic spin, k is the Boltzmann constant, and J_{ij} represents the exchange interaction between atoms i and j [44,45]. It implies that isothermal annealing induces the change of the exchange integral.

The attempt to eliminate the pure external stress effect by making samples out of toroidal shapes with various radius of curvature was performed (Fig. 1). As shown in Fig. 2a, the RSF value decreases with the increase of the mandrel-winded radius, which can be well-fitted by a power law. This suggests that a plastic deformation have been occurred after annealing. After re-winding, the internal stress increases logarithmically according to the magnitude of the mandrel-winded radius (see the inset of Fig. 2b). During re-winding, the bending can cause external layered stresses that are tensile near the outer surface and compressive near the inner [46]. Fig. 2 (b) shows the variations of induced strain and the internal stress with the radius of the mandrel-

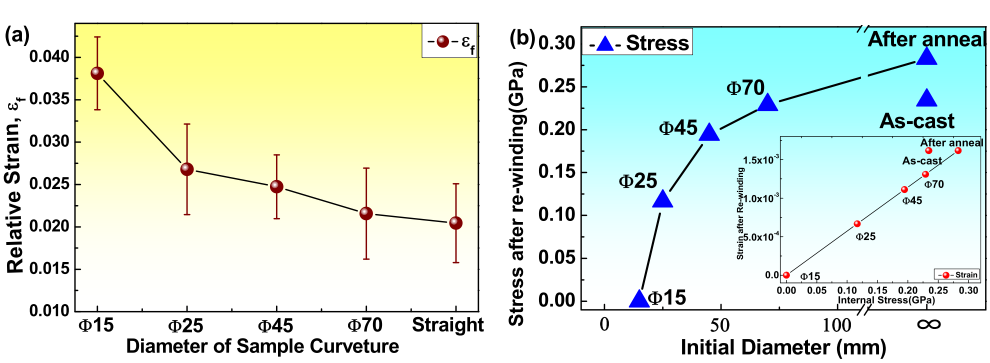

Fig. 2. (a) The relative strain at fracture on the bended samples with the different radius of curvature after annealing; (b) The relationship between partial internal stress and initial diameter which implies the logarithmic increase of stress to the initial diameter. The insert shows the induced partial strain of the samples after re-winding and partial internal stress in the minimum radius layer part of the mandrel-winded samples. Because of the different elastic moduli for the straight samples [48] the partial stress of as-cast samples is smaller than of the annealed samples. The values of the Young's moduli were referenced for as-cast ($E_{as}(H = 800A/m) \approx 145$ GPa) and annealed samples ($E_{t_{an}} = 350(H = 800A/m) \approx 175$ GPa) [47].