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Effect of heating rate on atom migration, phase structure and magnetic properties of the $Fe_{82}Si_2B_{11}P_4Cu_1$ alloy

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ABSTRACT

Atom migration, phase structure and magnetic properties in the $Fe_{82}Si_2B_{11}P_4Cu_1$ amorphous alloy isothermally annealed at various heating rates are investigated. Annealing the samples at a high heating rate promotes the formation of Cu-rich regions and delays the phase separation process to high temperature, which is conductive to increase the volume fraction of the nanocrystalline phase and refine nanograins during the crystallization. When the alloy is isothermally annealed at 758 K for 180 s, the saturation magnetic flux density increases from 1.74 T to 1.79 T and the coercivity decreases from 7.8 A/m to 4.4 A/m with the heating rate increasing from 0.5 K/s to 20 K/s. Meanwhile, it confirms that the occupation of Si atoms in the lattice of *bcc*-Fe can be suppressed by the annealing with high heating rate. Increasing the heating rate is widely regarded as an effective method to improve soft magnetic properties of Fe-based nanocrystalline alloys. However, it is found that the content of the nonmagnetic atom enriched regions rises with the increase of heating rate, which can reduce the magnetic interaction in the nanocrystalline ribbons. Therefore, the exorbitant heating rate reaching 40 K/s will dramatically promote the aggregation of nonmagnetic atoms and deteriorate the soft magnetic properties.

1. Introduction

Fe-based nanocrystalline soft magnetic materials have been widely studied and used as magnetic components [1, 2]. Optimizing the crystallization process to obtain the composite structure with uniform α-Fe nanograins embedded in the amorphous matrix is the key to improve their soft magnetic properties. Nevertheless, increasing Fe content is a common means to improve the saturation magnetic flux density (B_S) of the nanocrystalline alloys [3–5]. Previous results have demonstrated that high Fe content leads to the formation of non-uniform nuclei for α-Fe nanocrystallization in the as-quenched Fe-Si-B-P-Cu amorphous alloys [6, 7]. These pre-existing nuclei for α-Fe nanocrystallization will grow during the annealing, which leads to a sharp increase in the coercivity (H_C) according to Herzer's anisotropy model [8]. Fortunately, the rapid annealing method can mitigate this problem. Sharma et al. [7] have revealed that the growth of pre-existing nuclei for α-Fe nanocrystallization is suppressed at high heating rate and consequently obtained an ultrafine grain structure in the $Fe_{85}Si_2B_8P_4Cu_1$ nanocrystalline alloy. Recently, an extremely high heating rate of about 150 K/s has been successfully realized, which significantly improves the magnetic properties of nanocrystalline alloys [9, 10]. Controlling the heating rate opens the possibility for refining grains of nanocrystalline soft magnetic alloys with high Fe content. However, these studies on heating rate mainly focus on the grain growth and soft magnetic properties of nanocrystalline alloys. Hence a comprehensive and deep understanding about the effect of heating rate on atomic-level structural evolution during the annealing is still desired.

The atomic rearrangement induced by annealing leads to the formation of local ordered structure in the amorphous matrix, which affects the crystallization process and magnetic properties of amorphous alloys. In Cu-doped Fe-based amorphous alloys, Cu atoms play a vital role in reducing the grain size because they tend to aggregate and form Cu clusters serving as heterogeneous nucleation sites for α-Fe prior to the primary crystallization [11–14]. It is noted that, except the beneficial Cu clusters, some undesirable nonmagnetic atom enriched regions also form in the amorphous phase [15, 16]. The research of the complicate two-phase structure system induced by structural relaxation and crystallization under the annealing with high heating rate is still not enough. In this work, we investigate the effect of heating rate on the microstructure and soft magnetic properties of $Fe_{82}Si_2B_{11}P_4Cu_1$ alloy. Based on the analysis of Mössbauer spectra, the local ordered structure and atom migration under various heating rates are also discussed.

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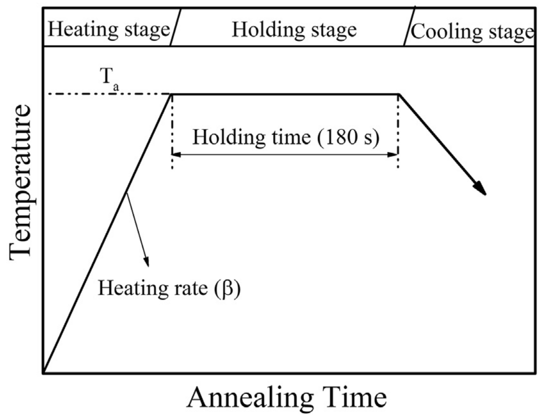

Fig. 1. Schematic diagram of isothermal annealing.

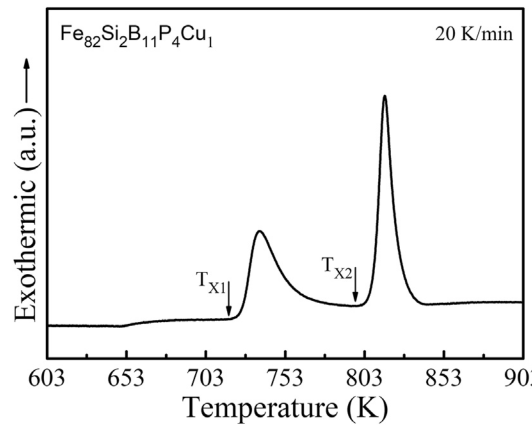

Fig. 2. DSC curve of the melt spun $Fe_{82}Si_2B_{11}P_4Cu_1$ alloy ribbons.

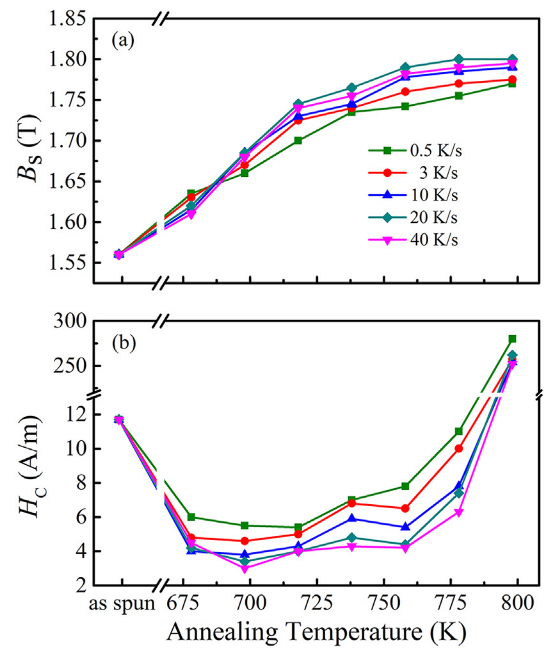

Fig. 3. Change in B_S (a) and H_C (b) of the $Fe_{82}Si_2B_{11}P_4Cu_1$ alloy ribbons as a function of the annealing temperature with heating rates ranging from 0.5 to 40 K/s.

2. Experimental details

The $Fe_{82}Si_2B_8P_4Cu_1$ alloy ingots were prepared by arc-melting industry raw materials of Fe (99.9%), Cu (99.99%), Si—Fe (Si: 99.586%, Fe: 0.27%), B—Fe (B: 17%, Fe: 82.9%) and P—Fe (P: 26.11%, Fe: 73.8%) in an Ar atmosphere. These ingots were remelted five times with magnetic stirring to ensure the chemical homogeneity. The amorphous precursor, i.e., about 3 mm wide melt-spun ribbons, were produced by rapid quenching method in an Ar atmosphere onto a copper wheel with a surface line velocity of 40 m/s. In order to study the effect of heating rate on the structural evolution and nanocrystallization in the $Fe_{82}Si_2B_8P_4Cu_1$ alloy, the melt spun ribbons were isothermally annealed at 678–798 K with various heating rates (β = 0.5, 3, 10, 20 and 40 K/s). Fig. 1 shows the schematic illustration of the isothermal annealing. T_a is the annealing temperature. The holding time and average cooling rate of all annealing processes are 180 s and 5 K/s, respectively.

The differential scanning calorimetry (DSC) was used to determine the primary and secondary crystallization temperature at a constant heating rate of 20 K/min under an Ar flow. The amorphous and annealed ribbons were characterized by X-ray diffraction (XRD) with Cu-Kα radiation and transmission electron microscopy (TEM). Transmission Mössbauer spectra were recorded at room temperature using a ^{57}Co source. All spectra were analyzed by the NORMOS program [17] and the relevant hyperfine parameters were also obtained. The B_S and H_C were measured by a DC B—H loop tracer.

3. Results and discussion

3.1. Thermal analysis

Fig. 2 shows the DSC curve of $Fe_{82}Si_2B_{11}P_4Cu_1$ melt-spun ribbons. The two obvious exothermic peaks indicate that the crystallization process includes two stages. The first and second onset crystallization temperature, marked as T_{X1} and T_{X2}, represent the formation of α-Fe (Si) phase and Fe-metalloid phase [5, 18], respectively.

3.2. Magnetic properties

Fig. 3 shows B_S and H_C of the $Fe_{82}Si_2B_{11}P_4Cu_1$ alloy as a function of annealing temperature (T_a) under various heating rates. Although the heating rate changes, the evolution of magnetic properties shows a similar trend during the annealing. The B_S gradually increases until T_a reaches 758 K and then remains unchanged at T_a = 758–798 K as shown in Fig. 3(a). It can be found in Fig. 3(b) that H_C first decreases before around 698 K due to the relaxation of internal stresses, then slightly fluctuates at T_a = 698–758 K, and finally increases with T_a further increasing to 798 K which can be attributed to the coarsening of α-Fe(Si) grains. One can see that B_S is enhanced and H_C is decreased by increasing the heating rate at any certain T_a in the range of 698–778 K, which indicates that regulating the heating rate can improve the magnetic properties of the $Fe_{82}Si_2B_{11}P_4Cu_1$ alloy.

3.3. Structural analysis

3.3.1. XRD and TEM studies

Fig. 4 shows the XRD, high resolution TEM (HRTEM) image and corresponding selected-area electron diffraction (SAED) pattern of the melt-spun $Fe_{82}Si_2B_{11}P_4Cu_1$ ribbons. The XRD pattern displays a broad peak without any crystallization peak indicating the formation of a practically complete amorphous structure. However, the HRTEM image indicates that the melt-spun ribbons contain minor small crystals with the diameter of 2–5 nm. These nanocrystals can be regarded as pre-existing nuclei [6, 7], which cannot be detected by XRD due to the small size and low content. The lattice fringes as indicated by white bars coincide to the bcc (110) plane distance of α-Fe(Si) phase.

The XRD patterns of the isothermally annealed (at 678 K for 180 s) $Fe_{82}Si_2B_{11}P_4Cu_1$ ribbons with various heating rates are shown in Fig. 5(a). The XRD patterns of the ribbons annealed at 678 K with the