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Original Research

Effect of Co addition on crystallization and magnetic properties of FeSiBPCu alloy

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Abstract

The effects of Co addition on the microstructure, crystallization processes and soft magnetic properties of $(Fe_{1-x}Co_x)_{83}Si_4B_8P_4Cu_1$ (x=0.35, 0.5, 0.65) alloys were investigated. The experimental results demonstrated that the addition of Co decreased the thermal stability against crystallization of the amorphous phase, and thus improved the heat treatment temperature of this alloy. FeCoSiBPCu nanocrystalline alloys with a dispersed α′-FeCo phase were obtained by appropriately annealing the as-quenched ribbons at 763 K for 10 min. The α′-FeCo with grains size ranging from 9 to 28 nm was identified in primary crystallization. The coercivity (H_c) markedly increased with increasing x and exhibited a minimum value at x=0.35, while the saturation magnetic flux density (B_s) shows a slight decrease. The $(Fe_{0.65}Co_{0.35})_{83}Si_4B_8P_4Cu_1$ nanocrystalline alloy exhibited a high saturation magnetic flux density B_s of 1.68 T, a low coercivity, H_c of 5.4 A/m and a high effective permeability μ_e of 29,000 at 1 kHz.
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Keywords: Crystallization; Soft magnetic properties; Microstructure; High temperature; Nanocrystalline alloy

1. Introduction

The Fe-based and FeCo-based nanocrystalline alloys have been widely investigated both experimentally [1–4] and theoretically [5–7] in physics, materials science [8–10], and engineering applications because of their excellent soft magnetic properties [11–13], including high saturation magnetic flux density (B_s), low coercivity (H_c), high effective permeability (μ_e), and low core losses. To date, three famous types of nanocrystalline alloy systems have been developed, and these systems are known as Finemet [14], Nanoperm [9,15], and Hitperm [16]. The Fe content of these nanocrystalline alloys is always low because a large amount of metal and metalloid elements, such as Nb, Zr, B, P, and Si, are added to prepare amorphous precursors and ensure a uniform nanocrystalline structure [2,17]. Therefore, high Fe content (above 80 at%), Fe-based, nanocrystalline, soft magnetic alloys are desirable for practical use. As a ferromagnetic element, it is well known that a high B_s can be obtained in FeCo-based nanocrystalline alloys via the substitution of ferromagnetic element Co for Fe in Fe-based nanocrystalline alloys [18]. Previously, researchers have developed FeSiBCu and FeSiBPCu softmagnetic alloys with high magnetic flux densities of approximately 1.8 T and 1.9 T, respectively [19]. It has been found that the marked improvement in the soft magnetic behavior is due to the reduction of the effective magnetic anisotropy, which occurs when the sizes of the nano-crystals become comparable with the magnetic exchange length [20,21]. These findings prompted us to consider whether the substitution of Co for Fe would improve

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the soft magnetic properties and thermal stability of FeSiBPCu nanocrystalline alloys, thereby permitting the formation of nanocrystalline alloys. Although the Co element may increase the cost of this alloy, it is very interesting to study the effect of Co addition on the microstructure, crystallization behavior, and magnetic properties of the amorphous and nanocrystalline phases.

Therefore, in this paper, Co was substituted for Fe in the FeSiBPCu amorphous alloy system. The FeCo at% is 83%, and the present FeCoSiBPCu glassy alloys are the so-called high Fe-content ones. Soft magnetic properties including the saturation magnetic flux density, coercivity, and permeability were investigated, and the grain size of the nano-scale crystalline phases with varying Co content were also studied.

2. Materials and methods

FeCoSiBPCu alloy ingots were prepared by arc melting mixtures of Fe (99.99% by mass), Co (99.99% by mass), Si (99.99% by mass), B (99.9% by mass), Cu (99.99% by mass), and pre-melted Fe_3P (99.9% by mass) in a highly purified Ar atmosphere. Amorphous $(Fe_{1-x}Co_x)_{83}Si_4B_8P_4Cu_1$ (x=0.35, 0.5, 0.65) alloy ribbons were produced via single-roller melt spinning. The width and thickness of the ribbons were approximately 1.5 mm and 20 μm, respectively. The chemical composition of the ribbon was checked using inductively coupled plasma spectroscopy, and no detectable deviation from the nominal composition was observed. The structures of the as-quenched and annealed ribbons were identified by X-ray diffraction (XRD) with Cu *K*α radiation and transmission electronic microscopy (TEM). The thermal stability of the as-quenched ribbons was evaluated using differential scanning calorimetry (DSC) at the heating rate of 0.67 K/s. A crystallization treatment was applied by treating the as-quenched amorphous specimens at different temperatures for 10 min under a vacuum, followed by water quenching. The average grain sizes were estimated by using Scherrer's equation and the distribution of the grain size was obtained by analyzing more than 100 spots with statistical analysis in software Gatan Digital Microscopy Suite. The saturation magnetic flux density (B_s) and coercive force (H_c) were measured under applied fields of 800 kA/m and 1 kA/m using avibrating sample magnetometer (VSM) and a dc B–H loop tracer. The effective permeability (μ_e) at 1 kHz was measured under a field of 1 A/m using a vector impedance analyzer. All property measurements were performed at room temperature.

3. Results and discussion

Fig. 1 shows the XRD patterns of the $(Fe_{1-x}Co_x)_{83}Si_4B_8P_4Cu_1$ (x=0.35, 0.5, 0.65) as-quenched ribbons, demonstrating the microstructural evolution of the alloy as a function of Co composition. All XRD spectra show one broad peak, signifying an amorphous structure in the as-quenched state.

The DSC curves measured at a heating rate of 0.67 K/s in a high-purity Ar flow for the as-quenched $(Fe_{1-x}Co_x)_{83}Si_4B_8P_4Cu_1$ (x=0.35, 0.5, 0.65) ribbons are shown in Fig. 2. It is clear that the replacement of Fe by Co brings about a

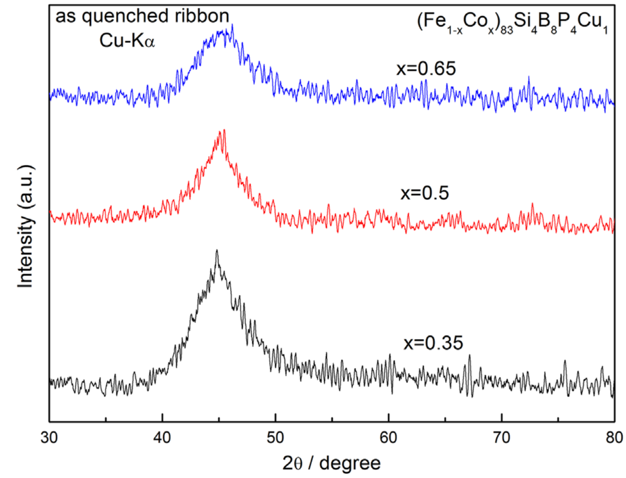

Fig. 1. XRD patterns of the $(Fe_{1-x}Co_x)_{83}Si_4B_8P_4Cu_1$ (x=0.35, 0.5, 0.65) as-quenched alloys.

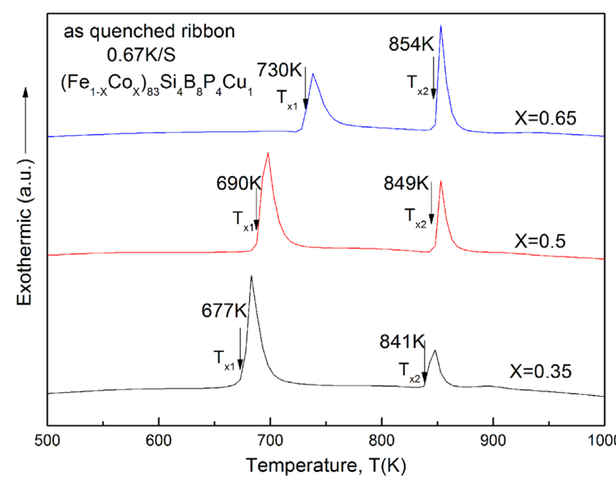

Fig. 2. DSC curves of the $(Fe_{1-x}Co_x)_{83}Si_4B_8P_4Cu_1$ (x=0.35, 0.5, 0.65) as-quenched ribbons.

change in the crystallization behavior. It can be observed that the crystallization process of these ribbons proceeded in two stages: in the first stage, partial transformation from the amorphous phase to the nanocrystalline α′-FeCo phase occured, and in the second stage, complete crystallization of the remaining amorphous phase occured. It may be noted that the initial crystallization onset temperature (T_{x1}) for $(Fe_{0.65}Co_{0.35})_{83}Si_4B_8P_4Cu_1$,$(Fe_{0.5}Co_{0.5})_{83}Si_4B_8P_4Cu_1$, and $(Fe_{0.35}Co_{0.65})_{83}Si_4B_8P_4Cu_1$ are 677 K, 690 K, and 730 K, respectively. However, the second crystallization onset temperatures (T_{x2}) are found to be 841 K, 849 K, and 854 K, respectively. With the additions of Co, the temperature interval $\Delta T_x(\Delta T_x = T_{x2} - T_{x1})$ enlarge compared with no Co content alloys [19]. Hence, in this alloy system, the Co addition leads to a significant increase in both the initial crystallization onset temperature (T_{x1}) and the second crystallization onset temperatures (T_{x2}). Thus, Co addition reduces the thermal stability against crystallization of the amorphous phase and improves the heat treatment temperature for this alloy.

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