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Influence of Co content on the structure and magnetic permeability of nanocrystalline $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloys

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ABSTRACT

Structural and soft magnetic response of nanocrystalline $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (x = 0, 0.25, 0.5, 0.75) alloys were investigated. Influences of heat treatment and Co content on the crystallization were analyzed through estimation of the crystalline volume fraction V_{cr} and thickness of the intergranular amorphous layer Λ from XRD patterns. High-temperature soft magnetic response was analyzed by the temperature evolution of the magnetic permeability from room temperature to 780 °C. It was found that the high-temperature soft magnetic behavior is improved with increasing Co content for $x \leq 0.5$, which is ascribed to reinforcing magnetic coupling between the crystals, however, a decrease of μ_i was observed for the Co richest sample. The observed μ_i behavior as a function of Co content is interpreted in terms of variations in the degree of exchange coupling.

1. Introduction

Nanocrystalline Fe–Cu–Nb–Si–B soft magnetic alloys show attractive magnetic properties such as high saturation magnetization, high permeability, and low core loss, simultaneously [1–3]. However, due to the relatively low Curie point of the residual amorphous matrix [4], they cannot be used at high temperature. Partial substitution of Co for Fe was carried out to meet the desire for high-temperature magnetic properties. When Co is added to the FeSiBNbCu alloys, crystalline phase with higher magnetic moment and Curie temperature determines their magnetic response at high temperatures [5–9]. Softness is mostly related to the exchange coupling between nanocrystalline grains through the amorphous matrix. The exchange coupling will be reinforced by increasing the saturation magnetization of the crystalline phases. As is shown [9], Co atoms selectively substitute Fe atoms into the nonequivalent Fe sites on the crystallization process and this substitution could affect the saturation magnetization of the crystalline phases and thus affect the magnetic interaction between crystals. Up to now, little attention has been paid to the discrepancy of magnetic moment of Co and Fe atoms and its effect on the magnetic exchange coupling and the soft magnetic response. In this paper, structural and soft magnetic response of nanocrystalline $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (x = 0, 0.25, 0.5, 0.75) alloys is analyzed by means of measuring μ_i–T curves [4,10–12], influence of variations in the degree of exchange coupling on the μ_i behavior as a function of Co content will be analyzed from the viewpoint of magnetic moment. In addition, changes in the crystalline volume fraction with Co content affecting the exchange coupling will also be discussed.

2. Experimental

Amorphous ribbons 5–10 mm wide, about 30 μm thick, were obtained by melt spinning technique with nominal composition $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloys (x = 0, 0.25, 0.5, 0.75). The toroidal samples with an outer diameter of about 22 mm and inner diameter of about 18 mm were fabricated by winding the ribbons into toroidal cores. In order to obtain the characteristic nanocrystalline structure, the samples were submitted to isothermal treatments (30 min) under vacuum atmosphere (10^{-3} Pa) in a tubular furnace. The phase structure of the ribbons was examined by X-ray diffraction (XRD) using D/max-2500/PC with Co Kα radiation (λ = 1.7889 Å). Differential scanning calorimetry (DSC) with a heating rate of 20 °C/min was used to determine the thermal processes of alloy studied. The permeability was in situ measured in a furnace with Ar atmosphere protection by using an HP4194A impedance analyzer at H = 0.4 A/m and f = 10 kHz and the heating rate was 10 °C/min.

3. Results and discussion

The nanocrystalline structure, mainly composed of $(FeCo)_3Si$ and α-FeCo(Si) crystals embedded in amorphous matrix, was obtained in these alloys after annealing at temperatures of

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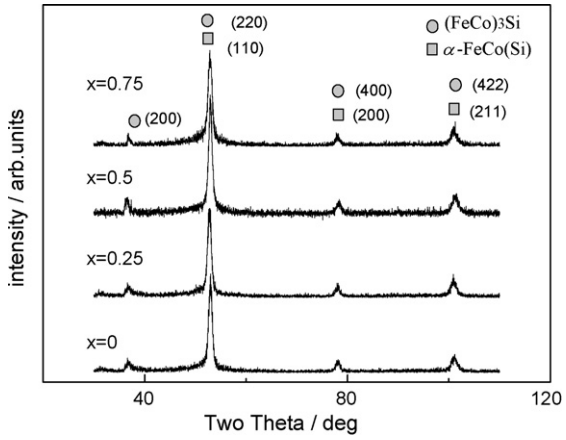

Fig. 1. XRD patterns for the 560 °C-annealed $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (x = 0, 0.25, 0.5, 0.75) alloys.

450–620 °C. The structure was monitored by X-ray diffraction. Fig. 1 shows the XRD patterns for the 560 °C-annealed $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (x = 0, 0.25, 0.5, 0.75) alloys. As presented, patterns of all the annealed samples show the characteristic diffraction peaks corresponded to crystalline precipitates. It is indicated that all the annealed samples partially crystallize after annealing and their microstructure consists of the residual amorphous phase and the nanocrystalline phase.

The crystalline volume fraction, V_{cr}, was calculated from the XRD spectra according to

$$V_{cr} = \frac{I_{cr}}{I_{cr} + I_{am}}$$

where I_{cr} and I_{am} are the integral intensities of diffraction peaks of crystalline phase and amorphous phase, respectively. These integral intensities are determined from peak areas [11,13,14] and the results are shown in Fig. 2. For the sample of x = 0.5, V_{cr} increases with T_a and the value of V_{cr} is relatively low for T_a = 470 °C because of the incomplete occurrence of the first stage of crystallization during the annealing treatment. Whereas, in the case of the alloy for x = 0.75, a higher V_{cr} of about 54% is obtained for the 470 °C-annealed sample and the V_{cr} has no significant increase with T_a in the range from 530 to 620 °C. This suggests that annealing at 530 °C for 30 min ensures the formation of stable structure for the sample of x = 0.75. In fact, V_{cr} values were obtained by numerous computations and a difference close to 5% is introduced as an error bar in data displayed in Fig. 2. It is noteworthy that the highest V_{cr} was obtained

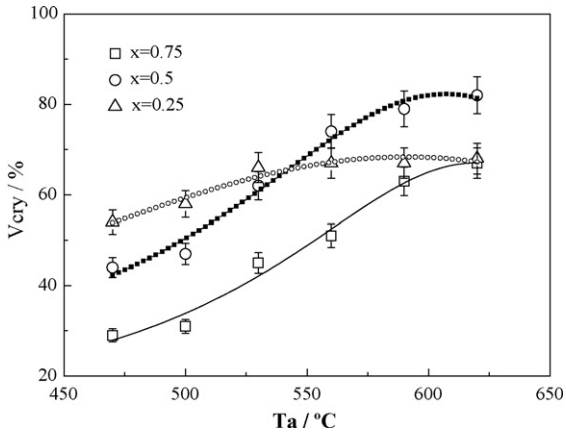

Fig. 2. The crystalline volume fraction (V_{cr}/%) for $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloys annealed at 470–620 °C.

Table 1
The average grain size, (D/nm), of $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloys annealed at 470–620 °C.

T_a (°C)	x = 0	x = 0.25	x = 0.5	x = 0.75
470		11	12	15
500	7	11	13	16
530	11	12	14	16
560	12	11	13	17
590	12	13	14	17
620	14	14	15	16

for the sample of x = 0.5 when $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloys were sufficiently crystallized. In addition, the obtained V_{cr} values are basically consistent with what were presented by Gercsi et al. [15].

The average grain size, D, was calculated from the X-ray diffraction patterns by means of the Scherrer formula. As is shown in Table 1, in principle, D increases with Co content. The calculated D values agree with the tendency of D upon Co content reported by Mazaleyrat et al. [14]. In addition, D increases slightly with T_a for $x \leq 0.5$, while it keeps nearly unchangeable with T_a from 470 to 620 °C for the sample of x = 0.75.

Additionally, the thickness of the amorphous layer between the surfaces of two adjacent nanocrystallites, Λ, can be approximately estimated as [16]:

$$\Lambda = D\left[\left(\frac{1}{V_{cr}}\right)^{1/3} - 1\right]$$

The calculated results were presented in Fig. 3. It can be seen that cobalt content in the alloys clearly affects Λ values, examined within the annealing temperature range of 470–620 °C. Taking into account the error in the employed method, a maximum deviation of 12% is computed from errors of V_{cr} and D according to the theory of errors. Such a maximum deviation is introduced as an error bar in data shown in Fig. 3. However, even within this range of estimated error, it can be clearly seen that the corresponding distance between the crystallites, Λ, becomes smaller as T_a increases, the lowest Λ value was obtained for the alloy with Co content of x = 0.5.

The analysis of the above graphs leads to conclusions that nanocrystalline structure formed during heat treatment is influenced by annealing temperature T_a, and also by the Co content. The addition of a larger amount of Co may lower the nanocrystallization temperature. With the optimal annealing conditions, the highest V_{cr} values and the lowest Λ values were found to be x = 0.5 for $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloys. With respect to the

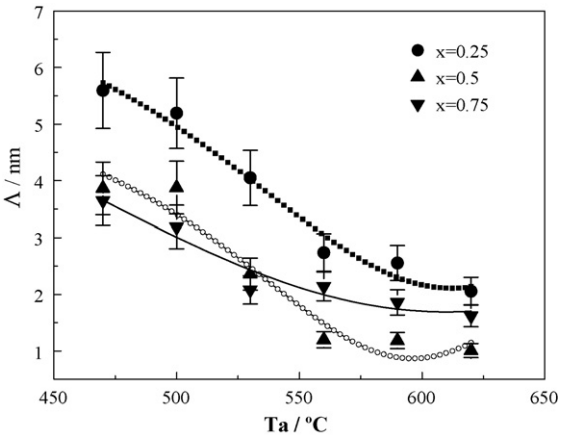

Fig. 3. The thickness of amorphous layer between the surfaces of two adjacent nanocrystallites, (Λ/nm), for $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloys annealed at 470–620 °C.