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Superior high-temperature magnetic softness for Al-doped FeCo-based nanocrystalline alloys

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A R T I C L E I N F O

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A B S T R A C T

The temperature dependence of initial permeability (μ_i-T), the average grain size (D), the Curie temperature of amorphous phase (T_c^{am}) of a series of annealed $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_{9-x}Al_x$ ($x = 0, 1, 2, 3$) alloys were investigated. The Curie temperature of amorphous phase (T_c^{am}) for the Al-contained alloys decreased compared with the Al-free alloy, but the high-temperature magnetic softness was improved. It was found that the increasing Al content enlarged the interval temperature (ΔT_x) between the onset crystallization temperature (T_{x1}) and the secondary crystallization temperature (T_{x2}). The μ_i of 530 °C vacuum annealed samples tended to increase both at room temperature (except for x = 3 alloy) and elevated temperature when Al replaced B element. The $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_6Al_3$ alloy exhibited excellent high-temperature magnetic softness, and its higher μ_i above 1000 can be maintained up to 670 °C.

1. Introduction

Finemet alloy is one of the most widely studied nanocrystalline alloys owing to its prominent soft magnetic properties, which was proposed by Yoshizawa et al. in 1988 [1]. This kind of nanocrystalline alloys, typically $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$, possesses the characteristics of high initial permeability (μ_i), high saturation magnetization (M_s), lower core loss and low coercivity (H_c). However the Curie temperature of amorphous matrix, T_c^{am}, for this alloy is not high, which limits its high temperature applications [2]. To raise the T_c^{am}, Co element was chosen to substitute for Fe partly in Fe-based nanocrystalline alloys, which improved the soft magnetic properties at elevated temperature [3–5]. Besides the T_c^{am}, Co addition broadened the temperature interval between the two crystallization temperatures, promoted the precipitation of α-(Fe,Co) crystalline phase and inhibits the precipitation of other hard magnetic phase [6]. But the Co doping into Finemet caused the saturation magnetostriction (λ_s) to increase [7–8]. It is reported that adding a small amount of Al decreased magnetic crystalline anisotropy (K_1), and therefore obtained higher initial permeability and lower coercivity (H_c) [9–10]. Replacing B with a small of Al could decrease primary crystallization temperature (T_{x1}) and increase the secondary crystallization temperature (T_{x2}) as well as temperature interval (ΔT_x), where the single soft magnetic crystalline phase precipitates from amorphous matrix [11]. It is unclear whether or not the FeCo-based nanocrystalline alloy with Al can exhibit improved soft magnetic properties. The changes made by Al element substitution for B in soft magnetic properties and microstructure for nanocrystalline $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_{9-x}Al_x$ ($x = 0, 1, 2, 3$) alloys were mainly investigated in present research.

2. Experimental procedure

$(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_{9-x}Al_x$ ($x = 0, 1, 2, 3$) amorphous ribbons with ~30 μm in thickness and ~2 mm in width were prepared by the single-roller melt spinning method with a copper wheel of 25 cm in diameter at an outer surface velocity of 3000 r/min. The ribbon samples were wound into toroidal cores with an outer diameter of about 18 mm and an inner diameter of about 16 mm. In order to achieve the dual-phase nanocrystalline structure, these samples were submitted to isothermal annealing at 530 °C for 0.5 h under vacuum atmosphere (10^{-3} Pa). The crystallization temperature of amorphous ribbons (T_x) was determined by the differential scanning calorimetry (*DSC*) with a heating rate of 10 °C/min from room temperature to 1000 °C in a Perkin-Elmer DSC7. The microstructure of annealed ribbons was examined by X-ray diffraction (*XRD*) using D/max-2500/PC with Cu-Kα radiation (λ = 1.54056 Å). The initial permeability μ_i was measured in situ in a furnace with Ar atmosphere protection using an HP4294A impedance analyzer at $H = 0.4$ A/m and $f = 10$ kHz, and the heating rate is 10 °C/min over the temperature range of 30–700 °C.

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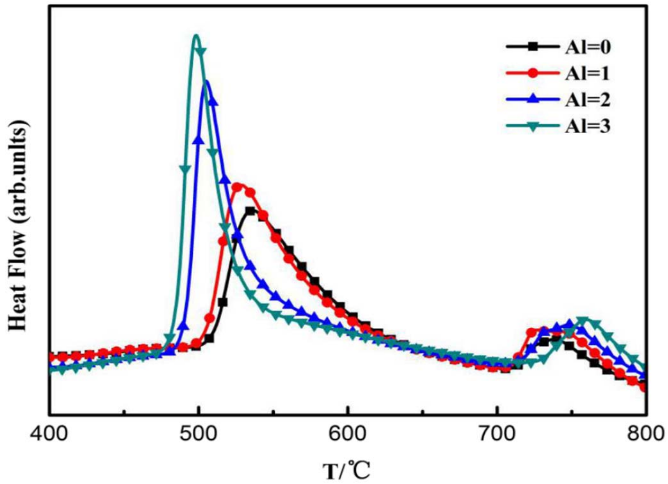

Fig. 1. DSC curve of as-quenched $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_{9-x}Al_x$ (x = 0, 1, 2, 3) alloys at a heating rate of 10 k/min.

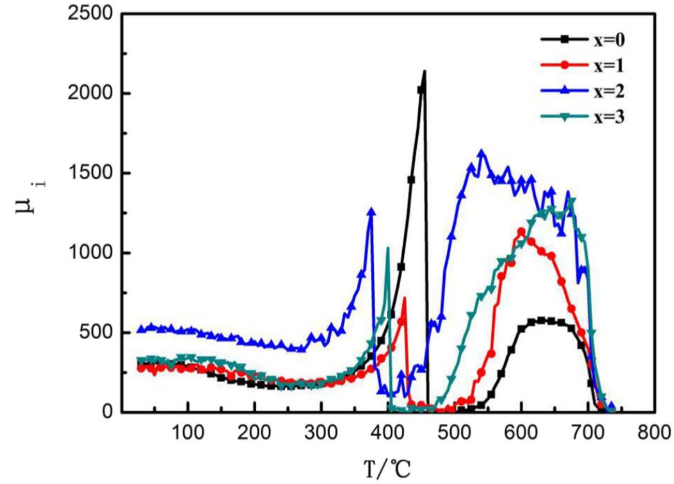

Fig. 2. μ_i-T curves of as-quenched $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_{9-x}Al_x$ (x = 0, 1, 2, 3) alloys.

3. Results and discussion

The consequence of the differential scanning calorimetry (*DSC*) measurement about the series of as-quenched $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_{9-x}Al_x$ (x = 0, 1, 2, 3) alloys is shown in Fig. 1. Two different stages were found during the process of the devitrification for all samples. The first exothermic peak, related with the precipitation of crystallized phases α-Fe(Si,Al) and Fe_3(Si,Al), appears when temperature (T_{x1}) is low. The second one is the hard magnetic phase separating out from the master alloy at the secondary crystallization temperature (T_{x2}). As shown in the graph, the addition of the Al component shifted the primary crystallization peaks towards lower temperatures from 510 °C at $x = 0$ to 475 °C at $x = 3$, but moved the position of the second crystallization peaks, which correspond to the formation temperature of the boride phase, to higher temperatures from 716 °C at $x = 0$ to 731 °C at $x = 3$, and therefore extended the interval temperature (ΔT_x) between the first and second crystallization temperature. When Al content gradually increased from Al-free to 3%, the ΔT_x varies from 206 °C to 256 °C. Distinctly, the sample with 3% Al content exhibits the largest ΔT_x about 256 °C guaranteeing the stability of the first crystallization phase in a wide temperature range. This can be attributed to the fact that the Al addition not only partitioned to the α-Fe(Si, Al) nanocrystallites, but also strongly concentrated in the Cu-rich particles. And the interaction of Al and Cu affected the kinetics of the early stages of the nucleation of the nanocrystallites [12].

The temperature dependence of initial permeability (μ_i-T curve) for as-quenched $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_{9-x}Al_x$ (x = 0, 1, 2, 3) alloys was measured and the results are shown in Fig. 2. A faster decrease of magnetic anisotropy than that of saturation magnetization with increasing temperature causes the classical sharp Hopkinson peak [13]. The μ_i suddenly decreases to zero at Curie temperature (T_c^{am}) owning to the ferro-paramagnetic transition of the amorphous alloy [14]. The result shows that the T_c^{am} of the Al-free alloy is about 460 °C. The T_c^{am} are about 430 °C for $x = 1$, 380 °C for $x = 2$ and 405 °C for $x = 3$, respectively. It can be found that the T_c^{am} of Al-containing alloys is lower than that of Al-free alloy. The reason is maybe that the excess Al destroys the exchange reaction between the Fe-Co atoms or the partial crystallization in the formation of amorphous alloys [11,15]. In addition, when the temperature is higher than T_c^{am}, the successive precipitating of soft magnetic crystal phase from the paramagnetic amorphous matrix causes an increasing tendency of the initial permeability to all the alloys. And the μ_i begins to decline once more after keeping relatively stable value for a period of time. Perhaps because the transformation from ferromagnetic to paramagnetic state of crystallization phase or Fe-B hard magnetic phase's precipitation deteriorates soft magnetic properties at elevated temperature [16] (Table 1).

Table 1
The primary crystallization temperature (T_{x1}), the secondary crystallization temperature (T_{x2}), the interval temperature (ΔT_x) and the Curie temperature (T_c^{am}) of the $(Fe_{0.5}Co_{0.5})_{73.5}Si_{13.5}Nb_3Cu_1B_{9-x}Al_x$ (x = 0, 1, 2, 3) alloys.

Alloys	T_{x1} (°C)	T_{x2} (°C)	ΔT (°C)	T_c^{am} (°C)
$x = 0$	510 ± 0.8	716 ± 0.6	206 ± 1.4	460 ± 2
$x = 1$	505 ± 0.8	712 ± 0.6	207 ± 1.4	430 ± 2
$x = 2$	483 ± 0.8	720 ± 0.6	237 ± 1.4	380 ± 2
$x = 3$	475 ± 0.8	731 ± 0.6	256 ± 1.4	405 ± 2

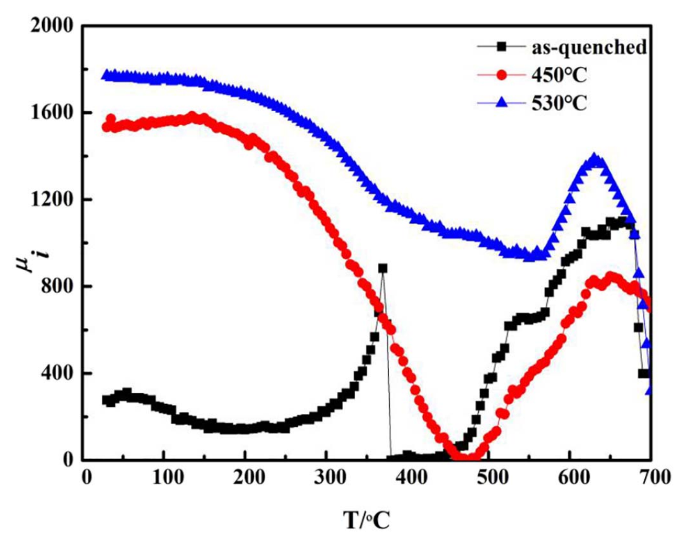

Fig. 3. μ_i-T curves of as-quenched 450 °C and 530 °C vacuum annealed $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_6Al_3$ alloy.

Fig. 3 depicted the evolution of the initial permeability μ_i with temperature T (μ_i-T curves) for the 450 °C and 530 °C annealed $(Fe_{0.5}Co_{0.5})_{73.5}Nb_3Si_{13.5}Cu_1B_6Al_3$ alloy. What is shown is that the Hopkinson peaks disappeared after annealing at 450 °C and 530 °C in virtue of the formation of dual-phase nanocrystal microstructure. For the 450 °C annealed alloy, the μ_i progressively decreases to zero from room temperature to 460 °C, and then gradually increases from zero to 800 during temperature increasing from 460 °C to about 650 °C. For the 530 °C annealed alloy, the μ_i only shows a slow decrease from room temperature up to 550 °C. Above 550 °C, it gradually increases until 650 °C, then it drops when temperature is higher than 650 °C. The