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Effects of minor precipitation of large size crystals on magnetic properties of Fe-Co-Si-B-P-Cu alloy

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ABSTRACT

Nanocrystalline $Fe_{81.3}Co_4Si_{0.5}B_{9.5}P_4Cu_{0.7}$ alloy shows low coercivity (H_c <10 A/m), but a minor increase in Co from 4 to 5 at.% ($Fe_{81.3}Co_5Si_{0.5}B_{8.5}P_4Cu_{0.7}$) results in a drastic increase in H_c (>60 A/m). In terms of structure both the alloys in as-quenched state exhibit similar X-ray diffraction patterns (i.e. X-ray amorphous). Similar to low concentration of Co in the alloy, it is possible to realize a fine nanocrystalline structure made from bcc Fe-Co phase for higher concentration of Co. However, existence of a very few large sized crystals (above 100 nm) in as-quenched state enhances the H_c. A large numbers of bcc Fe(-Co) nuclei (~2–5 nm) in as-quenched state results in a high magnetic flux density (B_s) after optimum annealing. It is possible to obtain a B_s of 1.88–1.90 T in this system but it is at the cost of an increase in H_c.

1. Introduction

Recently, power and energy problems have required the development of soft magnetic materials. The research in Fe-based soft magnetic alloys has been focused to nanocrystalline alloys because of their excellent performance, e.g. high magnetic flux density (B_s), high magnetic permeability (μ), low coercivity (H_c) and low core loss (W) [1–13]. NANOMET® (Fe-Si-B-P-Cu alloys), a typical nanocrystalline material with high Fe content (above 83 at.%) has attracted much attention as a high performance core material for magnetic applications [13–18]. NANOMET® alloys exhibit high B_s (~1.85 T), low H_c (<10 A/m), significantly low W ($W_{1.7/50} = 0.4$ W/kg) and relatively lower material cost compared to other nanocrystalline systems which usually contain Nb, Zr, Hf etc. as one of the elements. Afterwards, a small addition of Co (~4–5 at.%) is shown to be effective in producing wider ribbons for commercial applications (such as transformers, motors etc.) [19]. High concentration of magnetic elements (Fe+Co ~85.2 at.%) in the alloy is necessary to obtain high B_s ~ 1.84 T [20–22]. Simultaneous existence of high B_s similar to oriented steel, and H_c lower than it are the driving factors to increase the magnetic elements further.

Before changing the alloy composition, there may arise several questions, for example, “Can we improve magnetic properties further? What is the limit of high B_s without lowering the magnetic softness? What will be the obstacle in obtaining high B_s and low H_c simultaneously?” During our efforts to enhance the B_s of Fe-Co-Si-B-P-Cu alloys (by increasing the amount of Co in the alloy), we noticed that some of the ribbons exhibiting similar X-ray diffraction pattern (i.e. a broad halo in X-ray diffraction curve) in as-quenched state showed significantly higher H_c. Generally it is believed that the amorphous structure of as-quenched ribbon is very important for achieving a uniform nanocrystalline structure with low H_c after optimum heat treatment. Amorphous forming ability of an alloy depends on alloy composition. Metal to metalloid ratio is very important. In Fe-Si-B-P-Cu alloys (i.e. NANOMET®), we noticed that it is very difficult to produce amorphous ribbons with total magnetic elements higher than 86 at.%. From the magnetic point of view, FeCo alloys exhibit higher B_s than Fe. Therefore, we planned to examine the ribbons of $Fe_{81.3}Co_xSi_{0.5}B_{13.5-x}P_4Cu_{0.7}$ (x = 3–6 at.%) alloys. The concentration of B is relatively high among the metalloid elements present in the alloy, therefore replacement of B with Co seems to be a better option. Our results show that it is possible to make an amorphous ribbon with metallic elements (Fe+Co) higher than 86 at.%, but the H_c obtained after annealing is relatively high. It is difficult to understand because X-ray amorphous ribbons usually show low H_c after optimum annealing. Present study clarifies the reasons for higher H_c in X-ray amorphous like Fe-Co-Si-B-P-Cu

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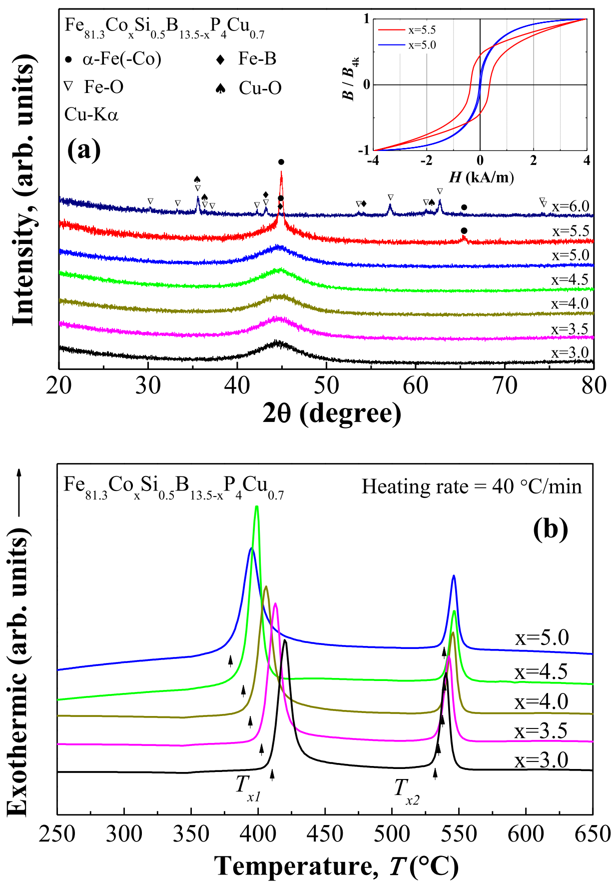

Fig. 1. (a) XRD profiles and (b) DSC curves for the as-quenched ribbons of $Fe_{81.3}Co_xSi_{0.5}B_{13.5-x}P_4Cu_{0.7}$ (x = 3, 3.5, 4, 4.5, 5, 5.5, and 6). Inset in Fig. (a) shows the B-H curves for as quenched ribbons with x = 5 and 5.5.

ribbons. We also emphasize on the upper limit and obstacles for obtaining high B_s and low H_c simultaneously. Based on experimental results and the model of nanocrystallization in NANOMET® type alloys [18], we figured out that the upper limit for simultaneous existence of high B_s and low H_c in Fe-Co-Si-B-P-Cu alloys is governed by the minor precipitation of larger sized α-Fe(-Co) crystals in as-quenched state.

2. Experiments

Alloy ingots with a nominal composition of $Fe_{81.3}Co_xSi_{0.5}B_{13.5-x}P_4Cu_{0.7}$ (x = 3, 3.5, 4, 4.5, 5, 5.5, and 6.0 at.%) were prepared by arc-melting a mixture of high purity metals (99.9 mass% Fe, 99.9 mass% Cu and 99.5 mass% Co), metalloid (99.9 mass% Si and 99.5 mass% B) and pre-melted Fe_3P (99 mass%). The alloy ingots were smashed to small pieces, so that they can be inserted into a quartz tube with a nozzle (width ~0.3 mm) at its one of the end. These

Table 1
The thermal parameters obtained from DSC curves as shown in Fig. 1 (b).

Composition $Fe_{81.3}Co_xSi_{0.5}B_{13.5-x}P_4Cu_{0.7}$	T_{x1} °C	T_{x2} °C	ΔT (=T_{x2} - T_{x1}) °C	1st Exothermic peak J/g
x = 3	410	532	122	84
x = 3.5	403	535	132	87
x = 4	394	538	144	96
x = 4.5	389	539	150	95
x = 5	380	539	159	104

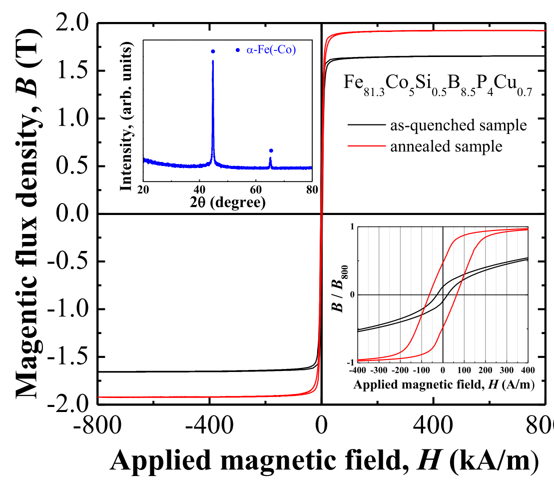

Fig. 2. Hysteresis curves of as-quenched and annealed ribbons of $Fe_{81.3}Co_5Si_{0.5}B_{8.5}P_4Cu_{0.7}$ alloy. Inset in the bottom shows the corresponding B-H loop tracer curves magnified in low field region. The VSM measurement focuses on B_s whereas B-H loop tracer is for precise measurement of H_c. Inset on the top is an XRD curve for the optimally annealed ribbon.

pieces of alloy in a quartz tube were melted by high-frequency induction melting, and then a jet of molten alloy was ejected (by argon gas ~0.05–0.06 MPa) through the quartz nozzle on a rapidly rotating copper wheel in air. This process produces ribbons (width ~4–6 mm and thickness ~20 μm) of desired alloy composition. The roller surface velocity was about 42 m/s.

The crystallization of amorphous ribbons was carried out in an infrared furnace (IR furnace) under the flowing argon gas. As-quenched ribbons were annealed in the temperature (T_a) range of ~350–500 °C for 10 min. Heating rate (*HR*) of the furnace/sample, starting from room temperature to required T_a was varied between ~10 and 800 °C/min. The optimum T_a (~420 °C) for each composition is the one which results in highest B_s and lowest H_c simultaneously. The microstructure (including crystal structure, grain size, and crystal distribution) of melt-spun and annealed ribbons was examined by X-ray diffraction (XRD) with Cu Kα radiation, and transmission electron microscopy (TEM). Standard TEM sample preparation method i.e. thinning of ribbon by Ion Milling (Model 1010 from E.A. Fischione Instruments. Inc.) under the flowing argon gas was used. Thermal properties were studied by using a differential scanning calorimetry (DSC, SII EXSTAR DSC6300) at a heating rate of 40 °C/min under flowing argon gas. Saturation

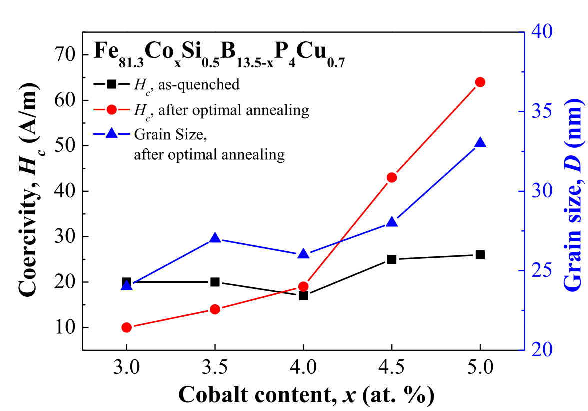

Fig. 3. Coercivity (H_c) and grain size (*D*) of bcc Fe(-Co) for as-quenched and annealed ribbons of $Fe_{81.3}Co_xSi_{0.5}B_{13.5-x}P_4Cu_{0.7}$ (x = 3, 3.5, 4, 4.5, and 5). The solid lines between the data points are only as a guide to the eye.