Improvement of Soft Magnetic Properties in (Fe$_{0.85}$B$_{0.15}$)$_{100-x}$Cu$_x$
Melt-Spun Alloys

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The Cu content dependence of magnetic properties in annealed (Fe$_{0.85}$B$_{0.15}$)$_{100-x}$Cu$_x$ alloys fabricated by melt-spinning was discussed. The value of $H_c$ markedly decreases with increasing x between $x = 1.0$ and 1.5, accompanied with significant reduction of crystalline grain size. The alloy with $x = 1.5$ showed excellent magnetic properties such as a small $H_c$ of about 7 A/m and a high $B_c$ of more than 1.8 T. For the present alloy system, more than 1.0% addition of Cu is effective for the formation of nano-scale grains and for the improvement of the soft magnetic properties.

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1. Introduction

Recently, the energy issue became one of most crucial problem. Therefore, Fe-based amorphous alloys and/or Fe-based nanocrystalline materials have been extensively studied because of their low core loss.\(^1\)\(^-\)\(^3\) However, there are difficulty to use them as a substitution of Si steels. The size of the core formed by these materials is larger than that formed by Si steel because $B_s$ in these materials is at most 1.7 T,\(^1\)\(^-\)\(^5\) whereas that in Si steels is about 2.0 T.\(^6\)\(^-\)\(^8\) From this point of view we have developed the nanocrystalline Fe-based soft magnetic materials with a lower core loss than Si steel and a higher $B_s$ than conventional Fe-based amorphous and/or nanocrystalline alloys.\(^9\) Such materials was fabricated by annealing the melt-spun Fe-Cu-B amorphous alloy.\(^9\)

In this alloy, the primary crystals in the as-quenched state are necessary for the appearance of excellent soft magnetic properties.\(^9\) It suggests that Cu plays a crucial role in the appearance of the nanocrystalline structure. To confirm the role of Cu addition, the Cu content dependence of magnetic properties and average crystalline grain size in annealed (Fe$_{0.85}$B$_{0.15}$)$_{100-x}$Cu$_x$ alloys formed by melt-spinning have been investigated in the present study. The effect of Cu on the magnetic properties, the nanocrystallization process and the microstructure are discussed.

2. Experimental

Amorphous (Fe$_{0.85}$B$_{0.15}$)$_{100-x}$Cu$_x$ ($x = 0.0$, 0.5, 1.0 and 1.5), alloy ribbons were prepared by a single-roller melt-spinning technique. The width and thickness of the ribbons were 5 mm and 21 µm, respectively. The specimens were annealed to form nano-scale grains. The typical annealing conditions which bring about most excellent soft magnetic properties were at 663 K for 3.6 ks in a N₂ atmosphere.\(^9\) The dc $B$-$H$ curves were measured using a $B$-$H$ curve tracer. The X-ray diffraction measurement was carried out using the Cu-K$_{α1}$ tube with wave length of $\lambda = 0.15406$ nm. The microstructure was observed by a transmission electron microscopy (TEM).

Fig. 1  $B$-$H$ curves for (Fe$_{0.85}$B$_{0.15}$)$_{100-x}$Cu$_x$ alloys annealed at 663 K for 3.6 ks.

3. Results and Discussions

The dc $B$-$H$ curves for (Fe$_{0.85}$B$_{0.15}$)$_{100-x}$Cu$_x$ alloy ribbons annealed at 663 K for 3.6 ks are shown in Fig. 1. The coercive force $H_c$ decreases significantly with Cu content, while the magnetic flux density at 1 kA/m, $B_{1k}$ shows a slight increase. Note that the saturation magnetic flux density $B_s$ shows little Cu content dependence and it was about $B_s = 1.83$ T. On the other hand, due to excellent saturation behavior of $B$-$H$ curve in the alloy with $x = 1.5$, the value of $B_{1k}$ for this alloy is higher than other concentration alloys. For the alloy with $x = 1.5$, the maximum permeability is larger than 60000 and $H_c = 6.9$ A/m. The Cu content $x$ dependence of $H_c$ for (Fe$_{0.85}$B$_{0.15}$)$_{100-x}$Cu$_x$ alloys annealed at 663 K for 3.6 ks is shown in Fig. 2. The value of $H_c$ decreases with increasing $x$ and shows drastic reduction between $x = 1.0$ and 1.5. Thus, the soft magnetic properties are improved by adding Cu. In particular, for concentration region of $x > 1.0$, Cu addition is very effective in improving soft magnetic properties. It is well known that microstructure greatly relates with soft magnetic behavior. The X-ray diffraction patterns for (Fe$_{0.85}$B$_{0.15}$)$_{100-x}$Cu$_x$ alloy with $x = 0.0$ and 1.5 annealed at 663 K for 3.6 ks are shown in Fig. 3. In both alloys, a bcc Fe...
phase was confirmed. However, in detailed, the halo of amorphous state is observed more significantly and peaks for bcc Fe phase are sharper in $x = 0.0$ than in $x = 1.5$. Therefore, difference in the average grain size of bcc Fe is suggested. In Fig. 4, the Cu content $x$ dependence of the average grain size $D$ for $(\text{Fe}_{0.85}\text{B}_{0.15})_{100-x}\text{Cu}_x$ alloys annealed at 663 K for 3.6 ks is shown. Here, $D$ is estimated from Sherrer’s relation using the 310 peak in the X-ray diffraction data. The $D$ decreases with increasing $x$. In the concentration region of $x \leq 1.0$, $D$ is larger than 30 nm, while it becomes about 20 nm at $x = 1.5$. In Fig. 5, the $D$ dependence of $H_c$ is shown. It has been pointed out in a study of Fe-Cu-Nb-Si-B system that $H_c$ decreases with decreasing the grain size to nano-order.\(^{10,11}\) The increment of $H_c$ is almost proportional to $D^6$ in the present alloy system, that behavior is similar to other nanocrystalline alloy systems.\(^{10,11}\) In Figs. 6 (A)–(D), the TEM images for the as-quenched and annealed alloy with $x = 0.0$ and 1.5 are shown. In the as-quenched state in the alloy with $x = 0.0$, the grains are hardly observed (Fig. 6 (A)). While nano-order particles are precipitated in the as-quenched state in the alloy with $x = 1.5$ (Fig. 6 (B)). In the annealed state alloys, significantly different crystalline state is observed. In the alloy with $x = 0.0$, the crystalline grains are enlarged by annealing. They are larger than 50 nm and surrounded by an amorphous phase (Fig. 6 (C)). On the other hand, a high number density of nanocrystalline grains smaller than 20 nm is observed in the annealed alloy with $x = 1.5$ (Fig. 6 (D)). The number density is about $10^{23} \sim 10^{24}/\text{m}^3$, which is comparable with the number density of Cu clusters in a Fe-Cu-Nb-Si-B nanocrystalline alloy estimated in an atom-probe study.\(^{12,13}\) Note that on the inside of the ribbon, randomly oriented bcc grains are confirmed by electron diffraction patterns observation. According to DSC measurement, the crystallization temperature $T_{X1}$ reduces and the precipitation temperature of Fe-B compounds $T_{X2}$ increases with increasing Cu content.\(^9\) Namely, the annealing temperature range for the formation of bcc Fe nanocrystals without precipitation of the Fe-B compounds becomes wider by increasing Cu content. Moreover, since B hardly solidifies in the bcc-Fe phase, the content of B in the remaining amorphous matrix increases with increasing number of nanocrystalline grains. This suggests that the grain growth is suppressed by the increase in B content in the remaining amorphous matrix.

Since the grains are large and their number density is low in the annealed Cu-free Fe-B alloy ($x = 0.0$) as shown in Fig. 6 (C), the effect of magnetocrystalline anisotropy remains. On the other hand, as shown in Fig. 6 (D), the nano-scale grains exist with high number density in the alloy with $x = 1.5$, and the effective magnetic anisotropy becomes small. In the present alloy system, Cu content brings about reduction of $D$ and improvement of soft magnetic behavior. However, the effect of Cu content is small in the concentration region of $x \leq 1.0$, and it becomes significant in $x > 1.0$. Unlike the Fe-Cu-Nb-Si-B alloy system,\(^{10}\) the existence of primary crystalline grains in the as-quenched
state is necessary for the appearance of the nanocrystalline state in the present alloy system, as observed in Fig. 6 (B). It is considered that the Cu clusters act as the nucleation sites of primary crystals, and the opportunities for nucleation increase with increasing Cu content. Since the exothermic peak corresponding to the crystallization observed in DSC curve for the alloy with $x = 1.5$ is broad and spread over a wide temperature range, crystallization occurs slowly, while crystallization in other concentration alloys occurs rapidly. In the alloy with $x = 1.5$, nucleation of primary crystals in high number density brings about simultaneous grain growth.

4. Conclusion

In conclusion, the high $B_s$ soft magnetic nanocrystalline Fe-B-Cu alloys were developed by annealing melt-spun ribbons. In the present study, the improvement of magnetic properties was discussed. The value of $H_c$ decreases with increasing Cu content and exhibits significant reduction between $x = 1.0$ and 1.5, accompanied with significant reduction crystalline grain size. Excellent magnetic properties such as a low $H_c$ of about 7 A/m and a high $B_s$ of more than 1.8 T were obtained in the alloy with $x = 1.5$. In the present alloy system, more than 1.0% Cu addition brings about nucleation of primary crystals in high number density in the as-quenched state and the reduction of the average grain size by simultaneous grain growth by annealing.

REFERENCES


Fig. 6 TEM images: (A) as-quenched alloy with $x = 0.0$ and (B) $x = 1.5$ and (C) alloy with $x = 0.0$ annealed at 663 K for 3.6 ks, and (D) alloy with $x = 1.5$ annealed at same condition.