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MAGNETIC PROPERTIES OF Fe-Al-B-Zr-Cu ALLOYS WITH FINE NANOCRYSTALLINE STRUCTURE

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The crystallization behaviors and magnetic properties for $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ (x=0, 1, 2 at%) alloys is investigated. By the addition of 1~2 Cu, the temperature range, where a single bcc phase exists, expands largely over 200 K and the grain size of bcc phase represents to less than 10 nm. For the optimally annealed Cu-added alloys, the high μ_e (1 kHz) above 20000 combined with the high B_{10} of about 1.4 T is obtained in nanocrystalline state. The low core loss of 95.8 W/kg at 0.1 T and 100 kHZ is confirmed for the nanocrystalline $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloy.

I. INTRODUCTION

Recently, we have carried out a systematic study on the development of new soft magnetic materials in Fe-Al-B-Zr alloy system. As was reported [1], the good soft magnetic properties for $Fe_{86-x}Al_4B_{10}Zr_x$ ($5 \leq x \leq 10$) alloys annealed from amorphous state were obtained below the onset temperature of the first-stage crystallization. As crystallization started, the soft magnetic properties were deteriorated rapidly through the coarsening of bcc phase and the formation of Fe-B compounds. For the optimally annealed amorphous alloys, the obtained magnetic properties were as follows, μ_e=17000~25000 at 1 kHz, H_c=20~30 mOe and B_{10}=0.6~1.1 T. However, because T_c and B_{10} in the amorphous state of these alloys are much lower than those in the crystallized state, it is needed to improve T_c and B_{10} characteristics by the proper addition of alloying elements to Fe-Al-B-Zr system and crystallization treatment.

It is well known that a metastable bcc phase with ultrafine grain size is obtained and hence a large improvement of their soft magnetic properties is caused by annealing of amorphous Fe-Si-B-Nb-Cu [2] and Fe-M-B-Cu (M=transition metal) [3] alloys in the vicinity of crystallization temperatures. It had subsequently been clarified that the addition of Cu to Fe-Si-B alloys enhanced the instability of the amorphous structure and gave an easy formation of the nanocrystalline bcc phase [4].

Similarly, the addition of Cu is also expected to bring about a fine-grained bcc structure in the Fe-Al-B-Zr amorphous alloys. This paper is intended to clarify the effect of Cu on the crystallization behaviors and magnetic properties of amorphous Fe-Al-B-Zr alloys.

II. EXPERIMENTAL PROCEDURES

Amorphous $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$(x=0,1,2 at%) alloys, about 1.5 mm wide and 18 μm thick, were prepared by a single-roller melt spinning method in Ar gas atmosphere. The crystallization behaviors of amorphous alloys were examined by a differential thermal analyzer (DTA) at a heating rate of 10 K/min. The Curie temperature (T_c) and thermomagnetization (σ_T) of amorphous alloys were studied in the temperature range from 298 to 1083 K in an applied field of 10 kOe using a vibrating sample magnetometer. The amorphous alloys were wound into toroidal cores with 21 mm inner diameter and annealed in the temperature range of 573 to 923 K for 1 h in the vacuum state of ~10^{-3} Torr. The structures of the annealed alloys were estimated by an X-ray diffraction (XRD) using Cu-K_α radiation and transmission electron microscopy (TEM). The effective permeabilities (μ_e) of the annealed alloys were measured by an impedance analyzer in an applied field of 10 mOe. The dc magnetic properties of the magnetic induction (B_{10}) and the coercive force (H_c) were measured by a hysteresis loop tracer under 10 Oe and 0.1 Oe, respectively. The core loss (W_L) was measured by using a B-H analyser.

III. RESULTS AND DISCUSSION

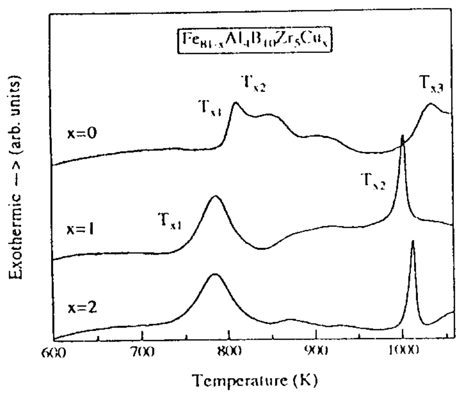

Fig.1 DTA curves of amorphous $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ (x=0,1 alloys heated at the rate of 10K/min.

Figure 1 shows the DTA curves, revealing the effect of Cu addition on the crystallization processes of amorphous $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$(x=0,1,2) alloys. The precipitated crystalline phases represented in Fig.1 were identified by X-ray diffraction. As reported separately [1], the amorphous alloy with x=0 crystallized through three stages, consisting of the first-, second- and third-stage changes from amorphous to bcc, mainly Fe_3B and Fe_3Zr respectively. However, the alloys with x=1 and 2 crystallize through two stages, corresponding to the changes from amorphous to bcc and mainly $Fe_2B(Zr)$, respectively. Here, the T_{x1}, T_{x2} and T_{x3} represent the onset temperatures of the precipitation of bcc, Fe_3B and Fe_3Zr in case of x=0, while T_{x1} and T_{x2} do bcc and $Fe_2B(Zr)$ phases in case of x=1 and 2, respectively. As shown in this figure, T_{x1} and T_{x2} for the Cu-free $Fe_{81}Al_4B_{10}Zr_5$ amorphous alloy are overlapped, while for the Cu-added alloys the temperature gap between the two peaks is expanded largely over 200 K. Furthermore, T_{x1} shifts to a lower temperature side. This result indicates that the addition of Cu in the amorphous Fe-Al-B-Zr alloy results in an extension of the temperature range where the single bcc phase field exists.

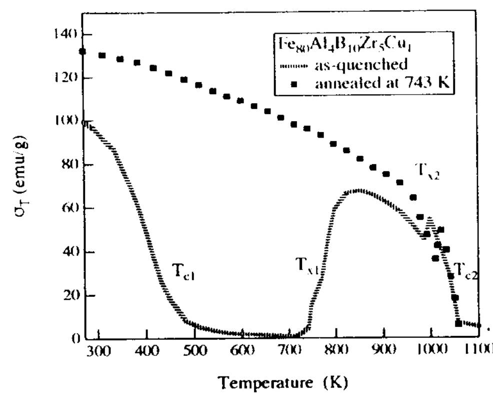

Fig.2 Thermomagnetic curves for amorphous $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloys as-quenched and annealed for 1 h at 743 K under an applied field of 10 KOe. The heating rate is 10 K/min.

Figure 2 shows the temperature dependence of saturation magnetization (σ_T) for the $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloys as-quenched and annealed at 743 K for 1h, respectively. The T_{x1} and T_{x2} correspond to the onset temperatures of the first- and second-exothermic peaks on the DTA curves measured at the same heating rate. As the temperature increases, σ_T for the as-quenched alloy decreases remarkably and approaches nearly zero value at 470 K, which is the Curie temperature (T_{c1}) for the amorphous alloy. The further rise of heating temperature results in the significant increase of σ_T in the vicinity of T_{x1}, which is due to the precipitation of the bcc phase from amorphous matrix. It is notable the discontinuous increase at T_{x2}, which is presumably due to the structural change from the remaining amorphous phase to the bcc α-Fe(Al) and $Fe_2B(Zr)$ ones, where pure α-Fe and Fe_2B phases have the T_c values of about 1043 and 1015 K [5,6], respectively. The bcc $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloy annealed at 743 K for 1 h is also recognized this phenomenon at T_{x2}. Furthermore, the Curie temperature (T_{c2}) of this alloy is about 1040 K and higher than those of other Fe-based nanocrystalline materials.

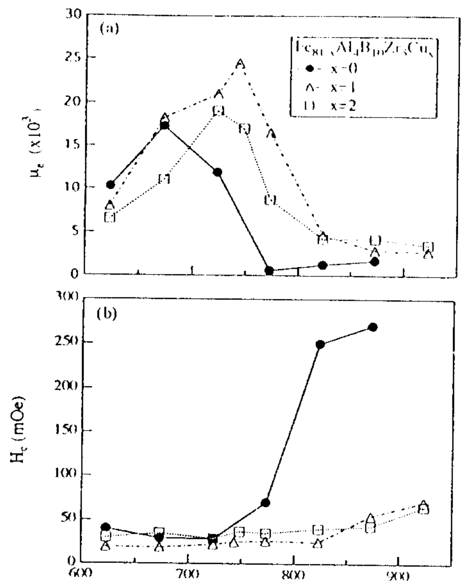

Fig.3 (a) Effective permeabilities, μ_e, at 1 kHz and (b) coercive force, H_c, as a function of T_a for amorphous $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ (x=0,1,2) alloys.

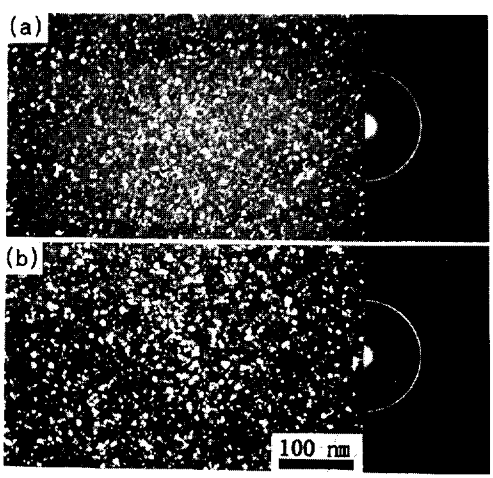

Fig.4 Dark-field microstructures and corresponding diffraction patterns of $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloys annealed for 1 h at (a) 743 K and (b) 823 K.

Figures 3(a) show the changes in μ_e at 1 kHz as a function of annealing temperature, T_a, for the amorphous $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ alloys. The maximum value of μ_e at x=0 is located below T_{x1} and decreases significantly to nearly zero in the crystallite state. However, the μ_e values of the alloys with x=1 and 2 increase steadily and then reaches the maxima as high as 20000~25000 in the T_a range of 723 to 743 K, where the structural change from the amorphous to the bcc phase is observed. With further increasing T_a, they decrease rapidly and approach about 5000 at 823 K. As shown in Fig.3(b), H_c for the alloy of x=0 increases largely in the T_a range above T_{x1}, while for those of above x=1 are constant in the T_a range of 673~873 K.

In order to clarify the reason for the rapid change of the μ_e values for the Cu-containing alloys in the range of $T_{x1} \leq T_a \leq T_{x2}$, the microstructure of the amorphous $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloy was examined by TEM. As shown in Fig.4, the sample annealed at 743 K consists of a bcc phase with a homogeneous and ultrafine grains of 5~8 nm. However, the one at 823 K includes partially the inhomogeneous and large grains of 8~15 nm. Accordingly, it is presumed that the excellent soft magnetic characteristics for the bcc $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ (x=1, 2) alloys are strongly dependent on the grain size of bcc phase

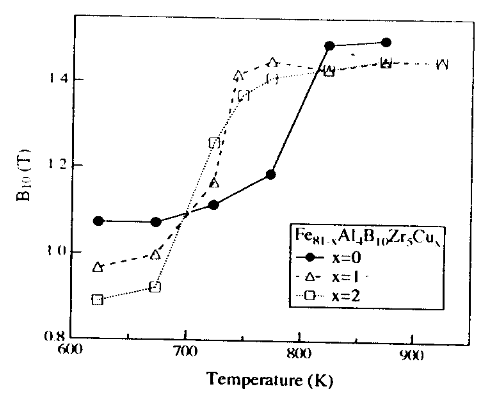

Fig.5 Magnetic induction, B_{10}, calculated by a hysteresis loop tracer at an applied field of 10 Oe as a function of annealing temperature for amorphous $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ (x=0,1,2) alloys.

On the other hand, although B_{10} for the Cu-containing amorphous alloys in Fig.4 is as low as 0.92~1.00 T in the T_a range below 673 K, it start to increase sharply at 723 K, corresponding to the temperature that the amorphous phase changes into the bcc one. With further increasing T_a, it increases gradually and then reaches 1.45 T at 873 K.

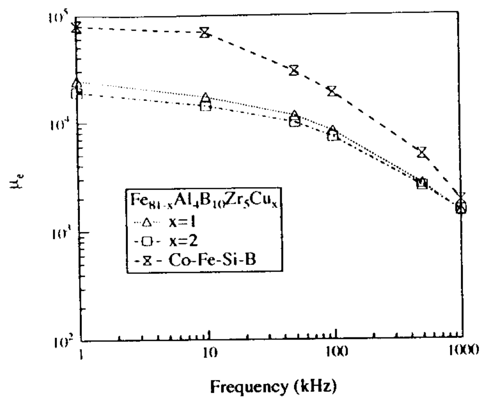

Fig.6 Changes in the μ_e values as a function of frequency for bcc $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ (x=0,1,2) alloys.

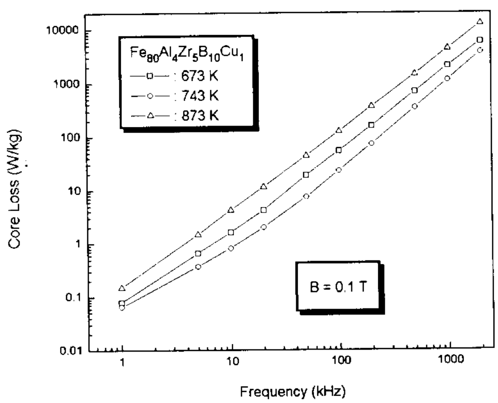

Fig.7 Changes in the W_L values as a function of frequency for the $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloy annealed at various temperatures.

The frequency dependence of μ_e for the bcc $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ (x=1, 2) alloys with the highest μ_e values at 1 kHz is exemplified in Fig. 6. The μ_e values decrease gradually to about 8000 at 100 kHz and significantly in the higher frequency range. In comparison with that for the amorphous Co-Fe-Si-B alloy, no district degradation is seen in the range of 1 to 1000 kHz and, furthermore, they keep the high level of 1500 at 1000 kHz.

Figure 7 shows the changes in core loss (W_L) as a function of frequency for the variously annealed $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloy. W_L has a similar tendency against T_a and shows minimum at T_a=743 K. The optimum T_a leading to the minimum value of W_L agrees with that at which the maximum μ_e is obtained in Fig. 3 (a).

Table 1 Magnetic properties (B_s, μ_e, H_c), structure and T_c for Fe-Al-B-Zr-Cu and other soft magnetic alloys.

alloy	struct.	B_s (T)	μ_e at 1 kHz	T_c (K)	W_L** (W/kg)
$Fe_{80}Al_4B_{10}Zr_5Cu_1$	bcc	1.42*	25000	1040	95.8
$Fe_{79}Al_4B_{10}Zr_5Cu_2$	bcc	1.37*	20000	1040	-
$Fe_{87}Zr_7B_5Cu_1$ [3]	bcc	1.55	20000	-	-
Co-Fe-Si-B [7]	amor.	0.53	80000	483	-
$Fe_{78}Si_9B_{13}$ [7]	amor.	1.55	9000	688	168
$Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$ [7]	bcc	1.24	90000	873	-
Fe-6.5 wt.% Si [8]	bcc	1.80	2400	973	1200

* H_{ex}=10 Oe, ** B=0.2T at 100 kHZ

The magnetic properties for the bcc Fe-Al-B-Zr-Cu alloys annealed at optimum annealing temperature are summarized in Table 1, where the data of some soft magnetic materials are also shown for comparison. The B_s and T_c values of the bcc Fe-Al-B-Zr-Cu alloys are considerably higher than those of bcc Fe-Si-B-Nb-B and Co-based alloys. The μ_e and W_L values at 1 kHz are also much higher than those of bcc Fe-6.5 wt.% Si and amorphous Fe-based alloys.

Ⅳ. CONCLUSIONS

With the aim of developing a new soft magnetic material combined with a high saturation magnetization, the magnetic properties and structures of $Fe_{81-x}Al_4B_{10}Zr_5Cu_x$ (x=0, 1, 2 at%) alloys annealed from amorphous state are investigated. By the addition of 1~2 Cu, the temperature range formed a single bcc phase expands largely more than 200 K. Furthermore, the addition of Cu results in the formation of bcc phase with a nanoscale grain size of 5~8 nm.

Especially, the bcc nanocrystalline $Fe_{80}Al_4B_{10}Zr_5Cu_1$ alloy produced by optimum annealing treatment exhibit high T_c, μ_e, B_{10} and W_L of 1040 K, 25000 at 1 kHz, 1.42 T and 95.8 W/kg at 100 kHz and 0.2 T, respectively.

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