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MAGNETIC PROPERTIES OF MELT-SPUN $Fe_{86-x}Al_4B_{10}Zr_x$ AMORPHOUS ALLOYS

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With the object of developing a new magnetic core materials for high frequency use, the crystallization behaviors and the soft magnetic properties of amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ ($5 \leq x \leq 10$ at%) alloys subjected to annealing treatment at wide temperature range were investigated. For optimally annealed $Fe_{86-x}Al_4B_{10}Zr_x$ alloys in amorphous state, rather good soft magnetic properties of μ_e=17000~25000, H_c=20~30 mOe and $B_{10} \geq 0.6$ T are obtained. However, as the alloys crystallize, the soft magnetic properties are largely deteriorated, which is attributed principally to the narrow temperature gap between T_{x1} and T_{x2}, which allows the nearly co-precipitation of bcc phase and Fe-B compounds in incipient crystallization stage.

I. INTRODUCTION

The soft ferromagnetic amorphous alloys have been intensively studied because of their application to electrotechnology, either for power transformers or high-frequency magnetic cores. The Fe-Si-B system in particular has been the object of major research and the best composition range [1] and the additive effects of transition metal elements [2] are well established.

In this case of Fe-Al based alloys, however, less attention has been given to core materials, in spite of the fact that their electrical and magnetic properties [3] are similar to those of Fe-Si based alloys [4] except their magnetostriction [5].

Recently, we have carried out a systematic study on the development of new high-frequency magnetic materials in Fe-Al-B based systems. This paper presents the structural changes and magnetic properties of amorphous Fe-Al-B-Zr alloys under various annealing temperatures.

II. EXPERIMENTAL PROCEDURES

Amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ (X=5, 7.5, 10 at%) alloys, 1~2 mm wide and 17~19 mm 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 under magnetic 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 K to 973 K for 1 h in the vacuum state of about 10^{-3} Torr. The structures of the annealed alloys were estimated by an X-ray diffraction (XRD) using Cu-K_α radiation. The effective permeabilities (μ_e) were measured by an impedance analyzer under an applied field of 10 mOe. The dc magnetic properties of the magnetic induction (B_{10}) and coercive force (H_c) were measured by a hysteresis loop tracer under an applied field of 10 and 0.1 Oe, respectively.

III. RESULTS AND DISCUSSION

In Fig. 1, the DTA curves of the amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ (X=5, 7.5, 10) alloys are shown. Three exothermic peaks appear in each DTA curve, indicating that the amorphous alloys crystallize through three distinct stages. The structural changes corresponding to the three exothermic peaks were examined by XRD for the samples heated at the heating rate of 10 K/min which is the same as that for the DTA measurement: the first-, second- and third-stage peaks, T_{x1}, T_{x2} and T_{x3}, were confirmed to be due to the structural changes from amorphous to mainly bcc, Fe_3B and Fe_3Zr phases respectively. T_{x1} increases monotonically with increasing Zr content, indicating that an addition of Zr into Fe-Al-B system affects largely the stability of the amorphous state. It is noteworthy that above X=7.5 the temperature range between T_{x1} and T_{x2} expands over 50 K. This result indicates that the formation of single bcc phase is possible by proper annealing treatment.

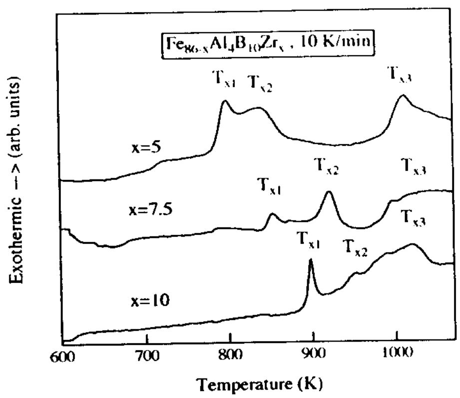

Fig. 1 DTA curves of amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ (x=5, 7.5, 10) alloys heated at the rate of 10 K/min.

Figure 2 shows the XRD patterns of the $Fe_{78.5}Al_4B_{10}Zr_{7.5}$ alloy heated up to 873, 973 and 1073 K at the heating rate of 10 K/min. The sample heated up to 873 K above T_{x1} shows diffraction peaks of only a bcc structure, while that heated up to 973 K above T_{x2} shows the mixed structure composed of bcc and Fe_3B phases. The lattice spacing d_{110} of $(110)_{bcc}$ for $Fe_{78.5}Al_4B_{10}Zr_{7.5}$ alloy heated up to 873 K is 0.2034 nm and decreases gradually to that (0.2027 nm) of pure α-Fe with increasing heating-temperature to 1073 K. This indicates that the solute elements in the bcc phase diffuse out with increasing heating-temperature through the decomposition of the non-equilibrium bcc phase, which are supersaturated with the solute elements.

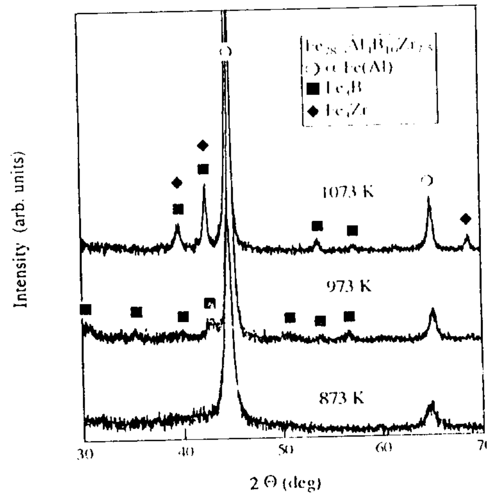

Fig. 2 X-ray diffraction patterns of amorphous $Fe_{78.5}Al_4B_{10}Zr_{7.5}$ alloys heated at the rate of 10 K/min.

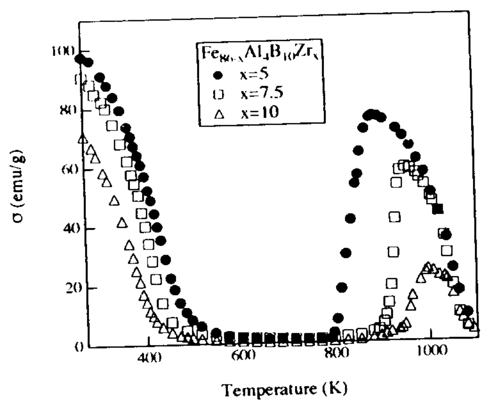

Fig. 3 Thermomagnetic curves for amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ (x=5, 7.5, 10) alloys. The heating rate is 10 K/min and the applied magnetic field is 10 kOe.

The temperature dependence of saturation magnetization (σ_T) of the amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ alloys is shown in Fig. 3. The Curie point, T_c, and σ_T decrease with increasing Zr content. The inflection of magnetization around 800 K for the alloy with X=5 is related to the formation of bcc phase from amorphous state and its temperature increases with Zr content, corresponding well to the DTA results of Fig. 1. On the other hand, the inflection temperature around 1050 K is constant irrespective to Zr content and agrees with the T_c of α-Fe phase. This result indicates that in this temperature range the B and Zr elements supersaturated into bcc phase are almost precipitated as the Fe-B and Fe-Zr compounds.

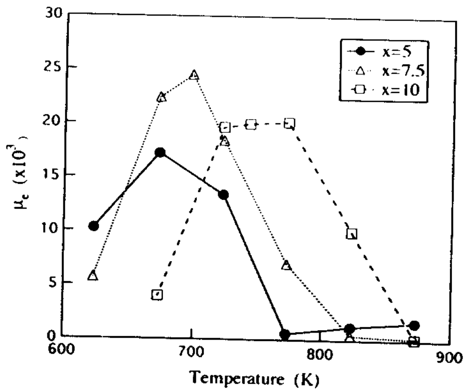

Fig. 4 Effective permeabilities, μ_e, at 1 kHz as a function of annealing temperature for amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ alloys.

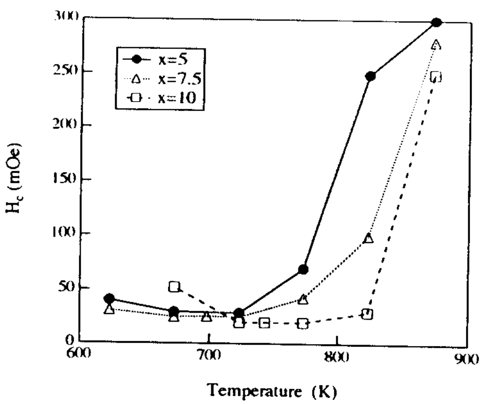

Fig. 5 Coercive force, H_c, calculated by a hysteresis loop tracer at an applied field of 0.1 Oe as a function of annealing temperature for amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ alloys.

The variations of μ_e at 1 kHz as a function of annealing temperature for the amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ alloys are shown in Fig. 4. The highest μ_e values are located below T_{x1} of each alloys. The optimum-annealing temperature, T_{opt}, which represents the maximum value of μ_e, increases with Zr content, corresponding to the increase of T_{x1}. This result seems to be due to the temperature difference occurring to the structural relaxation for amorphous alloys with different Zr content. However, above T_{x1}, the μ_e values of the alloy with x=5 decrease considerably, due to the nearly simultaneous formation of the bcc and Fe_3B phases. On the other hand, although the temperature gap between T_{x1} and T_{x2} for the alloy with x=7.5 expands largely over 50 K, the μ_e value around T_{x1} is much lower than that of below T_{x1}. The rapid decrease in μ_e associated with the formation of bcc phase is explained by coarsening of the grain size, leading to the increase in apparent anisotropy. In fact, the grain size of bcc phase in the $Fe_{78.5}Al_4B_{10}Zr_{7.5}$ alloy annealed for 1 h at 873 K, which is evaluated from the half width of the $(110)_{bcc}$ diffraction peak using Scherrer's equation, is as large as 35 nm and increases rapidly with further increasing temperature. The H_c values for the amorphous alloys annealed below T_{x1} in Fig. 5 sustain as low as 20~30 mOe, while they increase to hundreds of mOe as the crystallization proceeds.

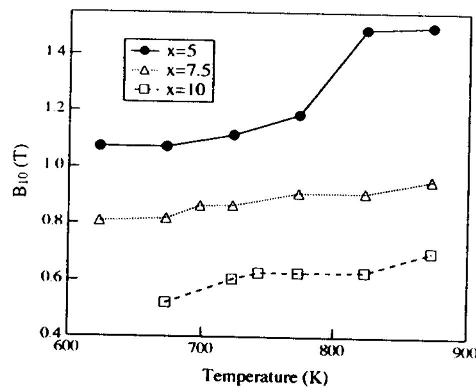

Fig. 6 Magnetic induction, B_{10}, calculated by a hysteresis loop tracer at an applied field of 0.1 Oe as a function of annealing temperature for amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ alloys.

As shown in Fig. 6, B_{10} for the amorphous alloys annealed below T_{x1} shows a tendency to increase a little with increasing temperature. However, it is by far lower than those annealed above T_{x1}, due to the Invar effect in Fe-Zr amorphous alloys [6]. The rapid increase in B_{10} and H_c for $Fe_{81}Al_4B_{10}Zr_5$ alloy annealed in the temperature range of 823~873 K is associated with the structural change from amorphous to the mixed structure composed of bcc and Fe_3B phase, having a larger magneto-crystalline anisotropy [7]. As a result, the good soft magnetic properties of optimally annealed amorphous alloys are obtained below T_{x1} and the values are summarized in Table 1.

Table 1 The magnetic properties (μ_e, H_c and B_{10}) for amorphous $Fe_{86-x}Al_4B_{10}Zr_x$ alloys obtained by optimum annealing for 1 h. The T_c, T_{opt} and T_{x1} of their amorphous alloys are also shown for reference.

Alloy	μ_e (1 kHz)	H_c (mOe)	B_{10} (T)	T_c (K)	T_{opt} (K)	T_{x1} (K)
$Fe_{81}Al_4B_{10}Zr_5$	17000	30	1.1	473	673	797
$Fe_{78.5}Al_4B_{10}Zr_{7.5}$	25000	25	0.8	443	698	853
$Fe_{76}Al_4B_{10}Zr_{10}$	20000	20	0.6	413	743	899

Ⅳ. CONCLUSIONS

The soft magnetic properties and structures of $Fe_{86-x}Al_4B_{10}Zr_x$ (x=5, 7.5, 10) alloys annealed from amorphous state were investigated. The amorphous alloys crystallize through three distinct stages: that is, the crystallization behaviors of the first-, second- and third-stage are confirmed to be due to the structural changes from amorphous to mainly bcc, Fe_3B and Fe_3Zr phases, respectively. The first-stage crystallization temperature increases monotonically with increasing Zr content.

For $Fe_{86-x}Al_4B_{10}Zr_x$ amorphous alloys annealed at various temperature, the good soft magnetic properties are obtained below the onset temperature of the first-stage crystallization. As the alloys crystallize, they are deteriorated considerably, due to the nearly co-precipitation of bcc and Fe-B phases. For the optimally annealed amorphous alloys, the obtained magnetic properties are as follows; μ_e=17000~25000 at 1 kHz, H_c=20~30 and B_{10}=0.6~1.1 T. However, because T_c and B_{10} in the amorphous state of these amorphous alloys are much lower than those in the crystallized state, it is needed to improve them by the proper addition of alloying elements into Fe-Al-B-Zr alloy system and crystallization treatment.

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