Compositional Effect on Soft Magnetic Properties of Co-Fe-Si-B Amorphous Alloys with Zero Magnetostriction

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The compositional effect on magnetic properties of Co-Fe-Si-B amorphous alloys was examined with the aim of finding an optimum composition for a magnetic head material. The lowest coercivity (H_c) and the highest permeability at 0.3 kHz ($\mu_{0.3}$) in the as-quenched state were obtained at 0.94Co/(Co+Fe) and in the metalloid range of 24 to 25%(Si+B) and 0.6Si/(Si+B). The composition range is in agreement with that in which the highest glass-forming capacity is obtained and hence the good soft magnetic properties in the limited composition range are presumably due to a high degree of amorphicity and structural homogeneity. The permeability at 100 kHz (μ_{100}) shows a maximum value in the range of 27%(Si+B) and 0.65 to 0.70Si/(Si+B) and the difference of the optimum compositions between $\mu_{0.3}$ and μ_{100} is due to the necessity of high electrical resistivity for μ_{100} . The annealing results in the change of the optimum composition to 0.6Si/(Si+B) for μ_{100} and 0.5Si/(Si+B) for H_c . The annealing-induced change is interpreted from the difference in the compositional effect on the homogeneity and thermal stability of the amorphous structure. Thus, the combination of high magnetization values of 98×10^{-6} to 113×10^{-6} wb.m.kg $^{-1}$ and high permeability at 0.3 kHz reaching 19.21×10^4 in the annealed state is achieved at 0.6Si/(Si+B) where the amorphous structure has high thermal stability and hence the metalloid composition is concluded to be optimum for an amorphous magnetic head material.

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I. Introduction

Since ferromagnetic properties of melt-spun Co-Fe-Si-B amorphous alloys were reported for the first time in 1974⁽¹⁾, a number of studies on the magnetic properties of (Fe, Co, Ni)-Si-B amorphous alloys have been carried out⁽²⁾⁽³⁾, leading to the finding of excellent soft ferromagnetic properties for a Co-Fe-Si-B amorphous alloy with zero magnetostriction. The zero magnetostrictive Co-Fe-Si-B amorphous alloy has attracted great attention in a practical use as a magnetic head material⁽⁴⁾⁻⁽⁷⁾ because of high electrical resistivity and high hardness as well as good soft magnetic properties. Although various kinds of characteristics are required for magnetic head materials, the most important characteristics are due to the simultaneous satisfaction of high saturation magnetization and high permeability⁽⁸⁾. However, little has been known about the compositional effect on the magnetic properties of zero magnetostrictive Co-Fe-Si-B amorphous alloys. This paper is intended to examine the compositional dependence of magnetic properties (coercivity, permeability and saturation magnetization) in rapidly solidified and annealed states for zero magnetostrictive Co-Fe-Si-B amorphous alloys and to find an optimum alloy composition at which high saturation magnetization and good soft magnetic properties are obtained simultaneously.

II. Experimental Procedure

Co-Fe-Si-B ingots in the composition range of 0.9 to 1.0Co/(Co+Fe), 22 to 27 at% (Si+B) and 0.2 to 0.7Si/(Si+B) were produced by induction melting a mixture of pure cobalt (99.9 mass%) and iron (99.9 mass%) metals, silicon (99.999 mass%) and boron (99.8 mass%) in an alumina crucible in an argon atmosphere, followed by casting into a copper mold with an inner diameter of 30 mm. From the ingots, rapidly solidified ribbons with a thickness ranging from 45 to 50 μ m and a width of 24 mm were produced in air by a single roller melt spinning method. The amorphicity of the ribbons was examined by X-ray diffractometry using Co $K\alpha$ radiation and transmission electron microscopy (TEM). The permeability (μ_e) was measured at a frequency of 0.3 or 100 kHz under an applied field of 0.239 or 0.796 A/m by the transe method for the ring samples with an inner and an outer diameter of 6 and 10 mm, respectively, produced by either chemical etching or mechanical punching the wide amorphous ribbons. Measurement of coercivity (H_c) was made for the same ring samples by using a B-H loop tracer. Saturation magnetization (σ_s) was measured under an applied field of 39.8 kA/m (500 Oe) at room temperature with a vibrating sample magnetometer. Magnetostriction was evaluated by measuring the elongation of sample under an applied field of 79.6 kA/m with a strain gauge meter. Electrical resistivity was measured

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at room temperature by the DC four-probe technique. Annealing treatment was made for 600 s at different temperatures in an argon atmosphere followed by water quenching. Crystallization temperature and heat of crystallization were measured at a scanning rate of 0.33 K/s by differential scanning calorimetry (DSC) and the onset temperature of exothermic peak due to crystallization was defined as crystallization temperature (T_x).

III. Results and Discussion

1. Magnetic properties in a rapidly solidified state

Figure 1 shows the changes in μ_e at 0.3 kHz and 0.239 or 0.796 A/m ($\mu_{0.3}$) and H_c as a function of concentration ratio of Co to Co+Fe for Co-Fe-Si-B amorphous alloys. The compositional dependences of $\mu_{0.3}$ and H_c are considerably larger for the alloys with higher ratios of Si/(Si+B) than for the alloys with lower Si concentrations and the former alloys exhibit an increase of $\mu_{0.3}$ and a decrease of H_c at a limited composition of 0.940Co/(Co+Fe). Similarly, the latter alloys also tend to have high $\mu_{0.3}$ and low H_c in the Co/(Co+Fe) range of 0.940 to 0.945. It is known that magnetostriction affects significantly soft magnetic properties and the approach of magnetostriction to zero value is an essential factor to achieve good soft magnetic properties of high μ_e and low H_c . Considering previous data⁽⁹⁾⁽¹⁰⁾ that zero magnetostriction is obtained for $(\text{Co}_{0.96}\text{Fe}_{0.04})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys, the good soft magnetic properties in the vicinity of 0.94Co/(Co+Fe) seem to result from nearly zero magnetostriction. The systematic compositional dependence of $\mu_{0.3}$ suggests that the concentration ratio of Co to Co+Fe exhibiting zero magnetostriction remains almost constant of 0.94 in a rather wide metalloid composition range of 23 to 27 at% (Si+B) and 0.20 to 0.64Si/(Si+B), in agreement with the previous data⁽¹¹⁾⁽¹²⁾ on the effect of metalloid composition on the optimum ratio of Co/(Co+Fe) exhibiting zero magnetostriction for Co-Fe-Si-B amorphous alloys. On the basis of the compositional dependences of $\mu_{0.3}$ and H_c shown in Fig. 1, a subsequent study of finding an optimum metalloid composition for the production of an excellent soft magnetic material was performed in the series of $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ alloys with a constant Co/(Co+Fe) value of 0.94.

Figures 2 and 3 show the changes in H_c and $\mu_{0.3}$ for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys with metalloid composition. As is evident from the contour lines shown in Fig. 2, the H_c value is the lowest (0.96 A/m) at 25%(Si+B) and 0.60Si/(Si+B) and increases with a deviation from the composition. The increase in H_c is more remarkable in the higher Si/(Si+B) range. Similarly, the $\mu_{0.3}$ also shows the highest value of 18700 at 24%(Si+B) and 0.6Si/(Si+B) and decreases with a deviation from the composition. It is notable that the combined characteristics of low H_c and high $\mu_{0.3}$ are obtained in the vicinity of 24 to 25%(Si+B) and 0.6Si/(Si+B). In order to clarify the reason for the com-

Fig. 1 Changes in the permeability ($\mu_{0.3}$) at 0.3 kHz and coercivity (H_c) as a function of Co/(Co+Fe) for as-quenched Co-Fe-Si-B amorphous alloys.

Fig. 2 Change in the coercivity (H_c) as a function of Si+B and Si/(Si+B) for as-quenched $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

positional effects of H_c and $\mu_{0.3}$ for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys, the measurement of magnetostriction was made and the magnetostriction was confirmed to be of the order 10^{-7} for almost all the quaternary alloys examined in the present study. However, the compositional dependence of the magnetostriction in the very

Fig. 3 Change in the permeability at 0.3 kHz and 0.796 A/m ($\mu_{0.3}$) as a function of Si+B and Si/(Si+B) for as-quenched $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

small values of 10^{-7} could not be clarified because of significant scattering resulting from the very small magnetostriction values. Judging from the very small magnetostriction, which can be regarded as a nearly zero magnetostriction, in the Co-Fe-Si-B amorphous alloys with a constant Co/(Co+Fe) value of 0.94, the compositional dependences of $\mu_{0.3}$ and H_c shown in Figs. 2 and 3 seem to reflect the degrees of amorphicity and microscopic structural homogeneity in the rapidly solidified amorphous state.

Here, it appears important to point out that the metalloid composition of 24 to 25% (Si+B) and 0.6Si/(Si+B) agrees well with that⁽¹³⁾ having the largest glass-forming capacity in Co-Si-B ternary alloys. As shown in Fig. 4, the large critical ribbon thickness (t_c) for the formation of an amorphous phase exceeding 145 μm has been obtained in the range of 25 to 27% (Si+B) and 0.60Si/(Si+B)⁽¹³⁾. The total amount of Si and B in the amorphous alloy exhibiting the largest t_c value is slightly larger than that (24 to 25%) for the amorphous alloy with the best soft magnetic properties. It has previously been reported⁽¹³⁾ that the largest t_c value in Co-Si-B and Fe-Si-B ternary systems is obtained at $\text{Co}_{72.5}\text{Si}_{12.5}\text{B}_{15}$ and $\text{Fe}_{77.5}\text{Si}_{10}\text{B}_{12.5}$ and the optimum metalloid content leading to the largest glass-forming capacity is considerably less for Co-Si-B alloys. Accordingly, the slight difference in the metalloid compositions at which the largest t_c value and the best soft magnetic properties are obtained seems to result from the replacement of Co by 6%Fe. It is generally thought that the larger the glass-forming capacity the higher are the noncrystallinity and homogeneity of the amorphous phase in the melt-spun ribbon with a constant thickness. Accordingly, the agreement of the metalloid composition between the best soft magnetic

Fig. 4 Change in the critical ribbon thickness (t_c) for formation of an amorphous phase as a function of Si+B and Si/(Si+B) for as-quenched Co-Si-B amorphous alloys.

properties and the largest glass-forming capacity allows us to presume that the best soft magnetic properties in the range of 24 to 25% (Si+B) and 0.6Si/(Si+B) are due to the reduction of magnetic anisotropy resulting from a high degree of noncrystallinity and homogeneity in the amorphous phase caused by the largest glass-forming capacity. However, the details in the variations of H_c and $\mu_{0.3}$ with metalloid composition do not always agree with that for the t_c . The difference in the variations of t_c and H_c or $\mu_{0.3}$ with metalloid composition appears to result from the differences in metallic component and liquid-quenching condition.

Figure 5 shows the change in μ_e at 100 kHz (μ_{100}) for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys with metalloid composition. The μ_{100} value at the high frequency was very sensitive to sample thickness and hence the data shown in Fig. 5 were obtained for the amorphous ribbons with a nearly constant thickness ranging from 45 to 50 μm . As the metalloid content increases, the μ_{100} increases and shows high values above 6000 for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{73}\text{Si}_{27-x}\text{B}_x$ with Si/(Si+B) ratios ranging from 0.65 to 0.70. In comparison with the $\mu_{0.3}$ values shown in Fig. 3, the increase of frequency from 0.3 to 100 kHz brings about the shift of the composition, at which the largest μ_e value is obtained, to the higher composition range. This shift is explained by taking the compositional dependence of electrical resistivity into consideration. As shown in Fig. 6, the electrical resistivity at room temperature (ρ_{RT}) increases significantly from about 110 to $150 \times 10^{-8} \Omega\text{m}$ with an increase of total metalloid content and the ratio of Si/(Si+B). Although the $\mu_{0.3}$ is independent of ρ_{RT} , the μ_{100} at the high frequency is usually dependent on ρ_{RT} and tends to increase with increasing ρ_{RT} because

Fig. 5 Change in the permeability at 100 kHz and 0.796 A/m (μ_{100}) as a function of Si+B and Si/(Si+B) for as-quenched $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

Fig. 6 Change in the electrical resistivity at room temperature (ρ_{RT}) as a function of Si+B and Si/(Si+B) for as-quenched $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

of a decrease of eddy current loss. Accordingly, the achievement of the largest μ_{100} in the higher metalloid range is presumably due to the overlap of the compositional dependences of μ_e and ρ_{RT} .

2. Magnetic properties in an annealed state

(1) Change in magnetic properties with annealing temperature and alloy composition

It is generally known that as-quenched amorphous alloys do not exhibit the best soft magnetic properties

because of the hindrance of the movement of magnetic walls due to quenching-induced internal stress. The annealing treatment consisting of heating in a temperature range between Curie temperature (T_c) and the onset temperature of crystallization (T_x) and subsequent water quenching has been reported to bring about an improvement of soft magnetic properties⁽¹⁴⁾. A similar heating treatment was made for the $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys with the aim of improving soft magnetic properties. Figure 7 shows the change of the $\mu_{0.3}$ as a function of annealing temperature (T_a) for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{75}(\text{Si}_x\text{B}_y)_{25}$ amorphous alloys having T_x values considerably higher than T_c . The heating time at each T_a was fixed to be 600 s. When T_a is lower than T_c , the $\mu_{0.3}$ decreases as compared with that of the as-quenched samples. However, the increase of T_a to the temperature range between T_c and T_x gives rise to $\mu_{0.3}$ values higher than those of the as-quenched samples. The highest $\mu_{0.3}$ value in an annealed state is 14.11×10^4 for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{75}\text{Si}_{15}\text{B}_{10}$ annealed at 700 K. It is thus notable that the alloy with the highest $\mu_{0.3}$ in an as-quenched state exhibits the largest $\mu_{0.3}$ even in the annealed state. The data shown in Fig. 7 also indicate that the optimum T_a leading to the highest $\mu_{0.3}$ value tends to decrease with an increase of the ratio of Si/(Si+B).

An optimum metallic composition in an annealed state was also investigated for Co-Fe-Si-B amorphous alloys with different metalloid compositions. Figure 8 shows the changes in $\mu_{0.3}$ and H_c as a function of Co/(Co+Fe) for Co-Fe-Si-B amorphous alloys annealed at temperatures between T_c and T_x . The combined characteristics of the highest μ_e and the lowest H_c are obtained at 0.94Co/(Co+Fe), in agreement with that for the as-quenched samples. It is therefore concluded that there is no appreciable difference in the metallic composition

Fig. 7 Change in $\mu_{0.3}$ for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{75}\text{Si}_x\text{B}_{25-x}$ amorphous alloys with annealing temperature (T_a).

Fig. 11 Change in crystallization temperature (T_x) as a function of Si+B and Si/(Si+B) for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

Fig. 12 Change in maximum permeability μ_{100} at 0.796 A/m as a function of Si+B and Si/(Si+B) for annealed $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

ed state to 0.5Si/(Si+B) in the annealed state is also due to the higher T_x value in the vicinity of 0.5Si/(Si+B) as compared with that at 0.6Si/(Si+B).

In addition, Fig. 12 shows the compositional dependence of the maximum permeability at 100 kHz (μ_{100}) for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys subjected to an optimum annealing treatment. The μ_{100} shows a maximum value of 13700 in the vicinity of 26%(Si+B) and 0.6Si/(Si+B) and hence the annealing is said to result in the shift of the Si/(Si+B) ratio exhibiting the highest μ_{100} from 0.67 to 0.63. The shift is also interpreted from the result shown in Fig. 11 that T_x increases with a decrease of Si/(Si+B) from 0.7 to 0.6.

(2) Influence of metalloid composition on the saturation magnetization (σ_s) and permeability (μ_c)

Figure 13 shows the metalloid dependence of σ_s for as-quenched $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys. The σ_s decreases gradually from 105 to 70 emu/g with an increase of the total metalloid content and the concentration ratio of Si to Si+B. The gradual decrease of σ_s is thought to originate from the hybridization among 3d-electrons in Co and Fe, 3p-electrons in Si and 2p-electrons in B. On the basis of the data shown in Figs. 10, 12 and 13 on the changes in the $\mu_{0.3}$, μ_{100} and σ_s with metalloid composition for annealed Co-Fe-Si-B amorphous alloys, the relations between σ_s and $\mu_{0.3}$ or μ_{100} are shown in Figs. 14 and 15, respectively. The combined

Fig. 13 Change in saturation magnetization (σ_s) as a function of Si+B and Si/(Si+B) for as-quenched $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

Fig. 14 Relation between σ_s and $\mu_{0.3}$ for annealed $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

Fig. 15 Relation between σ_s and μ_{100} for annealed $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys.

properties of high σ_s and high μ_e are obtained at both frequencies of 0.3 and 100 kHz for the $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys with 0.6Si/(Si+B). It is therefore concluded that the best soft magnetic properties with combined characteristics of high σ_s , low H_c and high μ_e at 0.3 and 100 kHz are obtained for $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys with 24 to 27%(Si+B) and 0.6Si/(Si+B). Here, it is important to describe that the optimum metalloid composition to obtain good soft magnetic properties is completely different from that (Si/(Si+B)=0.2)⁽⁷⁾ which has been practically used by other groups as soft magnetic materials in Co-Fe-Si-B system. The optimum of the alloy composition found in the present study is also supported from the recent trend in which the practical use of magnetic heads made from the amorphous alloy with 0.6Si/(Si+B) has increased significantly, in good contrast to the tendency for the amorphous magnetic head with a metalloid composition of Si/(Si+B)=0.2 to reduce the amount of application⁽⁷⁾.

IV. Summary

In order to search an optimum alloy composition exhibiting excellent soft magnetic properties which are required for an amorphous magnetic head material, the compositional dependences of σ_s , H_c and μ_e were examined for Co-Fe-Si-B amorphous alloys ranging from 0.9 to 1.0Co/(Co+Fe), 22 to 27%(Si+B) and 0.2 to

0.7Si/(Si+B). The results obtained are summarized as follows:

(1) Low H_c and high $\mu_{0.3}$ in a rapidly solidified state were obtained for Co-Fe-Si-B amorphous alloys with 0.94Co/(Co+Fe). The lowest H_c and the highest $\mu_{0.3}$ for as-quenched $(\text{Co}_{0.94}\text{Fe}_{0.06})_{100-x-y}\text{Si}_x\text{B}_y$ amorphous alloys were obtained in the ranges of 25 to 26%(Si+B) and 0.6Si/(Si+B) and 24 to 25%(Si+B) and 0.6Si/(Si+B), respectively. The glass-forming capacity in Co-Si-B ternary alloys is the largest in the vicinity of 27%(Si+B) and 0.6Si/(Si+B), in agreement with that at which the lowest H_c and the highest $\mu_{0.3}$ are obtained. The agreement allows us to presume that the good soft magnetic properties in the limited composition range are due to a high degree of noncrystallinity and homogeneity in the as-quenched amorphous structure. In addition, the μ_{100} shows a maximum value in the high metalloid range of 27%(Si+B) and 0.65 to 0.70Si/(Si+B) because of the necessity of high electrical resistivity.

(2) The optimum ratios of Si/(Si+B) for μ_e and Co/(Co+Fe) for μ_e and H_c remain unchanged even after optimum annealing, though the optimum Si/(Si+B) ratio for H_c decreases to 0.5. The compositional effect on the soft magnetic properties for the annealed samples is explained by taking the compositional dependences of noncrystallinity, homogeneity and thermal stability of the amorphous alloys into consideration.

(3) The combined properties of high σ_s , low H_c and high μ_e are obtained in the range of 24 to 27%(Si+B) and 0.6Si/(Si+B) and hence the metalloid composition is concluded to be applicable to an amorphous magnetic head material.

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