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FePBNbCr soft-magnetic glassy alloys with low loss characteristics for inductor cores

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ABSTRACT

Fe-based glassy alloys with both high saturation magnetization and low magnetic anisotropy have attracted interest in recent study on Mr. Inoue, 2004, and we have succeeded in developing novel glassy $Fe_{97-x-y}P_xB_yNb_2Cr_1$ (x=5-13, y=7-15) alloys for an inductor material. The glassy alloy series of $Fe_{97-x-y}P_xB_yNb_2Cr_1$ (x=5-13, y=7-15) have high glass-forming ability with the large critical thickness of 110–150 µm and low coercivity of 2.5–3.1 A/m caused by the structural homogeneity. The $Fe_{77}P_7B_{13}Nb_2Cr_1$ glassy alloy exhibits the largest critical thickness of 150 µm related to the wide super cooled liquid region and the high saturation magnetic flux density (B_s) of 1.3 T, both of which are higher than those of the conventional amorphous $Fe_{75}Si_{10}B_{12}Cr_3$ alloy. Hence, Fe-P-B-Nb-Cr powder/resin composite core has much lower core loss of 650 kW/m³ which is approximately 1/3 lower than the conventional amorphous Fe-Si-B-Cr powder/resin composite core that are annealed at 623 K. In addition, the optimum annealing temperature of 623 K for these glassy alloys was lower than that (723 K) for the ordinary Fe-Si-B-Cr amorphous alloy, which is a significant advantage for the efficiency in mass production of inductor core using soft-magnetic glassy alloy powder.

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1. Introduction

Recently, the achievement of a high level of power efficiency in the power supply circuit has become a high-priority issue. The inductor in the power supply is required to be capable of dealing satisfactorily with the high-current supply and to improve the power loss characteristic. In order to improve the power supply characteristics, soft-magnetic material for inductor core must strike a balance between high Bs and low core loss characteristic. But ordinary Ni-Zn ferrite and Mn-Zn ferrite core can not use the inductor core material for large current power supply because of the B_s is too lower compare with other soft-magnetic material consisting of metallic alloys. On the other hand, ordinary soft-magnetic materials consisting of metallic alloys have the subject owing to high core loss that is attributed to large magnetic anisotropy. Therefore we focused on the glassy metal alloy with both high saturation magnetization and low magnetic anisotropy, and developed Fe-based glassy metal alloys with a chemical composition Fe₇₇P₇B₁₃Nb₃ for inductor core material [1–3]. However the glassy metal alloy Fe77P7B13Nb3 has not acquired high corrosion resistance and demonstrated a performance in practical use

well vet. Corrosion resistance is important factor in practical use as well as the soft-magnetic properties for inductor core material. Thus we improved the corrosion resistance owing to adding optimum content of Cr to Fe77P7B13Nb3 and have succeeded in developing novel glassy metal alloys with chemical compositions of $Fe_{97-x-v}P_xB_vNb_2Cr_1$ for a new class material of inductor core. Optimum content of Cr in $Fe_{97-x-y}P_xB_yNb_2Cr_1$ was decided to the 1 at% that was able to control the decrease of B_s which is accompanied by added Cr and the maximum thickness for glass formation (T_{max}) , known to be one of the indicators for GFA [4], was improved also up to 150 μ m owing to the change in total amount of Fe–Nb₂–Cr₁ from 80 at% to 77 at%. We report on a novel glassy metal alloys $Fe_{97-x-v}P_xB_vNb_2Cr_1$ with both high B_s of 1.3 T and low core loss of 650 kW/m³ compare with ordinary amorphous alloy Fe–Si–B–Cr. An additional remark which should be made here is that optimum annealing temperature is lower compared with the typical Fe₇₅Si₁₀B₁₂Cr₃ amorphous alloy and can acquire high GFA even if using the raw material of low cost ferroalloys.

2. Experimental

 $Fe_{97-x-y}P_xB_yNb_2Cr_1$ and $Fe_{75}Si_{10}B_{12}Cr_3$ were prepared by induction melting the mixture of pre-melted Fe–P, Fe–B, Fe–Nb, Fe–Cr, Fe–Si and pure metal of Fe in an argon atmosphere. The alloy compositions represent nominal atomic percent. The glassy metal alloys $Fe_{97-x-y}P_xB_yNb_2Cr_1$ and amorphous alloy $Fe_{75}Si_{10}B_{12}Cr_3$ were produced in a ribbon form having a width of about 1 mm and thickness of about 25–150 μ m by a single roller melt-spinning method in an argon atmosphere. As-

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Fig. 1. The change of as-quenched structure with the thickness for $Fe_{77}P_7B_{13}Nb_2Cr_1$ and $Fe_{75}Si_{10}B_{12}Cr_3$ ribbons with a width of 1 mm.

melt span structures were examined by X-ray diffraction (XRD) with Mo K α line. Curie temperature (T_c) was estimated by differential thermal analysis (DTA) at a heating rate of 0.67 K/s. The glass transition temperature (T_g) and crystallization temperature (T_x) were estimated by differential scanning calorimetry (DSC) at a heating rate of 0.67 K/s. The saturation magnetic flux density (B_s) was measured under an applied field of 400 kA/m with a vibrating sample magnetometer (VSM). The coercivity (H_c) was measured under a field of 1 kA/m with a DC B-H loop tracer after annealing of 623 K. All measurements were carried out at room temperature. The glassy metal alloys Fe_{97-X-y}P_xB_yNb₂Cr₁ and amorphous alloy Fe₇₅Si₁₀B₁₂Cr₃ powder were produced by a water atomization. The powders with an average particle size of 12 μ m were mixed with a packing density of 71–3% by molding pressure of 8 GPa after granulation. The core loss (W) was measured under a frequency of 300 kHz and a field of 50 mT by B–H loop analyzer for the ring core shape after annealing of 423–723 K.

3. Results and discussion

It is well known that iron-based amorphous is one of the inductor material having low core loss characteristic. Some kind of Fe-metalloid amorphous alloys for inductor core material have been developed and been in practical use. Among them, Fe–Si–B–(Cr) system is now the major alloy widely utilized by industries.

The characteristic of amorphous forming ability for the typical $Fe_{75}Si_{10}B_{12}Cr_3$ amorphous alloy and glassy metal alloy $Fe_{77}P_7B_{13}Nb_2Cr_1$ was investigated. Fig. 1 shows the change in as-quenched structure with the thickness for $Fe_{73}Si_{10}B_5Cr_2$ and $Fe_{77}P_7B_{13}Nb_2Cr_1$ continuous ribbons with a width of 1 mm produced by the single roller melt-spinning method. The as-quenched structure was examined for freely solidified surface of the ribbon by the XRD. The amorphous alloy and glassy metal alloy ribbons have an amorphous phase up to thickness of about 50 μ m and 80 μ m, respectively. Therefore, it is noted that the glassy metal alloy $Fe_{77}P_7B_{13}Nb_2Cr_1$ have significantly high forming ability compared with ordinary amorphous alloy $Fe_{75}Si_{12}B_{10}Cr_3$. The use in glassy metal alloy probably gives a new possibility to recent subject of the soft-magnetic material for inductor in mass production.

Fig. 2 shows the compositional dependence of the maximum thickness for glass formation (T_{max}) for melt-spun Fe_{97-x-y}P_xB_yNb₂Cr₁ and Fe₇₉P₇B₁₁Nb₃ alloys without Cr. The T_{max} for Fe₇₉P₇B₁₁Nb₂Cr₁ glassy alloy was observed to decrease from 130 µm to 80 µm accompanying by addition of 1 at% Cr. However, the T_{max} of Fe–P–B–Nb–Cr system of glassy alloy can be improved up to 150 µm owing to the change in total amount of Fe–Nb₂–Cr₁ from 80 at% to 77 at%. And a glassy phase was observed in a wide compositional range and the large T_{max} of over 100 µm is observed in the enclosed range of 5–13 at% of P, 7–15 at% of B and 78–81 at% of Fe+Nb₂Cr₁, indicated with line of 100 µm. The largest T_{max} is 150 µm for Fe_{97-x-y}P_xB_yNb₂Cr₁ (x=5–8, y=12–15).



Fig. 2. The compositional dependence of the maximum thickness for glass formation (T_{max}) for melt-spun Fe_{97-x-y}P_xB_yNb₂Cr₁ alloys.

Fig. 3 shows X-ray diffraction patterns of various Fe-P-B-Nb-Cr compositions of the compositional area with large T_{max} . The XRD pattern reveals only typical halos, and no peaks corresponding to crystalline phases are visible. Thus it is to be noticed that a single amorphous phase is produced in the thickness range up to 110–150 μ m. The core loss of Fe₇₇P₇B₁₃Nb₂Cr₁ glassy alloy with a ΔT_x of 28 K, a T_{max} of 150 μ m, a H_c of 4.3 A/m, Fe₇₇P₁₁B₉Nb₂Cr₁ glassy alloy with a ΔT_x of 29 K, a T_{max} of 130 µm, a H_c of 6.9 A/m at as-quenched and Fe₇₅Si₁₀B₁₂Cr₃ amorphous alloy without glass transition, a T_{max} of 50 µm, a H_c of 28.1 A/m at as-quenched as function of annealing temperature is shown in Fig. 4. The specimens were heated under an argon atmosphere of 10 kPa at a rate of 0.05 K/s and isothermal annealed at prescribed temperature for 1.8 ks, and then cooled to 295 K by the furnace cooling. The extremely low core loss of 653–783 kW/m³ was obtained for 1.8 ks at 623 K for the glassy alloys Fe77P7B13Nb2Cr1 and Fe₇₇P₉B₁₁Nb₂Cr₁. The core loss of the ternary amorphous alloy reaches a minimum value of 1450 kW/m³ at 723 K. On the other



Diffraction angle, 20 / degree

Fig. 3. X-ray diffraction patterns of melt-spun $Fe_{97-x-y}P_xB_yNb_2Cr_1$ alloys, respectively.

Table 1

The curie temperature (T_c), the glass transition temperature (T_g), the crystallization temperature (T_x), the parameter of glass-forming ability (ΔT_x , T_{max}), magnetic properties (B_s , H_c) and the core loss (W) for Fe_{97-x-v}P_xB_vNb₂Cr₁ glassy alloys and the typical Fe-based amorphous.

Characteristic material	T_c (K)	T_g (K)	$T_{x}(\mathbf{K})$	ΔT_{x} (K)	T_{max} (mm)	B_s (T)	$H_c \left(A/m \right)$	$W(kW/m^3)$	Formation
Fe ₇₇ P ₇ B ₁₃ Nb ₂ Cr ₁	556	767	795	2\$	150	1.31	2.5	653	Glass
Fe ₇₇ PgB ₁₁ Nb ₂ Cr ₁	553	759	789	31	130	1.33	2.5	810	Glass
Fe ₇₇ B ₁₃ Cr ₁	549	754	783	29	130	1.31	3.1	783	Glass
Fe ₇₇ P ₁₃ B ₇ Nb ₂ Cr ₁	536	740	771	31	110	1.28	3.1	-	Glass
Fe ₇₆ B ₁₁ Nb ₂ Cr ₁	554	762	793	31	115	1.27	2.8	706	Glass
Fe ₇₉ P ₇ B ₁₁ Nb ₃	522	754	792	38	115	1.30	2.3	913	Glass
Fe ₇₅ Si ₁₀ B ₁₂ Cr ₃	648	-	848	-	50	1.25	14.8	1989	Ordinary amorphous

 H_c and W are indicated the data after annealing temperature of 623 K.



Fig. 4. Core losses (*W*) of $Fe_{77}P_7B_{13}Nb_2Cr$, $Fe_{77}P_{11}B_9Nb_2Cr_1$ glassy alloys and $Fe_{75}Si_{10}B_{12}Cr_3$ amorphous alloy as a function of annealing temperature.

hand, the core loss of glassy alloys almost reaches minimum values of $653-783 \text{ kW/m}^3$ at 623 K, 2 times less than the minimum core loss of 1450 kW/m^3 for amorphous alloy, and then is slightly improved to $501-716 \text{ kW/m}^3$ at 723 K as it approaches 754 K and 767 K of T_g , respectively. The optimum annealing temperature of the core loss is 723 K for the $\text{Fe}_{75}\text{Si}_{10}\text{B}_{12}\text{Cr}_3$ amorphous alloy and 623 K for glassy alloys.

As above results, the excellent low core loss characteristics of the glassy alloy can be obtained at 623 K, 100 K less than the optimum annealing temperature of 723 K for amorphous alloy. This result is considered to have a low magnetic anisotropy due to the high stability of amorphous structure is achieved in feature super cooled liquid region of the ΔT_x of 28–31 K glassy alloys. Considering the minimum value of core loss for the alloys, as described above, the extremely low core loss of 653-783 kW/m³ at 623 K for $Fe_{97-x-y}P_xByNb_2Cr_1$ can be explained as due to the high stable glassy structure based on super cooled liquid region featured glassy metal alloy. Taking account of the difference in the optimum annealing temperature for core loss, 623 K for glassy metal alloy and 723 K for the amorphous alloy, we can see that the glassy alloy exhibits the higher structural stability of magnetic softness, which should indicate the higher stability of the glassy phase than the amorphous phase [5,6]. In fact, a profound effect of having low magnetic anisotropy for glassy alloys is explained by the low H_c values of 2.5-3.1 at 623 K shown in Table 1.

Table 1 summarizes T_c , T_g , T_x , ΔT_x , magnetic properties (B_s , H_c) and the core loss for the Fe_{97-x-y}P_xByNb₂Cr₁ glassy alloys and the

typical Fe-based amorphous alloy for Fe₇₅Si₁₀B₁₂Cr₃. Here, T_{max} is adopted as the parameter for GFA. The larger parameters exhibit the higher GFA. The Fe_{97-x-y}P_xB_yNb₂Cr₁ glassy alloys have significant high T_{max} of 110–150 µm, comparable to ordinary amorphous alloy Fe₇₅Si₁₀B₁₂Cr₃ without glass transition, which are probably one of the reasons for the high GFA, leading to the ΔT_x of 28–31 K. The Fe_(97-x-y)P_xB_yNb₂Cr₁ glassy alloys though added Cr exhibit relatively high B_s of 1.27–1.33 T compared with the previously reported amorphous alloy Fe₇₅Si₁₀B₁₂Cr₃ due to the high Fe content of 77 at% and the excellent stability leading to the T_{max} of 110–150 µm. Therefore, we can say that this is the first achievement for simultaneous realization of high B_s and low core loss in Fe-based glassy alloy which Cr is added. In addition, the Fe_{97-x-y}P_xByNb₂Cr₁ glassy alloys also exhibit very excellent corrosion resistance which should enable ultra-high efficient and quality inductor product.

4. Conclusions

- 1. High glass-forming ability with obvious glass transitions was observed in the wide composition range, and the large T_{max} of over 100 µm is observed in 5–13 at% of P, 7–15 at% of B and 78–81 at% of Fe + Nb₂Cr₁ for melt-spun Fe_{97–x–y}P_xB_yNb₂Cr₁ alloys. The largest T_{max} is 150 µm for Fe_{97–x–y}P_xB_yNb₂Cr₁ (x=5–8, y=12–15).
- 2. The extremely low core loss of 653–783 kW/m³, 2 times less than the minimum core loss of ordinary Fe-based amorphous alloy, was obtained for the glassy alloys $Fe_{77}P_7B_{13}Nb_2Cr_1$ and $Fe_{77}P_9B_{11}Nb_2Cr_1$ having super cooled liquid region of a ΔT_x of 28–31 K leading to low magnetic anisotropy with H_c of 2.5–3.1 A/m.
- 3. The optimum temperature for Fe_{97-x-y}P_xB_yNb₂Cr₁ glassy alloy, 100 K less than the previously noted Fe-based amorphous, was 623 K almost reaching the minimum value of core loss.
- 4. The relatively high B_s of 1.3 T than typical Fe-based amorphous alloy was obtained for $Fe_{97-x-y}P_xB_yNb_2Cr_1$ that added Cr of 1 at% to Fe-P-B-Nb system and high corrosion resistance was obtained for $Fe_{97-x-y}P_xB_yNb_2Cr_1$ by added Cr.

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