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Novel FePBNbCr glassy alloys "SENNTIX" with good soft-magnetic properties for high efficiency commercial inductor cores

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ABSTRACT

According to a recent study, Fe-based glassy alloys are expected good soft-magnetic properties such as high saturation magnetization and lower coercive force. We focused on Fe-based glassy alloys and have succeeded in developing novel glassy $F_{97-x-y}P_xB_yNb_2Cr_1$ (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 μ m and high B_s of 1.25–1.35 T. The glassy alloy powder with chemical composition $Fe_{77}P_{10.5}B_{9.5}Nb_2Cr_1$ exhibits an excellent spherical particle shape related to the lower melting point and liquid phase point. In addition, Fe–P=B–Nb–Cr powder/resin composite core has much lower core loss of 653–881 kW/m³, which is approximately 1/3 lower than the conventional amorphous Fe–Si–B–Cr powder/resin composite core and 1/4 lower than the conventional crystalline Fe–Si–Cr powder/resin composite core due to the lower coercive force of 2.5–3.1 A/m. Based on above results, the glassy Fe₇₇P_{10.5}B_{9.5}Nb₂Cr₁ alloy powder enable to achieve ultra-high efficient and high quality products in a commercial inductor. In fact, the surface mounted inductor using Fe–P–B–Nb–Cr powder/resin exhibits the high efficiency of approximately 2.0% compared with the conventional inductors made of the crystalline Fe–Si–Cr powder/resin composite core.

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1. Introduction

According to the technical trend of small and high performance devices, "Note-PC, Mobile-Phones" and so on, the achievement of a high level of power efficiency is required in power supply circuits. The surface mount device type inductor in the power supply is expected to deal satisfactorily with high-current supply and to improve the power loss characteristic. In order to improve these characteristics of power supply, the soft magnetic material for inductor cores is required to have high B_s and low core losses. However, the conventional Ni-Zn ferrite and Mn-Zn ferrite core cannot be used as core materials of inductors for large current power supplies because the $B_{\rm s}$ of 0.5 T is much lower as compared with other soft magnetic materials consisting of metallic alloys. On the other hand, commercial soft magnetic materials consisting of metallic alloys, especially Fe-based materials, have the subject associated with high hysteresis loss that is attributed to large magnetic anisotropy. Hence, we focused on glassy metallic alloys with both high saturation magnetization and low magnetic anisotropy, and developed the Fe-based glassy metallic alloy with

a composition of Fe77P7B13Nb3 for inductor core materials [1]. Corrosion resistance is also one of the important factors in practical use for commercial inductor core materials, as same as the soft magnetic properties. Thus, we tried to improve the corrosion resistance and succeeded in developing a novel glassy alloy with a chemical composition of Fe₇₇P₇B₁₃Nb₂Cr₁ for a new class material of inductor cores [2]. In this paper, we report on a novel glassy alloy $Fe_{97-x-y}P_xB_yNb_2Cr_1$ with high B_s of 1.3 T, low H_c of 2.0 A/m by the low crystalline anisotropy, a low core loss of 881 kW/m³, a high corrosion resistance and excellent spherical particle shape in the produced powder. The surface mounted inductor using the glassy Fe₇₇P₁₀₅B₉₅Nb₂Cr₁ alloy powder/resin composite core exhibits a higher efficiency of about 2.0% compared with the conventional inductor made of crystalline Fe85Si9.5Cr5.5 powder/resin composite core. In addition, high glass-forming ability (GFA) is obtained even in case of using the raw ferroalloy materials of low costs.

2. Experimental procedures

Fe_{97-x-y}P_xB_yNb₂Cr₁, Fe₇₇P_xB_{20-x}Nb₂Cr₁ and Fe₈₅Si_{9.5}Cr_{5.5} were prepared by induction melting the mixture of pre-melted Fe–P, Fe–B, Fe–Nb, Fe–Cr, Fe–Si and pure Fe in an argon atmosphere. The alloy compositions represent the nominal atomic percents. Glassy Fe_{97-x-y}P_xB_yNb₂Cr₁, Fe₇₇P_xB_{20-x}Nb₂Cr₁ alloys and amorphous Fe₇₅Si₁₀B₁₂Cr₃ alloy were produced as ribbon form having a width of about 1 mm and thicknesses of 25–150 μ m by a single roller melt-spinning method in an argon atmosphere. The melting point (*T_m*) and the liquid phase point (*T_l*) were measured by differential thermal analysis (DTA) at a heating rate of 0.17 K/s. The glass

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transition temperature (T_g) , crystallization temperature (T_x) and Curie temperature (T_c) were determined 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 coercive force (H_c) was measured under a field of 1 kA/m with a DC B-H loop tracer at room temperature and after annealing at 623 K. The glassy Fe_{97-x-v}P_xB_vNb₂Cr₁, amorphous Fe₇₅Si₁₀B₁₂Cr₃ and crystalline Fe₈₅Si_{9.5}Cr_{5.5} alloy powders were produced by water atomization. The particle shape of these alloys is observed by scanning electron microscopy (SEM). Powders with an average particle size of 12 μ m were mixed with a resin binder and produced in a ring core form of φ 13 mm $\times \varphi$ 8 mm \times 6.5 mm and a surface mounted device (SMD) type choke coil of $10 \text{ mm} \times 10 \text{ mm} \times 4 \text{ mm}$ with a packing density of 71–73% under a molding pressure of 8.0 GPa after granulation. The inductance of a SMD choke coil using the glassy Fe77P10.5B9.5Nb2Cr1 alloy powder is 0.38 µH, 0.42 µH 0.47 µH and using the crystalline Fe₈₅Si_{9.5}Cr_{5.5} alloy powder is 0.41 μ H. The core losses (W) of the glassy Fe_{97-x-y}P_xB_yNb₂Cr₁ alloys and the amorphous Fe₇₅Si₁₀B₁₂Cr₃ alloy were measured under a frequency of 300 kHz and a field of 50 mT with B-H loop analyzer for the ring core shape after annealing of 623 K. The crystalline Fe₈₅Si_{9.5}Cr_{5.5} alloy is after annealing of 423 K. The efficiency of SMD choke coil of the glassy Fe77P10.5B9.5Nb2Cr1 was measured by a voltage step down DC/DC converter evaluation board of MAXIM 1717 evolution kit input voltage of 12.0V output voltage of 1.0 V and frequency of 300 kHz after annealing of 623 K. The SMD choke of the crystalline $Fe_{75}Si_{10}B_{12}Cr_3$ alloy was measured with same condition after annealing of 423 K.

3. Results and discussion

It is well known that many kind of soft-magnetic materials have used in inductor core material. The iron-based amorphous alloys are one of inductor materials having low core losses. Some kind of iron-metalloid amorphous alloys for inductor core materials have been developed and used in practical devices. Especially, Fe–Si–B–(Cr) amorphous alloy system is now a major material widely utilized for industrial products. But iron-based amorphous alloys are not used to the SMD choke coil material due to the higher annealing temperature of 773 K. Because insulating coating of inner-cupper wire in SMD choke coil is posed big damage with a higher annealing temperature of 773 K. Therefore, we focused on Fe-based glassy alloys [3] and developed the novel glassy alloy composition of Fe–P–B–Nb alloy system exhibiting high performance at the lower annealing temperature of 623 K [4].

Fig. 1 shows the compositional dependence of the maximum thickness for glass formation (T_{max}) of melt-spun Fe_{97-x-y}P_xB_yNb₂Cr₁ alloys. The glassy phase is obtained in a wide compositional range of over 100 µm lies in the enclosed range of 5–13 at% P, 7–15 at% B and 78–81 at% Fe+Nb₂Cr₁. In particular, the largest T_{max} of 150 µm can be obtained in compositional range of Fe_{97-x-y}P_xB_yNb₂Cr₁ (x=5–8, y=12–15). Higher



Fig. 1. The compositional dependence of the maximum thickness for glass formation (T_{max}) for melt-spun Fe_{97-x-y}P_xB_yNb₂Cr₁ alloys.



Fig. 2. The compositional dependence of the saturation magnetic flux density (B_s) for melt-spun Fe_{97-x-y}P_xB_yNb₂Cr₁ alloys.

 T_{max} is a significantly important characteristic to produce powders for an inductor core, using water atomization.

Fig. 2 shows the compositional dependence of the saturation magnetic flux density (B_s) for glassy $Fe_{97-x-y}P_xB_yNb_2Cr_1$ alloys. The high B_s of over 1.25 T is obtained in a wide compositional range of the enclosed range of 5–15 at% P, 5–16 at% B and 78–82 at% Fe + Nb₂Cr₁, as indicated with line of 1.25 T. The largest B_s of 1.35 T can be obtained in compositional range of Fe_{97-x-y}P_xB_yNb₂Cr₁ (x=4–8, y=12–16).

Fig. 3 shows the phosphorus compositional dependence of melting point (T_m) and liquid phase point (T_l) . T_m and T_l lowering trends with phosphorus content are significantly observed in a wide phosphorus range from 7.0 at% to 15.0 at%. Especially, T_l is lowered in the upper range of 9.5 at% P. Therefore, we decided that the optimum composition for making powders in glassy Fe_{97-x-y}P_xB_yNb₂Cr₁ systems is Fe₇₇P_{10.5}B_{9.5}Nb₂Cr₁ due to low T_m and T_l one can obtain good spherical shape particles by water atomization. Fe₇₇P_{10.5}B_{9.5}Nb₂Cr₁ also exhibits high B_s of 1.3 T.



Fig. 3. The phosphorus composition dependence of melting point (T_m) and liquid phase point (T_l) .





Particle shape of conventionally crystalline Fe₈₅Si₉₅Cr₅₅ alloy



Fig. 4. SEM micrographs of particle shapes of the glassy $Fe_{77}P_{10.5}B_{9.5}Nb_2Cr_1$ and crystalline $Fe_{85}Si_{9.5}Cr_{5.5}$ alloy powders.

Fig. 4 shows SEM micrographs of the glassy $Fe_{77}P_{10.5}B_{9.5}Nb_2Cr_1$ and the crystalline $Fe_{85}Si_{9.5}Cr_{5.5}$ alloy powders. The glassy $Fe_{77}P_{10.5}B_{9.5}Nb_2Cr_1$ powder consists of much spherical particles than the crystalline $Fe_{85}Si_{9.5}Cr_{5.5}$ alloy powder having a higher T_m of 1720 K and T_l of 1752 K. On the other hand, the crystalline $Fe_{85}Si_{9.5}Cr_{5.5}$ alloy powder with



Fig. 5. The compositional dependence of core losses (*W*) for the glassy $Fe_{97-x-y}P_xB_yNb_2Cr_1$ alloys powder/resin composite core after annealing temperature at 623 K.



Fig. 6. The output current dependence of the power load efficiency for the glassy $Fe_{77}P_{10.5}B_{9.5}Nb_2Cr_1$ and crystalline $Fe_{85}Si_{9.5}Cr_{5.5}$ alloys SMD choke coils.

high T_m of 1720 K exhibits spheroidal particle shape is no good.

Fig. 5 shows the compositional dependence of core losses (*W*) for glassy $Fe_{97-x-y}P_xB_yNb_2Cr_1$ alloys powder/resin composite core after annealing temperature of 623 K. For 80 at% Fe + Nb₂Cr₁, excellent low core losses of 653–881 kW/m³ are obtained in a wide compositional range of 7–11 at% P, and 9–13 at% B, as enclosed with in the line of 120 µm. This result is presumably the effect of the high glass formation resulting from the high stability of amorphous structure as evidenced from the existence of the super-cooled liquid region (ΔT_x) of 35 K featuring the glassy alloy.

Fig. 6 shows the output current dependence of the power load efficiency for the glassy $Fe_{77}P_{10.5}B_{9.5}Nb_2Cr_1$ and the crystalline $Fe_{85}Si_{9.5}Cr_{5.5}$ alloy SMD choke coil. Generally, the efficiency of the inductor is indicated by the quality factor (*Q*) giving by the following formula:

$$Q = \frac{\omega I}{R}$$

here Q is the quality factor, ω is the angular velocity, *L* is the inductance, and *R* is the magneto-resistance. It can be confirmed to the inductor that one with high inductance can obtain high efficiency from the above formula. However, the glassy Fe₇₇P_{10.5}B_{9.5}Nb₂Cr₁ alloy SMD choke coil with low inductance of 0.38 μ H exhibits a higher efficiency compared with the crystalline Fe₈₅Si_{9.5}Cr_{5.5} alloy SMD choke coil with high inductance of 0.41 μ H. For the glassy Fe₇₇P_{10.5}B_{9.5}Nb₂Cr₁ alloy SMD choke coil with an inductance of 0.42 μ H, one can achieve a higher efficiency of approximately 2.0% compared with the crystalline Fe₈₅Si_{9.5}Cr_{5.5} alloy SMD choke coil of 0.41 μ H. Thus, the high efficiency of SMD choke coil can be obtained by using the glassy Fe₇₇P_{10.5}B_{9.5}Nb₂Cr₁ alloy powders which named "*SENNTIX*". *SENNTIX* is a registered trademark for the glassy Fe₇₇P_{10.5}B_{9.5}Nb₂Cr₁ alloy powder.

Table 1 summarizes T_c , T_g , T_x , ΔT_x , T_m , T_l and T_{max} , magnetic properties (B_s , H_c) and core losses (W) for the glassy Fe_{97-x-y}P_xB_yNb₂Cr₁ alloys, amorphous Fe₇₅Si₁₀B₁₂Cr₃ and crystalline Fe₈₅Si_{9.5}Cr_{5.5} alloys. Here, T_{max} is applied as the parameter for GFA. The glassy Fe_{97-x-y}P_xB_yNb₂Cr₁ alloys have fairly larger T_{max} of 110–150 µm compared with the amorphous Fe₇₅Si₁₀B₁₂Cr₃ alloy without glass transition. This is consistent with the appearance of super-cooled liquid region with ΔT_x of 29–37 K. The glassy Fe_{97-x-y}P_xB_yNb₂Cr₁ alloys exhibit relatively high B_s of 1.26–1.33 T in comparison with the amorphous Fe₇₅Si₁₀B₁₂Cr₃ alloy with high Fe content. Additionally, this novel glassy Fe_{97-x-y}P_xB_yNb₂Cr₁ alloy has much lower core loss characteristics due to having lower H_c than the ordinary amorphous Fe₇₅Si₁₀B₁₂Cr₃ alloy. Hence one can conclude that the glassy Fe_{97-x-y}P_xB_yNb₂Cr₁ alloy is only one glassy alloy system of very few Fe-based glassy alloys with having both

Table 1

The Curie temperature (T_c) , glass transition temperature (T_g) , crystallization temperature (T_x) , melting temperature (T_m) , liquid phase point (T_l) , the parameter of glass-forming ability $(\Delta T_x, T_{max})$ and the magnetic properties (B_s, H_c) and core losses (W) for Fe_(97-x-y)P_xB_yNb₂Cr₁ glassy alloys, typical Fe-based amorphous alloy and the conventional crystalline alloy.

Characteristic material	T_c (K)	$T_{\varepsilon}.(K)$	T_{x} (K)	ΔT_x (K)	T_m (K)	T_l (K)	T_{max} (mm)	B_s (T)	$H_c (A/m)$	$W(kW/m^3)$	Formation
Fe77P7B13Nb2Cr1	556	767	795	36	1317	1402	150	1.33	2.5	653	Glass
Fe ₇₇ P ₉ B ₁₁ Nb ₂ Cr ₁	553	759	789	31	1315	1403	130	1.33	2.5	810	Glass
Fe77P10.5B9.5Nb2Cr1	544	750	785	35	1308	1396	130	1.30	3.8	881	Glass
Fe ₇₇ P ₁₁ B ₉ Nb ₂ Cr ₁	549	754	783	29	-	-	130	1.30	2.5	783	Glass
Fe ₇₇ P ₁₁ B ₇ Nb ₂ Cr ₁	536	740	771	37	1306	1366	110	1.26	3.1	-	Glass
Fe ₇₆ P ₁₀ B ₁₁ Nb ₂ Cr ₁	554	762	793	31	-	-	115	1.28	2.8	706	Glass
Fe ₇₅ Si ₁₀ B ₁₂ Cr ₃	648	-	848	-	1435	1469	50	1.25	14.8	1989	Amorphous
Fe ₈₅ Si ₉ Cr ₆	964	-	-	-	1720	1752	-	1.65	44.0	2361	Crystalline

 H_c and W are after annealing at 623 K.

high B_s and low core loss at lower annealing temperature [5,6]. Furthermore, the glassy $Fe_{97-x-y}P_xB_yNb_2Cr_1$ alloys with low T_m and T_l are suitable for producing powders consist of spherical particles to be used in inductor cores.

4. Conclusions

- 1. The large T_{max} of over 100 µm was obtained in 5–13 at% P, 7–15 at% B and 78–81 at% Fe + Nb₂Cr₁. The largest T_{max} of 150 µm was achieved in the composition range of Fe_{97–x–y}P_xB_yNb₂Cr₁ (x=5–8, y=12–15).
- 2. High B_s of 1.35 T was also obtained for the glassy $Fe_{97-x-y}P_xB_yNb_2Cr_1$ (x=4–8, y=12–16) alloys.
- 3. Low T_m and T_l were obtained for the glassy $Fe_{77}P_xB_{20-x}Nb_2Cr_1$ (x=7-15) alloys, and $Fe_{77}P_xB_{20-x}Nb_2Cr_1$ (x=7-15) alloys were exhibited to the optimum soft-magnetic material for a powder of an inductor core.

4. The excellent high efficiency of approximately 2.0% with the SMD choke coil were attained for the glassy $Fe_{77}P_{10.5}B_{9.5}Nb_2Cr_1$ alloy to leading in conjunction with the minimum value of core loss of 881 kW/m³.

References

- [1] H. Matsumoto, A. Urata, Y. Yamada, A. Inoue, J. Alloys Compd. 504S (2010) 139-141.
- [2] H. Matsumoto, A. Urata, Y. Yamada, A. Inoue, IEEE Trans. Magn. 46 (2) (2010) 373.
- [3] A. Inoue, B.L. Shen, H.M. Kimura, J. Metastable Nanocryst. Mater. 3 (2004) 20–21.
 [4] H. Matsumoto, A. Urata, Y. Yamada, A. Makino, J. Appl. Phys. 105 (2009)
- [4] H. Matsumoto, A. Urata, Y. Yamada, A. Makino, J. Appl. Phys. 105 (2009) 07A317.
- [5] T. Bitoh, A. Makino, A. Inoue, Mater. Trans. 44 (2003) 2020.
- [6] T. Bitoh, A. Makino, A. Inoue, J. Appl. Phys. 99 (2006) 08F102.