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RAPID PUBLICATION

Soft Magnetic Properties of Ring Shape Bulk Glassy Fe–Al–Ga–P–C–B–Si Alloy Prepared by Copper Mold Casting

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Structural and soft magnetic properties were investigated for a bulk glassy $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ alloy in a ring shape form with an outer diameter of 10 mm, an inner diameter of 6 mm and a thickness of 1 mm prepared by copper mold casting. The bulk sample composes of an amorphous single phase. The supercooled liquid region (ΔT_x) defined by the difference between the crystallization temperature (T_x) and glass transition temperature (T_g) for the bulk glassy alloy is about 60 K, being in agreement with that of the corresponding amorphous alloy ribbon. Saturation magnetization (σ_s) and Curie temperature (T_c) for the bulk sample are about 1.2 T and 620 K, respectively, both of which agree with those of the melt-spun ribbon. Furthermore, the maximum permeability (μ_{max}) and the coercivity (H_c) for the bulk glassy sample in as cast state are about 110000 and 2.2 A/m, respectively. These excellent soft magnetic properties are presumably due to a higher degree of structural homogeneity resulting from its high glass-forming ability. It is concluded that the good soft magnetic properties combined with high glass-forming-ability and castability of the Fe–Al–Ga–P–C–B–Si glassy alloy hold promise for its development as a future engineering magnetic material.

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Keywords: *iron-based glassy alloy, super-cooled liquid region, bulk amorphous alloy, soft magnetic properties, copper mold casting*

I. Introduction

Since the finding of good soft magnetic properties for the Fe-based amorphous alloys, many applications for these alloys have been developed(1). However, the shapes of amorphous alloys have been usually limited to sheet, wire, and film because of their low glass forming ability. For that, further extension of its application fields has been restricted. For instance, it is much difficult to laminate such thin amorphous ribbons to make transformers and/or inductors and also to improve a lamination factor.

In the last few decades, great efforts have been devoted to prepare a bulk amorphous alloy by powder metallurgy. However, no bulk amorphous alloys exhibiting good soft magnetic properties have not been synthesized yet because of their structural inhomogeneity(2).

Recently, bulk amorphous alloys have been formed in multicomponent Mg-(3)(4), Ln-(5)(6), Zr-(7)(8) and Zr–Ti-based(9) (Ln = lanthanoide metal) alloy systems by copper mold casting. These bulk amorphous alloys have a large supercooled liquid region of over 60 K before crystallization. The appearance of the large supercooled liquid region implies a high resistance against crystallization, leading to a high glass forming ability. In addition, it has been reported that deformation and working processes are much easier in the supercooled liquid region because of its low viscosity and ideal Newtonian flow(10). The above-described bulk amorphous alloys always satisfy the following three empirical rules for the achievement of high glass-forming ability(11)-(14); *i.e.*, (1) multicomponent alloy systems consisting of more than three elements, (2) significantly different atomic size ratios above about 12% among the main constituent elements, and (3) negative heats of mixing among their elements. According to these empirical rules, we have subsequently searched a new Fe-based amorphous alloy with high glass-forming ability. So far, we have found that Fe–Al–Ga–P–C–B–Si amorphous alloy sheets have a large supercooled liquid region exceeding 60 K before crystallization and a maximum thickness of 280 μm for glass formation (t_{max}) prepared by single roller melt spinning(15). We have subsequently prepared directly a bulk amorphous ring sample by the copper mold casting process with the aim of eliminating an intermediate working process to a core shape and investigated the relation between glass-forming-ability and soft magnetic properties.

In this paper, we present the structure, thermal stability of supercooled liquid region and soft magnetic properties for Fe–Al–Ga–P–C–B–Si glassy alloys in a ring shape form prepared by the copper mold casting method.

II. Experimental Procedure

A multicomponent alloy with composition of $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ was used for searching studies because the largest supercooled liquid region before crystallization in Fe-Al-Ga-P-C-B-Si system was obtained at the composition(16). Its alloy ingot was prepared by induction melting the mixtures of pure Fe, Al, Si and Ga metals, premelted Fe-P and Fe-C alloys and pure crystal boron in an argon atmosphere. The ring shape bulk sample with an outer diameter of 10 mm, an inner diameter of 6 mm and a thickness of 1 mm was prepared by copper mold casting method. The amorphous character was examined by X-ray diffraction, OM, SEM and TEM. Thermal stability associated with glass transition, supercooled liquid region and crystallization was examined at a heating rate of 0.67 K/s by differential scanning calorimetry (DSC). Saturation magnetization (σ_s), coercive force (H_c) and maximum permeability (μ_{max}) were measured at room temperature with a vibrating sample magnetometer (VSM) under 800 kA/m and a *B-H* loop tracer under 0.8 kA/m. Magnetic domain structure was observed by BMG method.

III. Results and Discussion

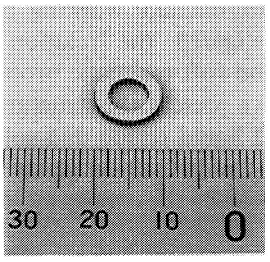
1. Structure and thermal properties

Figure 1 shows an external appearance of the ring-shape $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ alloy sample with the thickness of 1 mm. This bulk alloy has a good luster for typical metallic amorphous alloys and no distinctive contrast revealing the precipitation of crystalline phase is seen.

Figure 2 shows the XRD pattern for the ring-shape bulk sample. The XRD pattern consists only of a broad peak at a wave vector ($K_p = 4\pi \sin\theta/\lambda$) of about 31 nm^{-1}.

Figure 3 shows the optical micrograph and SEM image of the cross section for the bulk ring sample. The bright field image and selected-area diffraction pattern (SAED) at the central region of the bulk sample observed by TEM are also shown in Fig. 3. Only featureless contrast and the halo pattern based on the homogeneous amorphous phase can be seen.

Figure 4 shows the DSC curves of the ring-shape bulk sample, together with the data for the melt-spun sheets with the thicknesses of 20 μm prepared by the single roller melt-spinning method. The glass transition temperature (T_g), the onset temperature of crystallization (T_x) and the ΔT_x for the bulk sample are 748, 807 and 59 K, respectively. These values of the bulk samples are



$Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$

Fig. 1 Outer morphology of the bulky sample in a ring shape form with a thickness of 1 mm.

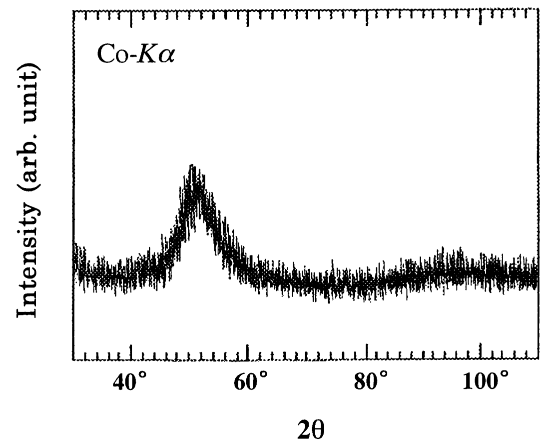
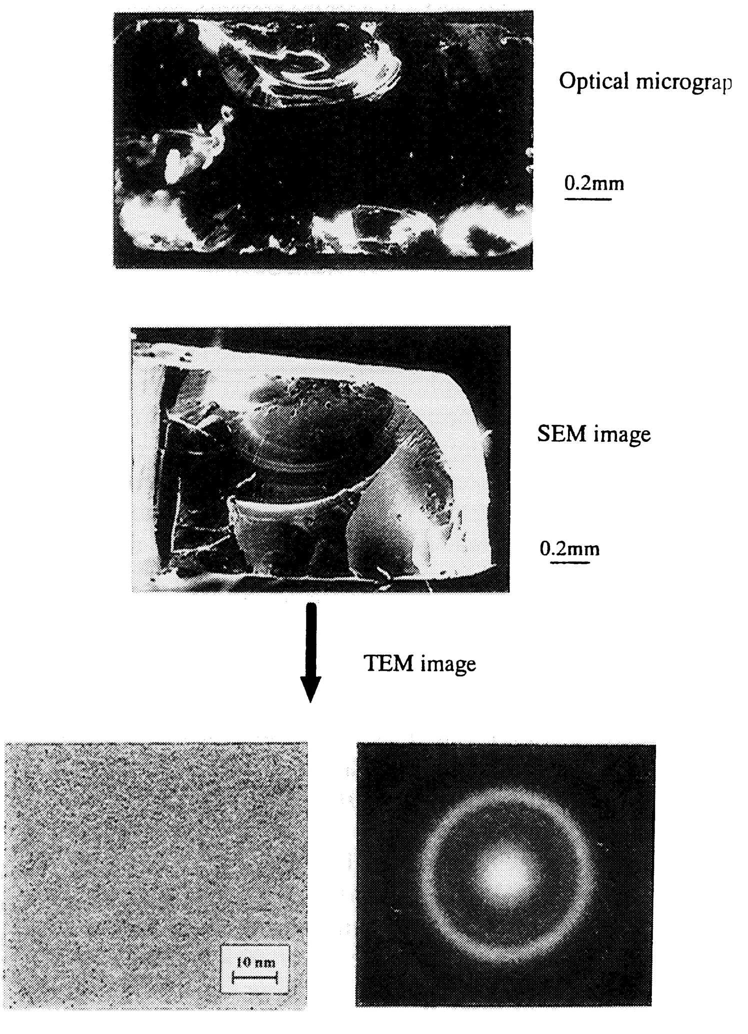


Fig. 2 XRD Pattern for the bulk $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ glassy alloy in a ring shape form.



Bright field image SAED

Fig. 3 OM, SEM, TEM and SAD images of the cross section for the bulk $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ glassy alloy in a ringed shape form.

almost the same as those (743, 806 and 63 K) for the melt-spun ribbons. The shape of the curve is also similar to that of the melt-spun ribbons. This result indicates that the bulk sample consists of a homogeneous amorphous structure which is almost the same as that of the melt-spun sheets.

2. Magnetic properties

Figure 5 shows the D.C. *B-H* loops of the ring shape bulk glassy alloy in as cast state and the ring shape sheet sample (ϕ10 μm × ϕ6 μm × 20 μm) in an annealed state fabricated by stamping the corresponding melt-spun ribbon measured by VSM. The enlarged loops in the low magnetic field and magnetization range measured by D.C. *B-H* loop tracer also shown. The saturation magnetization (σ_s) and coercivity (H_c) for the ring-shape bulk sample are about 172×10^{-6} Wbm·kg^{-1} and 2.2 A/m, respectively, both of which are approximately the same as those of the ring shape sheet sample. It should be noted that the soft magnetic properties for the bulk sample, however, is more excellent than those of the sheet sample.

Figure 6 shows the micrographs of the magnetic

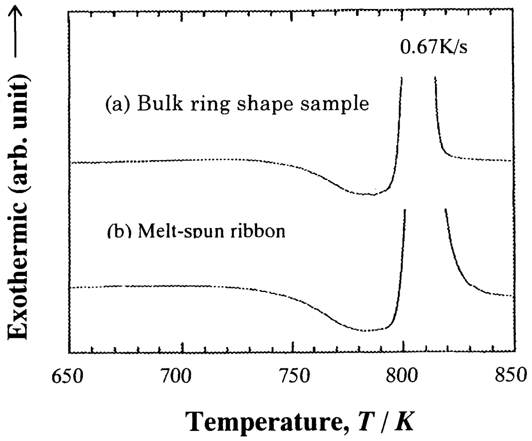


Fig. 4 DSC curves for the bulky $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ glassy alloy in a ring shape form and in a melt-spun ribbon with the thickness of 20 μm.

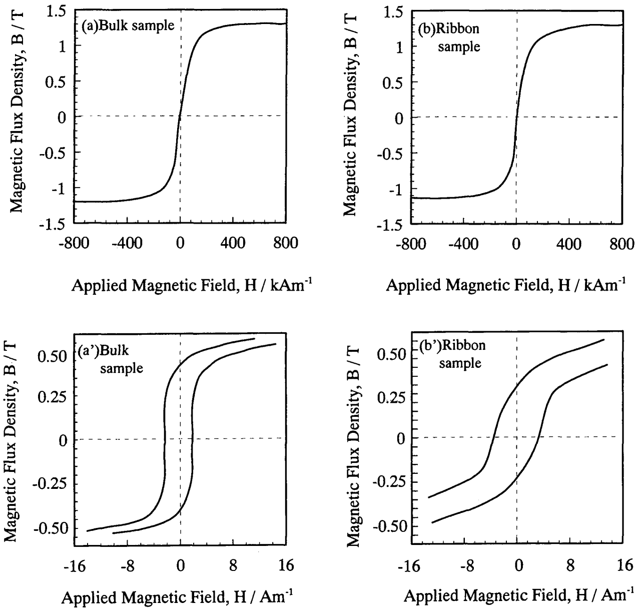
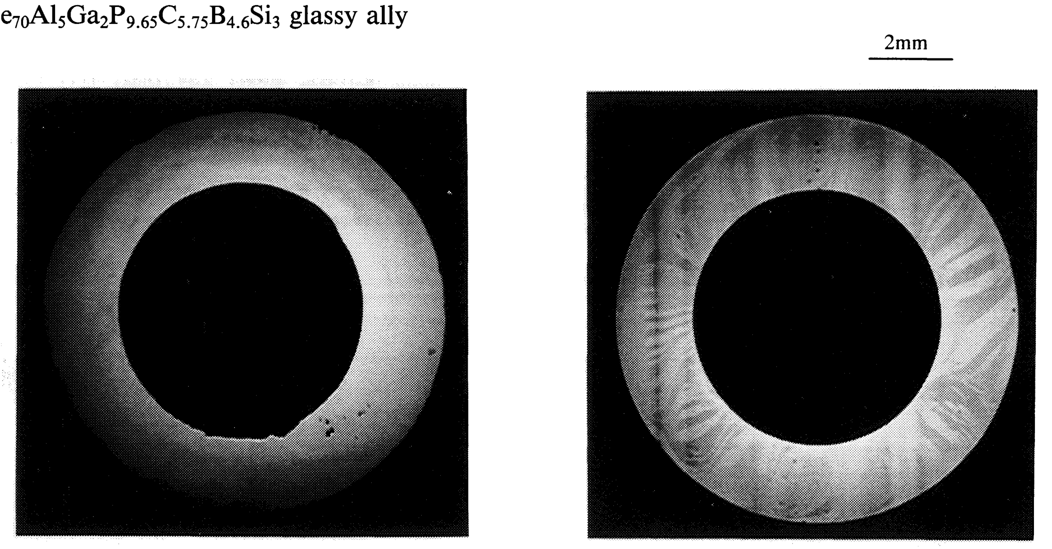


Fig. 5 D.C. *B-H* loops of (a) the bulky $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ glassy alloy in a ring shape form with the thickness of 1 mm in as-cast state and of (b) a corresponding melt-spun ring shape sheet sample with the thickness of 20 μm in an annealed state, and enlarged D.C. *B-H* loops in the low magnetic field and magnetization range for the hysterisis *B-H* loops shown in (a) and (b).



(a)Bulk sample
(ϕ10×ϕ6×1^t)

(b)Sheet Sample
(ϕ10×ϕ6×0.02^t)

Fig. 6 Magnetic domain structure of (a) the bulk $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ glassy alloy in a ring shape form with the thickness of 1 mm in as-cast state and of (b) a corresponding melt-spun ring shape sheet sample with the thickness of 20 μm in an annealed state.

Table 1 Magnetic properties for the bulk $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ glassy alloy in a ring shape form in as-cast state and in a corresponding melt-spun ribbon with the thickness of 20 μm in an annealed state.

	Saturation magnetization, σ_s ($\times 10^{-6}$ Wbm·kg^{-1})	Coercivity H_c (Am^{-1})	Maximum permeability μ_{max}
Cast bulk sample	172	2.2	110,000
Melt-spun ribbon	172	3.7	27,000

domain structures for the bulk ring sample and the ring shape sheet sample. The domain structure for the bulk sample looks like a coarse concentric circle pattern in spite of vague outline. On the other hand, that for the sheet sample forms a fine radial pattern clearly.

This indicates that the concentric circle pattern is more favorable to magnetize at the circumference direction than the fine radial pattern in the low magnetic field. The reason is that, at the concentric circle pattern, not all of the domain wall displacement are needed on account of a part of the easy axis having already fitted toward that direction. The difference in the magnetic domain pattern between the bulk sample and the sheet sample arises presumably due to the difference in distribution of inner stress caused by the preparation methods and their sample thicknesses. A detailed investigation of the relation between the domain structure and inner stress for these samples will give rise to some insight in the clarification of the reason for the achievement of the large μ_{max} and low H_c of the ring-shape bulk Fe-based glassy alloys.

Table 1 summarizes the σ_s, H_c and μ_{max} for the ring-shape bulk sample and the melt-spun ribbon. As is evident from this table, it is concluded that the Fe-Al-Ga-P-C-B-Si glassy alloys are very useful materials for practical use because of their excellent soft magnetic properties as well as high glass-forming-ability and high castability.

IV. Summary

The bulk glassy $Fe_{70}Al_5Ga_2P_{9.65}C_{5.75}B_{4.6}Si_3$ alloy in a ring shape form with an outer diameter of 10 mm, an inner diameter of 6 mm and a thickness of 1 mm was prepared by the copper mold casting method. The sample composed of a single amorphous phase and had a large supercooled liquid region of 59 K, which was the same as that of the melt-spun ribbon. Magnetic properties of σ_s, H_c and μ_{max} for the bulk glassy alloy were 172×10^{-6} Wb·m/kg, 2.2 A/m and 110000, respectively. The good soft magnetic properties for the bulk Fe-based glassy alloy are presumably due to good structural homogeneity resulting from its high glass-forming-ability.

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