

# Soft Magnetic Properties of Ring-Shaped Fe-Co-B-Si-Nb Bulk Metallic Glasses

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The reduction of the Nb content in the  $(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$  bulk metallic glass (BMG) has been studied. The glass-forming ability (GFA) is reduced by decreasing the Nb content, but it can be enhanced by replacing partially Fe by Co. Furthermore, the saturation magnetization of the  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  BMG is 1.35 T, being with 13% larger than that of the base alloy  $(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$ .  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  BMG exhibits slightly larger  $B_{800}$  (the magnetic flux density at 800 A/m) and smaller core losses (20%-30%) compared with the commercial Fe-6.5 mass% Si steel.

**Keywords :** bulk metallic glasses, soft magnetic properties, magnetization, coercivity, permeability, core losses

## 1. Introduction

The alloys in the bulk forms have historically been cast from the liquid state. Among them, the eutectic compositions are widely used because of their low melting temperature, which facilitates their processing. However, such alloys in bulk form usually exhibit crystalline structures with periodic atomic configurations, because of the extreme instability of the metallic supercooled liquid against crystallization [1, 2]. Such structural limitations were broken towards the end of the last century by the discovery of stabilized phenomenon of supercooled metallic liquids [3-5].

It is well known that many Fe-based alloys with an amorphous structure exhibit good soft magnetic properties. However, the glass-forming ability (GFA) of the ordinary Fe-based amorphous alloys is low and hence their formation has required higher cooling rates than  $10^5$  K/s. Therefore, the shape of the ordinary Fe-based amorphous alloys has been usually limited to thin sheet, wire and film, leading to the restriction of their application fields. In the last 15 years, a number of Fe-based soft magnetic bulk metallic glasses (BMGs) with a large supercooled liquid region ( $\Delta T_x$ ) between the glass transition temperature ( $T_g$ ) and the crystallization temperature ( $T_x$ ) have been found [6, 7]. The BMGs have

large GFA and, therefore, they can be used to prepare bulk amorphous alloys with thicknesses of few millimeters by casting. Furthermore, the soft magnetic properties of some Fe-based BMGs are better than those of ordinary amorphous alloys; therefore, these BMGs can be used in applications as soft magnetic materials [8-10].

One of the disadvantages of the Fe-based soft magnetic BMGs is the smaller saturation magnetization (typically 1.2 T or less) [6, 7] compared with the ordinary Fe-based amorphous alloys, such as the ternary Fe-B-Si alloys. However, in the development of the Fe-based BMGs, up to now the priority was given to the achievement of the large GFA. Therefore, there might be a possibility to improve additionally the saturation magnetization of Fe-based BMGs. In the present study, the influence of the reduction of the Nb content on the magnetization of Fe-B-Si-Nb BMGs has been examined. The soft magnetic properties of the ring-shaped bulk specimens have been investigated, too.

## 2. Experimental Procedure

The mother alloys were prepared by arc-melting a mixture of pure Fe (99.99%), Co (99.9%), B (99.5%), Si (99.999%) and Nb (99.9%) in an argon atmosphere. Ring-shaped specimens with the outer diameter of 10.0 mm, 6.0 mm the inner diameter and 0.50 mm in thickness were prepared by copper mold casting in an argon atmosphere. The specimens have been annealed for 600 s

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at  $0.97T_g$  in vacuum, a temperature higher than the Curie temperature ( $T_c$ ). No magnetic field was applied during annealing. After annealing, the Co-free specimens were cooled naturally. In order to avoid an induced magnetic anisotropy, the Co-containing alloys were quenched in water after annealing.

The amorphicity of the specimens was examined by X-ray diffractometry (XRD) with Cu  $K_\alpha$  incident radiation. The Curie temperature ( $T_c$ ), glass transition temperature ( $T_g$ ) and crystallization temperature ( $T_x$ ) of the melt-spun tapes, prepared by single-roller melt-spinning technique, of 1.5 mm in width and approximately 25  $\mu\text{m}$  in thickness were measured with a differential scanning calorimeter (DSC) at a heating rate of 0.67 K/s. The relative permeability ( $\mu_r$ ), the hysteresis curves and the core loss ( $P_{cm}$ ) of the ring-shaped bulk specimens were measured with a vector impedance analyzer, a DC  $B$ - $H$  loop tracer under a maximum magnetic field ( $H_m$ ) of 2.0 kA/m and an AC  $B$ - $H$  analyzer, respectively. The saturation mass magnetization ( $\sigma_s$ ) of the annealed melt-spun tapes was also measured with a magnetic balance under  $H_m$  of 0.80 MA/m. The density was measured by the Archimedian method. All the measurements were performed at room temperature.

### 3. Results and Discussion

The  $(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$  alloy was selected as the base composition because it has large GFA [11]. Since the addition of Nb to Fe-B-Si amorphous alloys remarkably decreases the magnetization [12], the decrease of the Nb

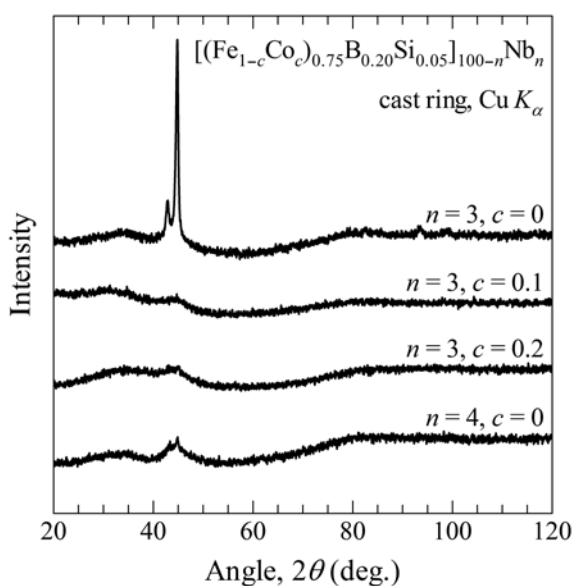


Fig. 1. X-ray diffraction profiles of ring-shaped as-cast  $[(\text{Fe}_{1-c}\text{Co}_c)_{0.75}\text{B}_{0.20}\text{Si}_{0.05}]_{100-n}\text{Nb}_n$  bulk specimens.

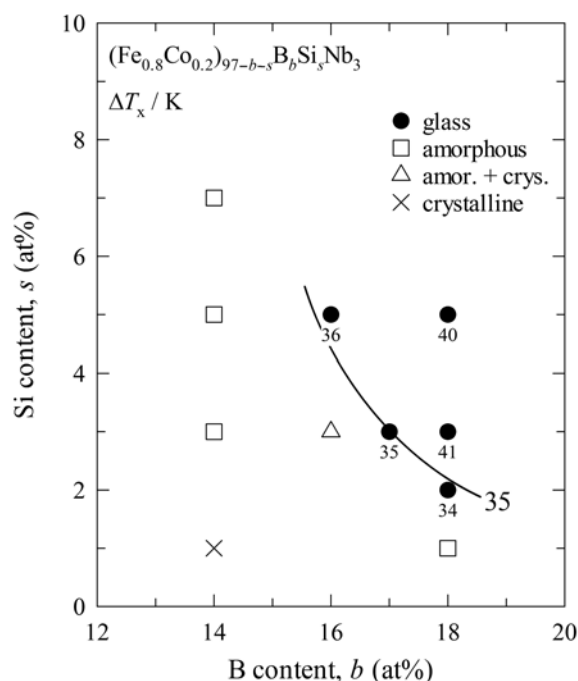


Fig. 2. Compositional dependence of the supercooled liquid region ( $\Delta T_x$ ) for melt-spun  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{97-b-s}\text{B}_b\text{Si}_5\text{Nb}_3$  tapes.

content was tried. Fig. 1 shows the XRD profiles of the ring-shaped as-cast  $[(\text{Fe}_{1-c}\text{Co}_c)_{0.75}\text{B}_{0.20}\text{Si}_{0.05}]_{100-n}\text{Nb}_n$  specimens. The  $(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$  alloy specimen has an almost single glassy structure. When the Nb content is decreased from 4 to 3 at%, the glassy structure cannot be obtained. If Fe is substituted partially by Co to compensate the decrease of the GFA [11], the glassy structure is achieved even for the alloys with 3 at% Nb. However, the alloy with  $n = 3$  and  $c = 0.1$  could not be cast as ring-shaped bulk specimens without cracks, because the alloy is too brittle. Therefore, it is necessary to have  $c \geq 0.2$  for  $n = 3$ .

$\text{Fe}_{67}\text{Co}_{18}\text{B}_{14}\text{Si}_1$  (Fe:Co  $\approx$  0.8:0.2) alloy is an example of Fe-based amorphous alloy with large saturation magnetization ( $= 1.8 \text{ T}$ ) [13]. Following this example, we optimized the content of B and Si to increase the magnetization, and Fig. 2 shows the compositional dependence of the supercooled liquid region ( $\Delta T_x$ ) of melt-spun  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{97-b-s}\text{B}_b\text{Si}_5\text{Nb}_3$  specimens. The glass transition is observed only in the compositional range of  $b \geq 16$  and  $b + s \geq 20$ .

Figs. 3 and 4 show the compositional dependence of the Curie temperature ( $T_c$ ) and saturation mass magnetization ( $\sigma_s$ ) at room temperature for the melt-spun tapes. The Curie temperature shows a tendency to decrease rapidly with increasing the Si content. This result suggests that Si content should be as low as possible to obtain large  $\sigma_s$  at room temperature. The maximum value of  $\sigma_s$  ( $185 \times 10^{-6}$

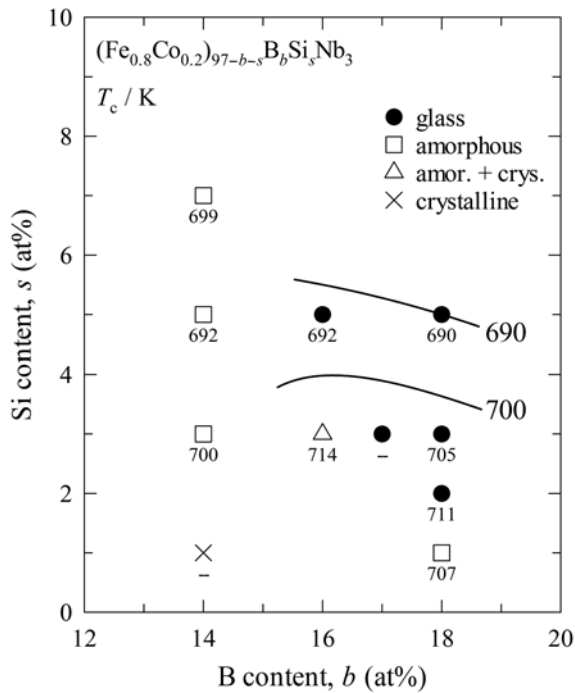


Fig. 3. Compositional dependence of Curie temperature ( $T_c$ ) for melt-spun  $(Fe_{0.8}Co_{0.2})_{97-b-s}B_bSi_sNb_3$  tapes.

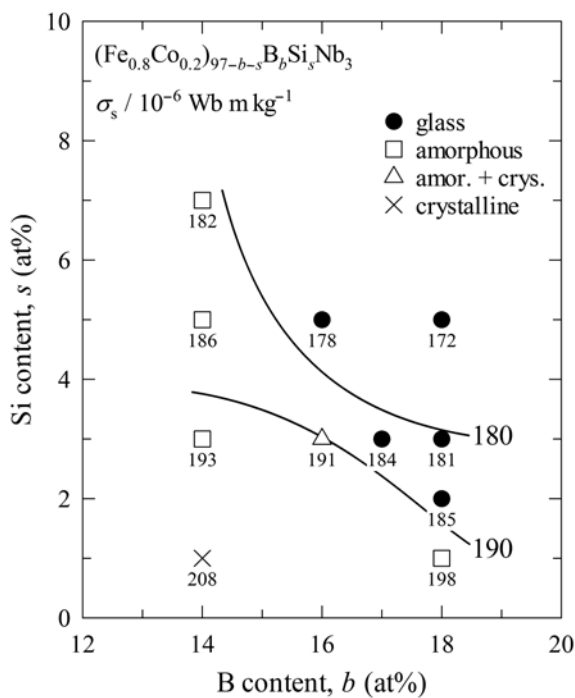


Fig. 4. Compositional dependence of saturation mass magnetization ( $\sigma_s$ ) at room temperature for melt-spun  $(Fe_{0.8}Co_{0.2})_{97-b-s}B_bSi_sNb_3$  tapes.

Wb m/kg) has been obtained for the glassy alloys with  $b = 18$  and  $s = 2$ .

The glass-forming ability of the alloys was confirmed

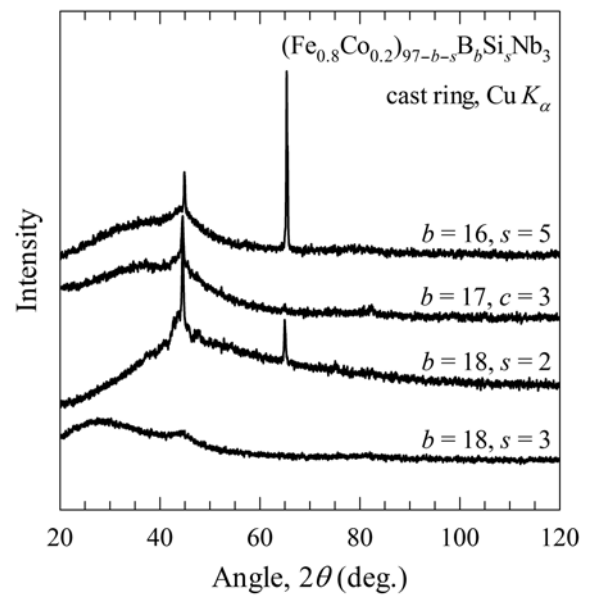


Fig. 5. X-ray diffraction profiles of ring-shaped as-cast  $(Fe_{0.8}Co_{0.2})_{97-b-s}B_bSi_sNb_3$  bulk specimens.

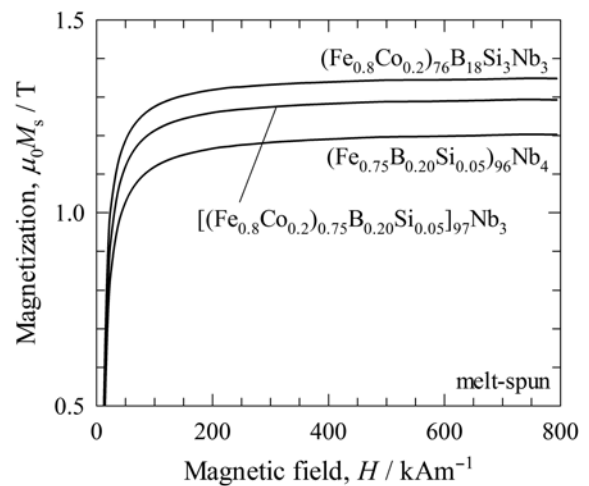


Fig. 6. Magnetization curves of  $(Fe_{0.75}B_{0.20}Si_{0.05})_{96}Nb_4$ ,  $[(Fe_{0.8}Co_{0.2})_{0.75}B_{0.20}Si_{0.05}]_{97}Nb_3$  and  $(Fe_{0.8}Co_{0.2})_{76}B_{18}Si_3Nb_3$  bulk metallic glasses.

by producing the ring-shaped bulk specimens. Fig. 5 shows the XRD profiles of the ring-shaped as-cast  $(Fe_{0.8}Co_{0.2})_{97-b-s}B_bSi_sNb_3$  specimens. The bulk specimen of the alloy with  $b = 16$  and  $s = 5$  was not produced because the magnetization of the alloy is obviously inferior to that of the others. It is only the alloy with  $b = 18$  and  $s = 3$  for which obtained the bulk specimen has a single glassy phase. Fig. 6 shows the magnetization curves of the  $(Fe_{0.75}B_{0.20}Si_{0.05})_{96}Nb_4$ ,  $[(Fe_{0.8}Co_{0.2})_{0.75}B_{0.20}Si_{0.05}]_{97}Nb_3$  and  $(Fe_{0.8}Co_{0.2})_{76}B_{18}Si_3Nb_3$  alloys. The saturation magnetization ( $\mu_0 M_s$ , where  $\mu_0$  is the permeability of a vacuum)

**Table 1.** Saturation magnetization ( $\mu_0 M_s$ ), magnetic flux density at 800 A/m ( $B_{800}$ ), initial permeability ( $\mu_i$ ), coercivity ( $H_c$ ) and core losses ( $P_{cm}$ ) of  $(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$  and  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  bulk metallic glasses of 0.50 mm in thickness. The data of Fe-6.5 mass% Si alloy with the same thickness are also shown for comparison [14].

	$\mu_0 M_s$ (T)	$B_{800}$ (T)	$\mu_i$	$H_c$ (A/m)	$P_{cm}$ (W/kg)			
					1 T, 50 Hz	1 T, 400 Hz	0.2 T, 1 kHz	0.2 T, 10 kHz
$(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$	1.20	1.18	33,000	4.0	0.24	10.8	1.17	57
$(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$	1.35	1.31	7,400	8.2	0.69	12.8	2.14	90
Fe-6.5 mass% Si <sup>a</sup>	1.80	1.27	58,000 <sup>b</sup>	~ 6	0.58	15.6	2.80	106

<sup>a</sup>Ref. 14.

<sup>b</sup> $\mu_{max}$ .

of  $(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$  (the base alloy) is only 1.20 T. On the other hand,  $\mu_0 M_s$  of the  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  alloy with optimized composition have been increased up to 1.35 T.

The magnetic properties of the  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  BMG are summarized in Table 1. The data of Fe-6.5 mass% Si steel (6.5Si) with the same thickness are also shown for comparison [14]. It should be noted that the magnetic flux density of the BMG at 800 A/m ( $B_{800}$ ) is slightly larger than that of the 6.5Si. Furthermore, the core losses of the BMG are approximately 20%-30% smaller than those of the 6.5Si.

## 4. Summary

The reduction of the Nb content in the  $(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$  BMG has been studied to increase the magnetization. In order to obtain the ring-shaped bulk specimens (0.50 mm thick) with a single glassy phase, a Nb content of at least 4 at% is required for Co-free alloys. However, the GFA increases by replacing partially Fe by Co. Furthermore, the saturation magnetization is improved from 1.20 T for the base alloy  $(\text{Fe}_{0.75}\text{B}_{0.20}\text{Si}_{0.05})_{96}\text{Nb}_4$  to 1.35 T for the  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  alloy, by optimizing the B and Si contents. It should be noted that the  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  BMG exhibits slightly larger  $B_{800}$  and 20%-30% smaller core losses compared with Fe-6.5 mass% Si. Therefore, it can be concluded that  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  BMGs have high potential to be used as core materials for various magnetic devices. The good magnetic properties of  $(\text{Fe}_{0.8}\text{Co}_{0.2})_{76}\text{B}_{18}\text{Si}_3\text{Nb}_3$  BMGs bring greater efficiency and miniaturization to magnetic devices.

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