

# Improving the Soft Magnetic Properties of Fe-Co-B-Zr-Nb Alloy by Tuning the Fe/Co Ratio

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**Amorphous  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_y\text{Nb}_{10-y}$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$  and  $y = 7$  or  $8$  at.%) ribbons were fabricated by the melt-spinning technique, and their thermal and magnetic properties were investigated. The results showed that all alloys had a fully amorphous structure, and the substitution of Zr for Nb resulted in high thermal stability and high saturation flux density ( $B_s$ ) with fairly low coercivity ( $H_c < 21$  A/m). Moreover, by varying the Fe/Co ratio, the optimum ratio with the highest  $B_s$  and low  $H_c$  was determined. As the Fe/Co ratio changed from 6:4 to 9:1,  $H_c$  decreased from approximately 20 to 9 A/m, and  $B_s$  also decreased gradually. A saturation flux density of 1.33 T was achieved with a coercivity of 9 A/m after optimization.**

**Keywords :** Fe-based, amorphous, soft magnetic material, ribbon

## 1. Introduction

Soft magnetic materials are essential for the efficient operation of next-generation power electronics and electrical machinery such as motors, generators, and transformers [1, 2]. Further, the demand for high efficiency, automation, and miniaturization of devices is increasing the use of soft magnetic materials with low core loss, high magnetic flux density ( $B_s$ ), and high permeability ( $\mu_c$ ) [3-5]. Over the past few decades, various types of soft magnetic materials have been used, including electrical steel, Fe, Ni-Zn/Mn-Zn ferrite, Fe-Ni, Fe-Si, Fe-Si-Al, nanocrystalline alloys, and metallic glassy metals [3, 6, 7].

Amorphous alloys have attracted much attention owing to their unique atomic arrangement that differs from those of typical crystalline materials [8, 9]. This unique arrangement affords a combination of mechanical characteristics, including high breaking strength, high elasticity, high wear resistance, high corrosion resistance, and excellent soft magnetic properties [10-12]. Furthermore, Fe-based amorphous/nanocrystalline alloys have approximately 100 times higher permeability, 100 times lower coercive force, and five times lower iron loss compared to existing crystalline soft magnetic materials such as Fe-Si and Fe-Si-Al alloys [13, 14]. Therefore, they are emerging as

optimal materials for producing high-performance and high-efficiency magnetic parts. Amorphous and nanocrystalline materials are constantly being improved through increases in  $B_s$  and the introduction of new alloys that are more amenable to fabricating large-scale parts [1, 15-18]. Well-known alloys include FINEMET (Yoshizawa *et al.*, 1988), NANOPERM (Suzuki *et al.*, 1988), HITPERM (Willard *et al.*, 1998), and NANOMET (Makino *et al.*, 2011). Among these, the Fe-Co-Zr-B-Cu alloy HITPERM exhibits high  $B_s$  and better soft magnetic properties than those of other families of materials at high temperatures [2, 19-21]. Škorvánek *et al.* experimentally demonstrated that Cu-free HITPERM exhibits a remarkably lower  $H_c$  than that of the original HITPERM [22]. Therefore, copper is not necessary to form the ultrafine structure of HITPERM. Additionally, an appropriate composition of Zr and Nb can be used to control the thermal stability while maintaining the  $B_s$ ,  $\mu_c$ , and mean grain size [23, 24].

The present study investigates the effects of Zr substitution for Nb with varying Fe/Co ratios in  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_y\text{Nb}_{10-y}$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$  and  $y = 7$  or  $8$  at.%) alloys on the magnetic and thermal properties.

## 2. Experimental Procedure

Alloys with a nominal composition of  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_y\text{Nb}_{10-y}$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$  and  $y = 7$  or  $8$  at.%)

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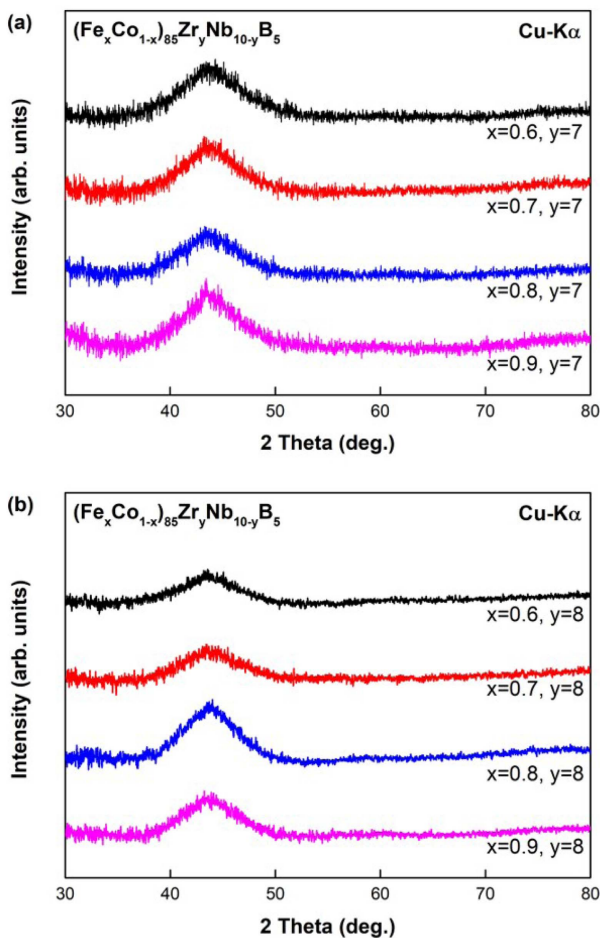
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were studied. The ingots were prepared by arc melting under Ar atmosphere with high-purity Fe (99.95 %), Co (99.95 %), Zr (99.5 %), Nb (99.95 %), and B (99.5 %). All ingots were remelted at least four times to ensure that the alloys were homogeneously mixed. The melt-spinning technique was used to fabricate rapidly solidified amorphous ribbons having a width and thickness of 2 mm and 20-30  $\mu\text{m}$ , respectively, with a roller speed of 56.3 m/s. Differential scanning calorimeter (DSC) was used to evaluate thermal stability at a heating rate of 0.34 K/s under Ar gas flow. The structural property of the amorphous ribbons was characterized by X-ray diffraction (XRD) with Cu-K $\alpha$  radiation. The magnetic measurement was performed using a vibrating sample magnetometer (VSM) and DC B-H loop tracer. The saturation magnetization ( $M_s$ ) was measured using a VSM under an in-plane applied field ranging from -800 to 800 kA/m, and  $H_c$  was measured using a DC B-H loop tracer.

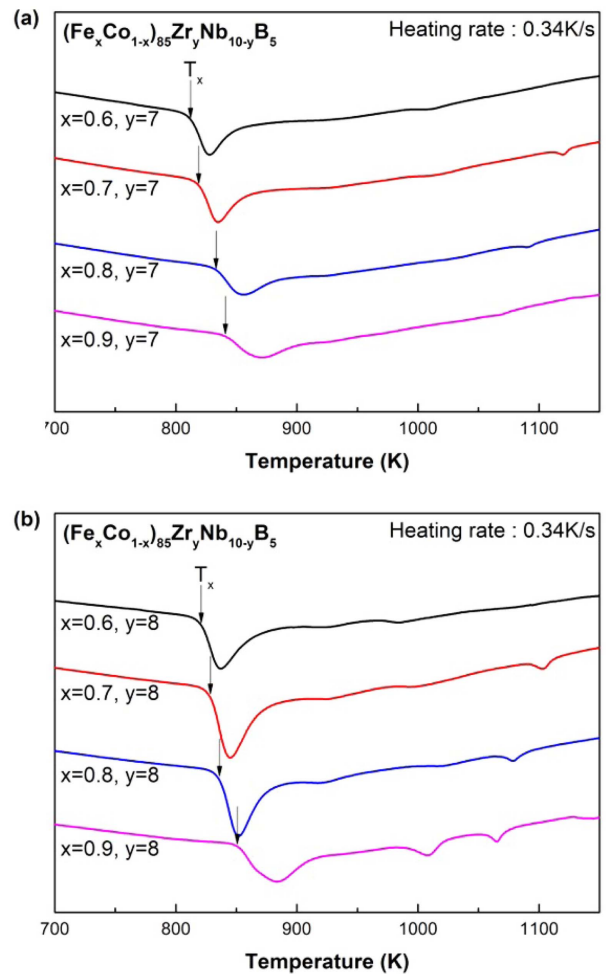
### 3. Results and Discussion

Figure 1 shows the X-ray Diffraction patterns of the melt-spun  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_y\text{Nb}_{10-y}$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$  and  $y = 7$  or  $8$  at.%) alloys with varying Fe/Co ratios. Broad diffraction peaks were observed at diffraction angles of  $2\theta \approx 44^\circ$ . No peaks correspond to any crystalline phase in all alloys. Fe/Co ratios of 6:4 to 9:1 had no effect on the amorphization procedure, and the alloy system retained an amorphous structure.

Figure 2 shows the DSC curves of the  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_y\text{Nb}_{10-y}$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$  and  $y = 7$  or  $8$  at.%) ribbons. All alloys show distinct exothermic peaks, indicating the precipitation of the  $\alpha$ -Fe phase [25, 26]. Figure 2 shows the crystallization onset temperatures ( $T_x$ ) of the alloys. With increasing Fe content, the  $T_x$  of  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_7\text{Nb}_3$  (alloys with  $y = 3$  at.% and  $x = 0.6, 0.7, 0.8,$  or  $0.9$ , hereinafter denoted as Zr7 alloys) and



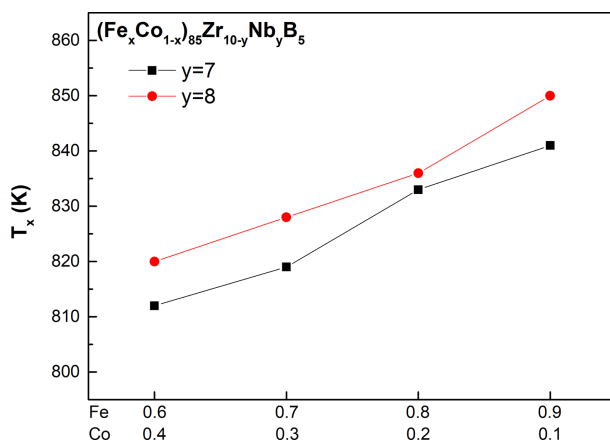
**Fig. 1.** XRD patterns of (a)  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_7\text{Nb}_3$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$ ) and (b)  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_8\text{Nb}_2$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$ ) as-spun ribbons.



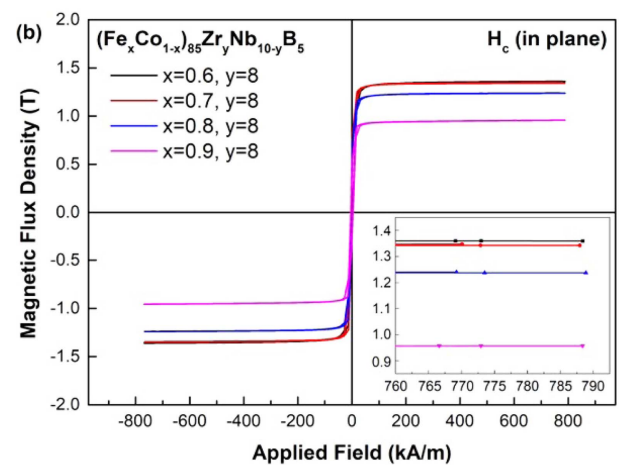
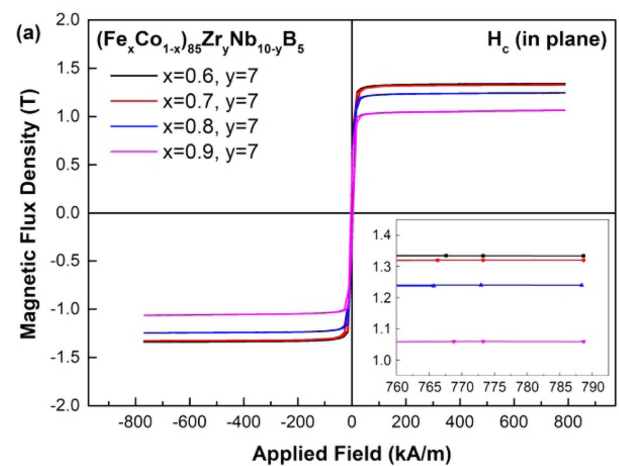
**Fig. 2.** DSC curves of (a)  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_7\text{Nb}_3$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$ ) and (b)  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_8\text{Nb}_2$  ( $x = 0.6, 0.7, 0.8,$  or  $0.9$ ) as-spun ribbons.

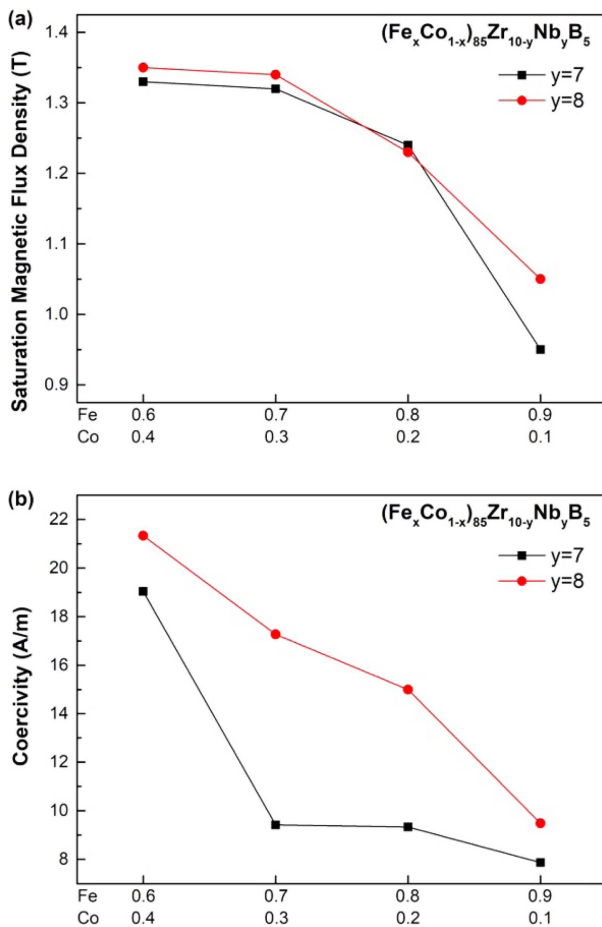
**Table 1.** Thermal and magnetic properties of as-spun  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_{10-y}\text{Nb}_y$  ( $x = 0.6, 0.7, 0.8, \text{ or } 0.9$  and  $y = 2$  or  $3$  at.%) ribbons.

Fe/Co Ratio	Alloys					Thermal Properties		Magnetic Properties	
	Alloy Composition					$T_x$ (K)	$H_c$ (A/m)	$M_s$ (emu/g)	$B_s$ (T)
6:4	Fe <sub>51</sub>	Co <sub>34</sub>	Zr <sub>8</sub>	Nb <sub>2</sub>	B <sub>5</sub>	820	21	139.80	1.35
7:3	Fe <sub>59.5</sub>	Co <sub>25.5</sub>	Zr <sub>8</sub>	Nb <sub>2</sub>	B <sub>5</sub>	828	17	137.05	1.34
8:2	Fe <sub>68</sub>	Co <sub>17</sub>	Zr <sub>8</sub>	Nb <sub>2</sub>	B <sub>5</sub>	836	15	128.37	1.23
9:1	Fe <sub>76.5</sub>	Co <sub>8.5</sub>	Zr <sub>8</sub>	Nb <sub>2</sub>	B <sub>5</sub>	850	9	100.15	1.05
6:4	Fe <sub>51</sub>	Co <sub>34</sub>	Zr <sub>7</sub>	Nb <sub>3</sub>	B <sub>5</sub>	812	19	135.78	1.33
7:3	Fe <sub>59.5</sub>	Co <sub>25.5</sub>	Zr <sub>7</sub>	Nb <sub>3</sub>	B <sub>5</sub>	819	9	135.70	1.32
8:2	Fe <sub>68</sub>	Co <sub>17</sub>	Zr <sub>7</sub>	Nb <sub>3</sub>	B <sub>5</sub>	833	9	128.57	1.24
9:1	Fe <sub>76.5</sub>	Co <sub>8.5</sub>	Zr <sub>7</sub>	Nb <sub>3</sub>	B <sub>5</sub>	841	7	110.86	0.95

**Fig. 3.** Variation of the crystallization temperature ( $T_x$ ) of the as-spun ribbons of  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_{10-y}\text{Nb}_y$  ( $x = 0.6, 0.7, 0.8, \text{ or } 0.9$  and  $y = 2$  or  $3$  at.%) ribbons.

$(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_8\text{Nb}_2$  (alloys with  $y = 2$  at.%) and  $x = 0.6, 0.7, 0.8, \text{ or } 0.9$ , hereinafter denoted as Zr8 alloys) ribbons increased from 812 to 841 K and from 820 to 850 K, respectively. Because  $T_x$  serves as a reference for the upper temperature limit for improving the magnetic properties, increasing the  $T_x$  value is highly desirable. In addition, a rapidly increasing  $T_x$  value is more likely to expand the supercooled region, which can increase the thermal stability. Based on DSC data, the crystallization temperature was plotted as a function of the Fe/Co ratios in Fig. 3. The graph shows that all  $T_x$  values for Zr8 alloys were higher than those for Zr7 alloys for a given Fe/Co ratio. Combined with previous research [27] that reports data for  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_9\text{Nb}_1$  ( $x = 0.6, 0.7, 0.8, \text{ or } 0.9$ ) alloys, increasing the Zr content in the Fe-Co-B-Zr-

**Fig. 4.** B-H curves of (a)  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_8\text{Nb}_2$  ( $x = 0.6, 0.7, 0.8, \text{ or } 0.9$ ) and (b)  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_7\text{Nb}_3$  ( $x = 0.6, 0.7, 0.8, \text{ or } 0.9$ ) ribbons in as-quenched state.



**Fig. 5.** Dependence of  $B_s$  and  $H_c$  on Fe/Co ratio of  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{Zr}_{10-y}\text{Nb}_y\text{B}_5$  ( $x = 0.6, 0.7, 0.8, \text{ or } 0.9$  and  $y = 2$  or  $3$  at.%) melt-spun ribbons.

Nb system can increase the crystallization temperature.

Figure 4 shows the B-H hysteresis loops of the  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_{10-y}\text{Nb}_y$  ( $x = 0.6, 0.7, 0.8, \text{ or } 0.9$  and  $y = 7$  or  $8$  at.%) melt-spun ribbons. All curves exhibit the typical squared loop of soft magnetic materials. The saturated parts of the graphs are enlarged in the upper left corner to compare  $B_s$  values with the Fe/Co ratio. Figure 5 shows the  $B_s$  and  $H_c$  values of  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_{10-y}\text{Nb}_y$  ( $x = 0.6, 0.7, 0.8, \text{ or } 0.9$  and  $y = 7$  or  $8$  at.%) ribbons as functions of the Fe/Co ratio. Overall, Zr8 alloys showed higher  $B_s$  values compared with those of Zr7 alloys; however, in the case of  $H_c$ , Zr7 alloys have lower values for a fixed Fe/Co ratio.  $B_s$  shows an increasing trend as the Fe:Co ratio changes from 9:1 to 6:4. Specifically, it changes from 0.95 to 1.33 T for Zr7 alloys and from 1.05 to 1.35 T for Zr8 alloys, indicating that the highest  $B_s$  is achieved for an Fe/Co ratio of 6:4 for both Zr7 and Zr8 alloys. Furthermore,  $H_c$  shows a downward trend with increasing Fe content (Fig. 5); it decreases from 19 to

7 A/m (Zr7 alloys) and from 21 to 9 A/m (Zr8 alloys). In particular, for Zr7 alloys, when the Fe/Co ratio is changed from 6:4 to 7:3,  $B_s$  remains almost unchanged, whereas  $H_c$  decreases greatly; these alloys show the best soft magnetic properties.

## 4. Conclusion

We investigated changes in the amorphous forming ability and soft magnetic properties of Zr substituted for Nb in Fe-Co-B-Zr-Nb amorphous ribbons. Further, we varied the Fe/Co ratios to optimize the thermal and magnetic properties, and we drew the following conclusions.

The effects of substituting Zr for Nb on the  $T_x$ ,  $B_s$ , and  $H_c$  values were observed. For all Fe/Co ratios,  $T_x$  and  $B_s$  increased with slightly increased  $H_c$ . Therefore,  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_7\text{Nb}_3$  alloys have better thermal and magnetic characteristics than those of  $(\text{Fe}_x\text{Co}_{1-x})_{85}\text{B}_5\text{Zr}_8\text{Nb}_2$  alloys.

As the Fe/Co ratio changes from 6:4 to 9:1,  $T_x$  increases by 30 K, indicating that the Fe/Co ratio affects the thermal properties of the alloys, and that the thermal stability improves as the Fe content increases.

$B_s$  and  $H_c$  both decrease as the Fe/Co ratio is varied from 4:6 to 9:1. The highest  $B_s$  value of 1.35 T is achieved when the Fe/Co ratio is 6:4; however, when the Fe/Co ratio reaches 7:3,  $B_s$  decreases slightly and  $H_c$  increases significantly, resulting in the optimum soft magnetic properties. For further improvement, heat treatment can be performed under appropriate conditions to reduce  $H_c$  and increase  $B_s$ .

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