

EFFECT OF Zr AND Nb ON THE ELECTRICAL AND MAGNETIC PROPERTIES OF THE Fe-Zr-Nb-B-Cu ALLOY

The present study, aims to investigate the effect of minor Zr and Nb alloying on soft magnetic and electrical properties of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys. The investigated alloys were prepared through the melt spinning process. Within the examined compositional range (Nb up to 5.25at%, respectively), the soft magnetic properties and electrical resistivity of the alloys continuously increase with increasing Nb content. However increasing the Nb content further decreases such properties. We could confirm the influence of ratio of Zr and Nb on grain growth and crystallization fraction during crystallization by using the soft magnetic properties and electrical properties.

Keywords: Soft magnet, Magnetic properties, Amorphous alloy, Nanocrystalline alloy, Electrical properties

1. Introduction

To increase high driving current and achieve miniaturization in wireless communication devices, the core of a power inductor requires high permeability and saturation magnetization at a high frequency range. However, the soft magnetic properties of metal materials decrease with increasing frequency because of their low electric resistance. Nanocrystalline alloys have been intensively investigated in the last decades because of their excellent soft magnetic properties [1]. Nanoperm alloy is a type of Fe-based soft magnetic alloy that exhibits excellent magnetic properties. Generally, the Nanoperm alloy is prepared through crystallization of amorphous precursors [2]. The soft magnetic properties of these materials is related to the macroscopic decrease in the magnetocrystalline anisotropy and very low magnetostriction. The low value of anisotropy is a consequence of the small grain size (10-15 nm) and random orientation of α -Fe crystallites [3]. The decreased value of effective magnetostriction can be achieved when the positive contribution of residual amorphous matrix is balanced by the negative contribution of the crystallites [4]. As reported earlier, transition metals and Cu are rejected from the primary α -Fe particles and partitioned in the residual amorphous matrix owing to the negligible immiscibility of transition metal in α -Fe. In addition, the existence of transition metal leads to the grain refinement of nanocrystalline alloys [4,5]. In fact, during the annealing of transition metals containing nanocrystalline alloys, transition-metal segregation in the residual amorphous matrix stabilizes the remaining amorphous phase and controls the grain size [5-7]. In this study, we investigated the influence of

Zr/Nb ratio change on the soft magnetic properties and electrical properties of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys.

2. Experimental

In this study, a Master alloy with the nominal composition of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) was prepared through arc melting in a Ti-gettered argon atmosphere. The composition of the alloy and the ratio of the transition metal are shown in Table 1. The mixture of pure Fe, Zr, Nb, Cu (purity 99.95%) and Fe-B (16at% B) were used as raw materials. Mother alloys were melted repeatedly at least four times to achieve homogeneity and were rapidly solidified (3 mm wide and 25-35 μm thick) in an argon atmosphere through the melt spinning technique, in which a copper roller with a diameter of approximately $\Phi 250$ was rotated at a constant circumferential speed of 48 m/s. The ribbons were annealed in the argon atmosphere at different temperatures ranging from 480 to 530°C for 1 h. The structure of the as-quenched and annealed ribbons was determined through X-ray diffractometry (XRD) using Cu K_α ($\lambda = 1.54056\text{\AA}$) radiation. Thermal analysis was conducted and the crystallization behavior of the amorphous alloys during continuous heating was determined through the differential scanning calorimetry (DSC) at a heating rate of 20°C/min. The saturation magnetization (M_s) and coercivity (H_c) were measured using a vibrating sample magnetometer (VSM). The electrical resistivity (ρ) and electrical conductivity of the alloys were measured using the two-point probe technique.

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TABLE 1

Composition of mother alloys and ratio of transition metal

	Fe	Zr	Nb	B	Cu	Zr/Nb ratio
Composition 1	86	7	0	6	1	1:0
Composition 2	86	5.25	1.75	6	1	0.75:0.25
Composition 3	86	3.5	3.5	6	1	0.5:0.5
Composition 4	86	1.75	5.25	6	1	0.25:0.75

3. Results and discussion

Fig. 1 shows the DSC curves of the as-quenched $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys. The characteristic temperatures and the glass-forming-ability parameters of the alloys, including the glass transition temperature (T_g), and onset temperature (T_x) of crystallization, are in the 350–500°C range. In particular, the glass-transition temperature is in the range of 362 to 393°C. All the compositions shows similar thermal properties. This indicates that Zr/Nb ratio do not affect thermal properties of alloy. The reason for this is considered because the atomic size of Zr and Nb are similar.

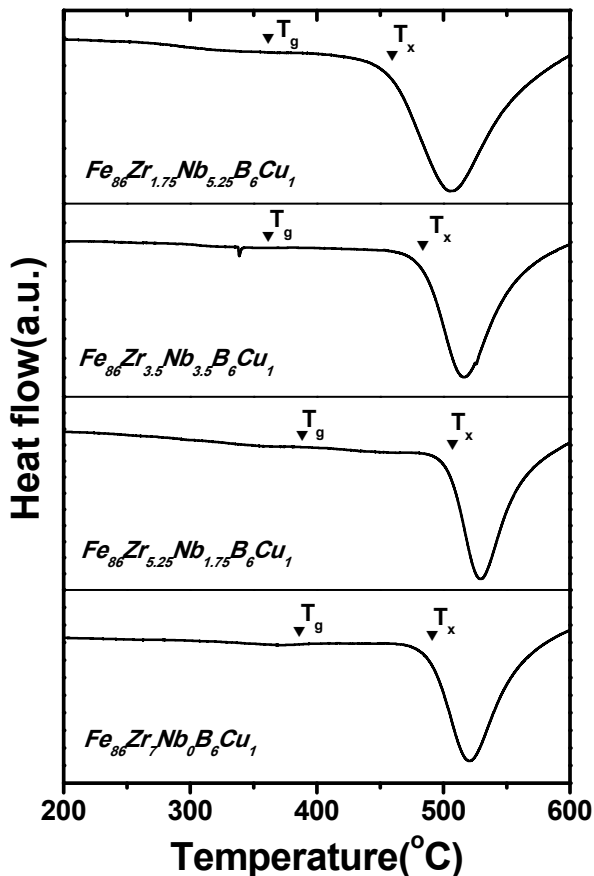


Fig. 1. DSC curve of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) amorphous alloy ribbons at the heating rate of 20 K/s

Fig. 2 shows the XRD patterns of the as-quenched and annealed (at 1st crystallization temperature) $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys. The heat treatment was conducted at the first crystallization temperature of the alloys. Fig. 2(a)

shows halo patterns without any distinct diffraction peaks indicating the formation of a fully amorphous phase. Fig. 2(b) shows the XRD spectrum of the $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloy annealed for 1 h at 457° to 503°C (which corresponds to the temperature just above the exothermic peak). The diffraction pattern is in good agreement with the body-centered cubic α -Fe.

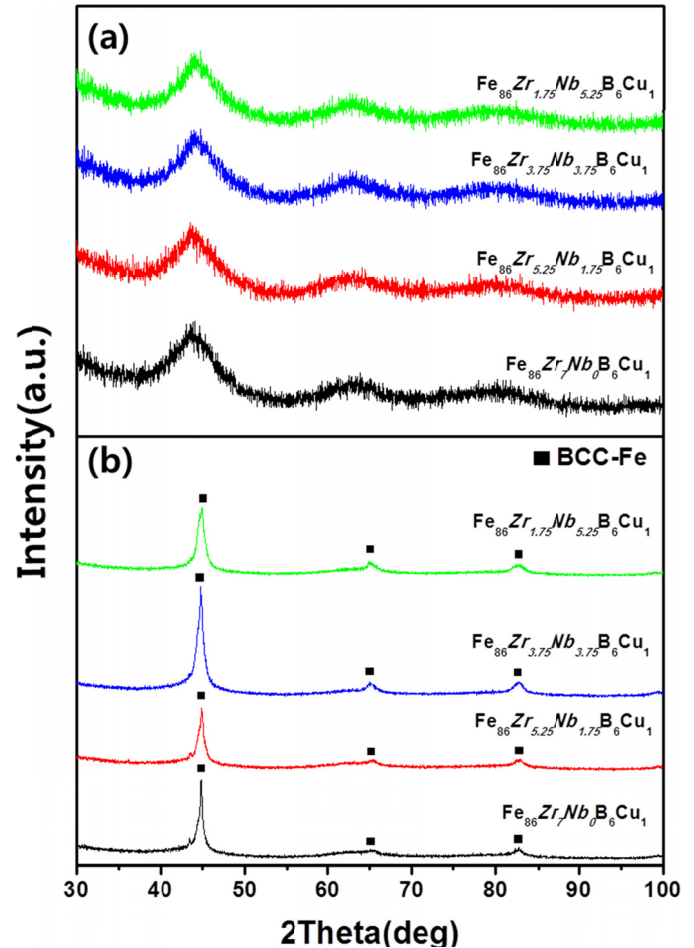


Fig. 2. (a) X-ray diffraction pattern of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) amorphous alloy (b) annealed for 3600 s

As shown in Fig. 3, the saturation magnetization of the $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys from 158.28 to 187.36 emu/g. The highest value of saturation magnetization of 187.36 emu/g was obtained for the composition of $\text{Fe}_{86}(\text{Zr}_{0.5}\text{Nb}_{0.5})_7\text{B}_6\text{Cu}_1$, for which the Zr/Nb ratio was 1:1. Although the overall composition shows a similar value of coercivity i.e., 3.5 Oe, the coercivity of the magnetic alloy depends on the size of the crystal phase. To calculate the initial permeability, $M-H$ curve was converted into a $B-H$ curve by using the following formula.

$$B = H + 4\pi M$$

The value of initial permeability of the alloys was determined through the magnetic flux density found in the formula. The variation of the initial permeability of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys is shown in Fig. 4. It is notable

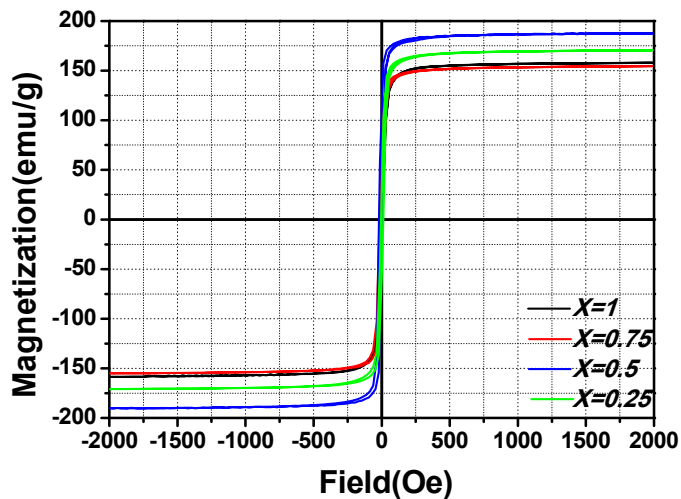


Fig. 3. $M-H$ hysteresis loop of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys annealed for 3600 s

that the alloys exhibit a distinct change in initial permeability with the alternation of the Zr/Nb ratio. With 1:0, 0.75:0.25, and 0.5:0.5, the initial permeability significantly increases showing a value in the range of 2781-9268. In contrast, the composition of Zr/Nb ratio of 0.75:0.25 showed a lower value: 4004. The properties of magnetically soft Fe-based nanocrystalline alloys are said to be the most structure sensitive [9]. The volume fraction of nanocrystals should be the fraction that compensates their negative magnetostriction contribution through the positive magnetostriction contribution of the amorphous matrix. A non-magnetostrictive Fe-based nanocrystalline material can be obtained for $V_{cr} = 70\%-75\%$, depending on an alloy composition. The transition metal hinders the growth of crystals at the crystallization stage of the amorphous system [8]. The transition metal seems to influence the crystal fraction of a nanocrystalline alloy by hindering the crystal grain growth, resulting in the difference of the soft magnetic properties of the alloys.

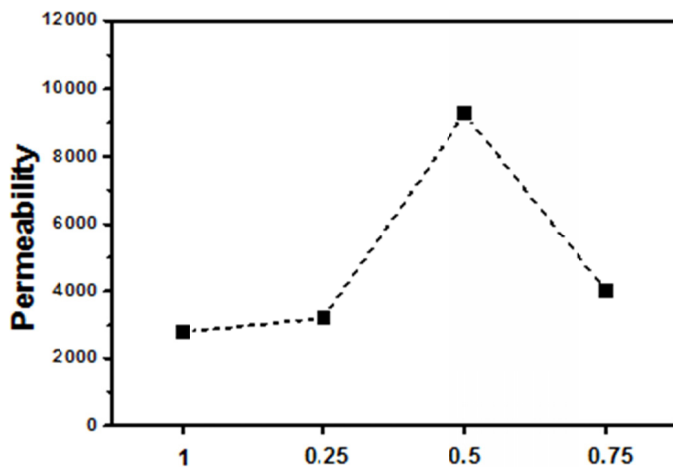


Fig. 4. Initial permeability of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys annealed for 3600 s

Lastly, the electrical properties of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys were measured. As shown in Fig. 5,

the electrical resistivity of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys ranges from 1.89×10^{-3} to $2.95 \times 10^{-3} \Omega \text{ cm}$. The electrical resistivity showed the same tendency as the saturation magnetization and magnetic flux density of the alloys. The amorphous materials have irregular atomic arrangement, and therefore do not allow the free electron to pass freely, implying a higher electrical resistivity. In contrast, the addition of the nanocrystalline phase allows the free electron to pass freely, thus decreasing the resistivity of the alloys. Therefore, this result suggests that the crystallization ratio based on the ratio of Zr and Nb (which are transition metals) has a strong influence on the electrical properties of these alloys. Generally, the decrease in grain size decreases the coercivity; however, in our case, the coercivity remained the same in all the compositions; this is probably due to the addition of transition metals in the alloy system.

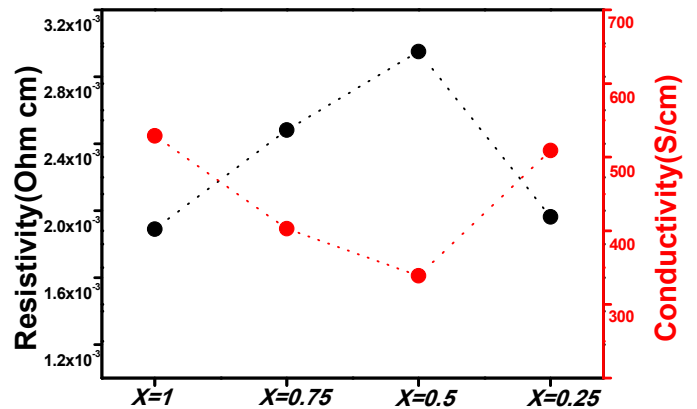


Fig. 5. Electrical properties of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys annealed for 3600 s

4. Conclusions

In this paper, we investigated the effects of Zr and Nb on the soft magnetic properties and electrical properties of $\text{Fe}_{86}(\text{Zr}_x\text{Nb}_{1-x})_7\text{B}_6\text{Cu}_1$ ($x = 1, 0.75, 0.5, 0.25$) alloys and the following conclusions can be drawn.

- 1) By changing the ratio of Zr and Nb, we showed the difference in the soft magnetic properties of the alloy. In particular, the change in the values of M_s , B_s , and initial permeability, which shows highest value with $\text{Fe}_{86}(\text{Zr}_{0.5}\text{Nb}_{0.5})_7\text{B}_6\text{Cu}_1$ composition, was shown.
- 2) We deduced a correlation between the magnetic and electric properties of the alloy. The electrical resistivity of the alloys showed a contrasting behavior to the soft magnetic properties.

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