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# Effects of annealing condition and Al content on novel $Fe_{73.5}Si_{13.5}B_9Cu_1Nb_{3-x}Al_x$ alloys

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**Abstract**

The annealing condition, Al content, and field amplitude dependences of the complex permeability for Nb-poor Finemet type alloys, $Fe_{73.5}Si_{13.5}B_9Cu_1Nb_{3-x}Al_x$ ($x$ = 0, 0.5, 1.5, 2.0, and 3.0), were investigated using an impedance analyzer and X-ray diffraction. The results show that different Al contents lead to different optimum annealing conditions, and the Al content exerts a distinct effect on microstructure thus resulting in a variety of real permeability value. For the samples annealed at 793 K for 0.5 h, the real permeability increases with an increase in Al content when the Al content is below 2.0 at.%; as for those annealed at 793 K for 1 h and at 813 K for 0.5 h, an overall increase in real permeability can be obtained compared to those annealed at 793 K for 0.5 h. The permeability under different field amplitudes is also studied and it is found that the relaxation frequency in the lower frequency region tends to moving toward a higher frequency with an increase in field amplitude. All these might be because of the role of Nb in the annealing process and the solubility of Al in the amorphous matrix and nanocrystallized crystallites.

*Keywords:* magnetism; magnetic property; impedance analysis; Fe-based alloys

## 1. Introduction

An increasing interest in nanocrystalline soft magnetic materials is observed in recent years because of their excellent soft magnetic properties and potential applications. Finemet is one among the outstanding alloys that possess an excellent soft magnetic property such as high permeability [1]. The excellent soft magnetic behavior is the result of the averaging out of magnetocrystalline anisotropy via magnetic coupling between the grains through the remaining ferromagnetic amorphous phase matrix [2]. These magnetic properties strongly depend on the grain phase and its average grain size $D$. The changing of the constituent elements and annealing condition are two main methods to modify the grain phase and $D$, so the magnetic properties can be tailored. In Finemet, the magnetic grain phase is α-FeSi crystallite, and Nb is generally considered as an essential composition element to inhibit crystallite growth, so the crystallites can be formed in a nanoscale [3]. However, the application of Finemet is not as popular as expected. One of the reasons is the high price of Nb. Therefore, transition metals and other elements have been used to substitute Nb [4-6], but no significant improvements have been reported. Because of the similar covalent radius between Al and Si, it is expected that Si can be partially replaced by Al in α-FeSi crystallites, and Tate *et al.* [7] reported that ternary FeSiAl crystals exhibit a lower crystalline anisotropy than the binary FeSi system. The appearance of α-FeSi(Al) in Finemet will lead to an increase in permeability, according to a random anisotropy model (RAM). The partial substitution of Fe or Si by Al has been reported and it shows that the coercivity, the saturation magnetization, and the crystallization temperature are decreased [7-10]. Although the appearance of α-FeSi(Al) and thus the improvement of soft magnetic properties are expected by the substitution of Al for Nb, this substitution has scarcely been researched.

In the present research, novel Finemet type alloys have been prepared by the substitution of Nb by Al and the effects of the annealing condition as well as Al content have been investigated.

## 2. Experimental

Nominal composition $Fe_{73.5}Si_{13.5}B_9Cu_1Nb_{3-x}Al_x$ ($x$ = 0,

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0.5, 1.5, 2.0, and 3.0) alloys were prepared by using a melt-spinning method on a Cu wheel in an argon atmosphere [11]. The ribbons were approximately 0.8-1 mm in width and 20 μm in thickness. The velocity of the Cu wheel was about 45 m/s. The samples were made by winding the ribbons on toroidal sample holders of 5 mm in diameter, made from ceramics, and were subsequently annealed at 793 K for 0.5 h and 1 h, at 813 K for 0.5 h, respectively, and then quenched in Ar atmosphere. The complex permeability of the samples, $\mu = \mu' - j\mu''$, was measured in a frequency range from 20 Hz to 1 MHz by an impedance analyzer (Agilent 4284A). Here, $\mu'$ and $\mu''$ were the real part and imaginary part of the complex permeability. They represented the magnetic energy storage and loss in the dynamic magnetization process, respectively. The structures of the ribbons were characterized by X-ray diffraction (XRD) with Cu $K_\alpha$ radiation. The grain size was estimated from X-ray diffraction patterns by using the Scherrer formula from the full width at half maximum (FWHM) of the peaks.

## 3. Results and discussion

The frequency dependence of the complex permeability of the samples were measured under an applied AC field of 0.265 A/m. Fig. 1(a) indicates the frequency dependence of the real part of the complex permeability (real permeability, $\mu'$) of the samples annealed at 793 K for 0.5 h. It can be seen that all the samples (except $x$ = 3.0) exhibit a similar behavior, although the values are different. $\mu'$ is almost constant and independent of the frequency in the low frequency region. As the frequency further increases, however, $\mu'$ decreases gradually; with the Al content increasing, an increase of $\mu'$ can be seen at this annealing condition. However, the permeability deteriorates for the Nb sample totally substituted by Al. To get a clear picture of the influence of the annealing condition, the frequency dependence of the real permeability of the sample with 0.5 at.% Al annealed at different annealing conditions is depicted in Fig. 1(b), and the inset shows the increase of $\mu'$ annealed at 793 K for 1 h and at 813 K for 0.5 h compared to that at 793 K for 0.5 h. It can be seen that $\mu'$ increases after a longer time or a higher temperature annealing. They result in higher permeability. The optimum annealing condition among these three annealing conditions should be 793 K for 1 h. Moreover, for the samples annealed at 793 K for 1 h, the increase of $\mu'$ is the biggest when the Al content is 0.5 at.%, but it decreases with the Al content further increasing. As for the samples annealed at 813 K for 1 h, the $\mu'$ value increases slightly with less Al content, and the sample with 1.5 at.% Al content has the maximum permeability; however, for the sample with 2.0 at.% Al, a slight decrease of $\mu'$ can be seen.

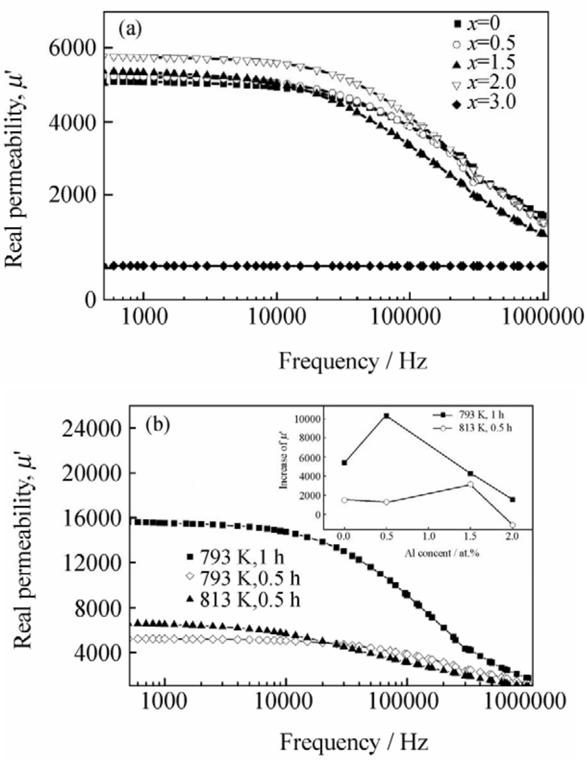


**Fig. 1. Dependences of real permeability spectra on Al content (a) and annealing condition (b): (a) annealed at 793 K for 0.5 h; (b) $x$ = 0.5, annealed at different conditions. The inset gives the increase of $\mu'$ for $Fe_{73.5}Si_{13.5}B_9Cu_1Nb_{3-x}Al_x$ ($x$ = 0, 0.5, 1.5, and 2.0) ribbons annealed at 793 K for 1 h and at 813 K for 0.5 h compared to that annealed at 793 K for 0.5 h (measured at 0.265 A/m and 1 kHz).**

The magnetic permeability is determined by a combination of chemistry, structure, and morphology (sample shape or grain shape). The variation in permeability is thus related to the microstructure of the alloys, and the crystallization products have been evaluated by XRD. Fig. 2 shows the XRD pattern of the samples. It indicates that the nanocrystallized α-FeSi or α-FeSi(Al) crystallites appear after annealing for every sample, and the iron boride phase appears subsequently, when the Al content is up to 2.0 at.%. The diffraction peak intensity increases with an increase in Al content, indicating a higher crystalline volume fraction ($V_{cry}$), with an increase in Al content [12]. The grain size $D$ is estimated by the Scherrer formula and the inset of Fig. 2 shows the variation in $D$. An overall increase of $D$ with the Al content can be seen, and $D$ is around 10 nm when the Al content is below 1.5 at.%, increases abruptly when the Al content is above 1.5 at.%, and then reaches a maximum value when Nb is totally substituted by Al. It is because that with more substitution of Nb by Al, the Nb content decreases and the crystallized process becomes easier because of less Nb to retard the crystallite growth, which leads to a relatively larger $D$ and a higher $V_{cry}$.