Electromagnetic Wave Absorption Characteristics of Nanocrystalline FeCuNbSiB Alloy Flakes/Polymer Composite Sheets with Different Flake Thickness

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This study examined the effects of a decrease in thickness of magnetic alloy flakes on the electromagnetic wave absorption characteristics of nanocrystalline $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ (at.%) alloy flakes/polymer composite sheets available for a quasi-microwave band. The thickness of FeCuNbSiB alloy flakes decreased to 1-2 μm with increasing milling time up to 24 h, and the composite sheet including alloy flakes milled for 24 h exhibited considerably enhanced power loss properties in the GHz range compared to the sheets having non-milled alloy powders. Although a considerable increase in loss factor upon milling was observed in the narrow frequency range of 4-6 GHz, there was no correlation between the complex permeability and flake thickness. However, the complex permittivity increased with increasing milling time, and there was good agreement between the milling time and the frequency dependences of the complex permittivity and power loss.

Keywords : electromagnetic wave absorption, nanocrystalline FeCuNbSiB alloy, composite sheets, flake thickness

1. Introduction

An increase in operation frequency and the highly integrated configuration of the electronic component parts have been realized with the rapid progress of high-speed processing and miniaturization of digital electronic equipment, such as personal computers and wireless communication system etc.. As a result, the frequency range of their electromagnetic noise has increased to the GHz range, and many serious problems of electromagnetic interference (EMI), including significant increases in the radiation and reflection of internal noise as well as cross-talk between circuit boards, have also been observed [1].

As one of technical responses to solve these EMI problems, composite sheets, where thin soft magnetic alloy powders, such as iron alloys, sendust, and silicon steel flakes, etc., are embedded in a dielectric polymer, have been recently developed [2-9].

These composite sheets are located near the noise sources and absorb the incident electromagnetic wave energy through magnetic loss.

The internal energy loss in an electromagnetic wave absorber that transforms the electromagnetic wave to heat energy via several loss mechanisms can be expressed using the following equation [10].

$$P = \pi f \mu''|H|^2 + \pi f \varepsilon''|E|^2 + 1/2 \cdot \sigma |E|^2 \quad (1)$$

where P, E, H, f, and σ are the electromagnetic wave absorption energy, electric field, magnetic field, frequency, and electrical conductivity, respectively. μ'' and ε'' are the imaginary part of the complex permeability ($\mu = \mu' - j\mu''$) and complex permittivity ($\varepsilon = \varepsilon' - j\varepsilon''$).

Therefore, magnetic loss-type absorbers (or sheets) using soft magnetic alloy powders need to have a high μ'' value for effective noise suppression and absorption.

Generally, the permeability of the magnetic materials decreases drastically in the GHz frequency range, which is higher than Snoek's limit resulting from natural resonance. A larger M_s is desirable to gain high complex permeability in the GHz range because the resonance frequency (f_r) at which the rapid drop in permeability takes place has a relationship with the saturation magnetization (M_s), gyromagnetic ratio (γ), and vacuum permeability (μ_0), as shown in Eq. (2) [11].

$$2\mu f_r \mu = 2\gamma M_s / 3\mu_0 \quad (2)$$

In order to obtain a high permeability in the GHz range, deformation of the soft magnetic alloy powders into thin flakes with a high aspect ratio is expected to be quite

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effective through the increase in f_r, which is derived from enhancement of shape anisotropy. In addition to the effect of an increase in f_r, the decrease in flake thickness up to the skin depth can possibly improve the magnetic properties at high frequencies by suppressing the eddy current flow and preventing formation of a closure domain.

From the above considerations, flakes of nanocrystalline (very fine grained in a scale of nanometers) FeCuNbSiB alloy, which has excellent soft magnetic properties at high frequency and large M_s over 1.2 T [12, 13], were used to develop an advanced composite sheet-type electromagnetic noise absorber available for the quasi-microwave band.

The relationship between the thicknesses of the flakes was examined according to the milling time, and the electromagnetic wave absorption characteristics were assessed.

2. Experimental Procedure

Amorphous $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ (at%) alloy powders as a precursor of nanocrystalline alloy flakes were prepared by pulverizing melt-spun ribbons with a thickness of 18 μm upon rotor-milling, and they were subsequently milled for 12 and 24 h using an attritor to fabricate the very thin flakes. The pulverized and milled alloy powders were classified into sizes less than 45 μm through mechanical sieving.

Subsequently, all amorphous flakes were annealed at 450 °C for 1 h in a vacuum to induce a transformation to a nanocrystalline state with a grain size of 10 nm or smaller [14], and they were then blended with polyurethane to a weight ratio of 65:35. Thin sheets, approximately 0.7 mm thick, were produced from the mixtures by tape casting.

The milling time dependence of the shape of the FeCuNbSiB alloy flakes and the cross-sectional views of the composite sheets were observed by scanning electron microscopy (SEM, JSM-8401).

The electromagnetic wave absorption characteristics of the nanocrystalline FeCuNbSiB alloy flakes/polymer composite sheets were evaluated by measuring the S parameter (reflection parameter S_{11}, transmission parameter S_{21}), complex permeability, and complex permittivity using a network analyzer (HP-8722D).

For obtaining the data of the S parameters, the samples were prepared in a rectangular shape, 5×5 cm in size, and set on a micro-strip line with a length, width and characteristic impedance of 79.53 mm, 2.24 mm and 50 Ω, respectively. The complex permeability and complex permittivity were measured in ring-type specimens whose inner and outer diameters were 3 and 7 mm, respectively.

3. Results and Discussion

Fig. 1 shows SEM images of amorphous FeCuNbSiB alloy flakes milled using an attritor for 0, 12, and 24 h. The thickness of the powder decreased largely by 12 and 24 h milling.

The X-ray diffraction patterns of the FeCuNbSiB alloy flakes showed a broad peak at approximately 45° of 2θ, as shown in Fig. 2, which indicates that the microstructure of the amorphous alloy annealed at 450 °C for 1 h had transformed to a bcc Fe-phase with very fine grains (i.e. nanocrystalline state).

Fig. 3 shows the cross-sectional views of the composite sheets, including the nanocrystalline FeCuNbSiB alloy

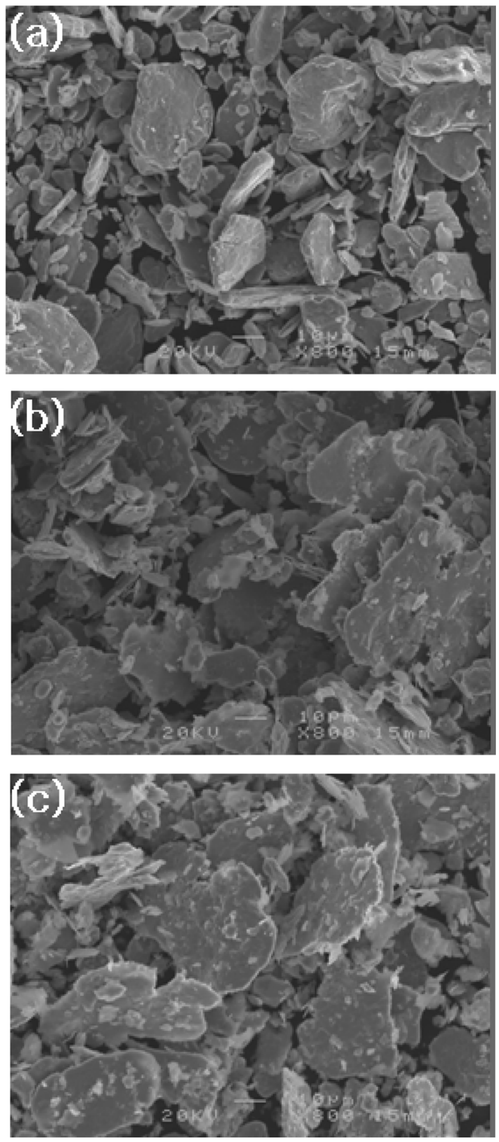

Fig. 1. SEM images of (a) amorphous FeCuNbSiB alloy powders milled by using an attritor for (a) 0 h, (b) 12 h, and (c) 24 h, respectively. (powder size <45 μm).

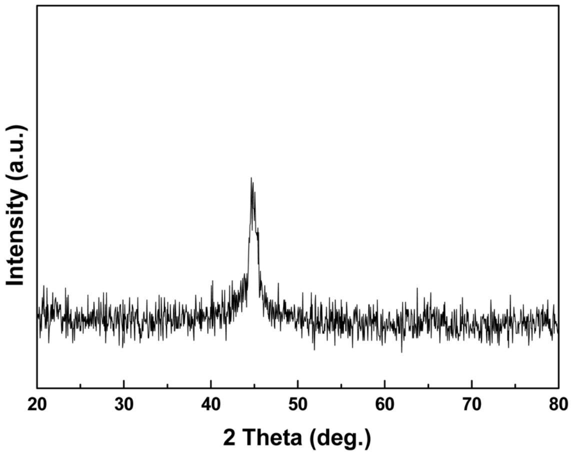

Fig. 2. X-ray diffraction pattern of the nanocrystalline FeCuNbSiB alloy flakes obtained by annealing the amorphous alloy powders at 450 °C for 1 h in a vacuum.

flakes milled for 0, 12, and 24 h. The thickness of the alloy flakes decreased from 18 μm to 3-4 μm after 12 h milling, and very thin flakes with a thickness of 1-2 μm were obtained after 24 h milling. In addition, the spacing between the alloy flakes and flake thickness decreased significantly.

Figs. 4 and 5 show the reflection parameter (S_{11}) and transmission parameter (S_{21}) of the composite sheets with flakes of different thickness, respectively. From the reflection coefficient Γ and transmission coefficient T of the electromagnetic wave in composite sheets, the S_{11} and S_{21} values were calculated using following equations [15].

$$S_{11} = 20 \log |\Gamma| \quad (3)$$

$$S_{21} = 20 \log |T| \quad (4)$$

As shown in Fig. 4, the reflection parameter, S_{11}, increased with increasing milling time (i.e. decrease in flake thickness) at frequencies lower than 1 GHz, while the frequency dependence of the S_{11} values were quite complicated at frequencies higher than 1 GHz. However, the S_{21} values decreased with increasing milling time at frequencies of 1-6 GHz.

From the S parameters presented in Figs. 4 and 5, the power loss (P_{loss}/P_{in}) defined by the following equation was calculated and given in Fig. 6 [15].

$$P_{loss}/P_{in} = 1 - (|\Gamma|^2 + |T|^2) \quad (5)$$

In this figure, the power loss increased significantly with increasing milling time (i.e. the thickness reduction) at f = 1-6 GHz, indicating excellent electromagnetic wave absorption performance of the composite sheet containing very thin flakes in the GHz range. Moreover, Figs. 5 and 6 suggest that the large electromagnetic wave energy

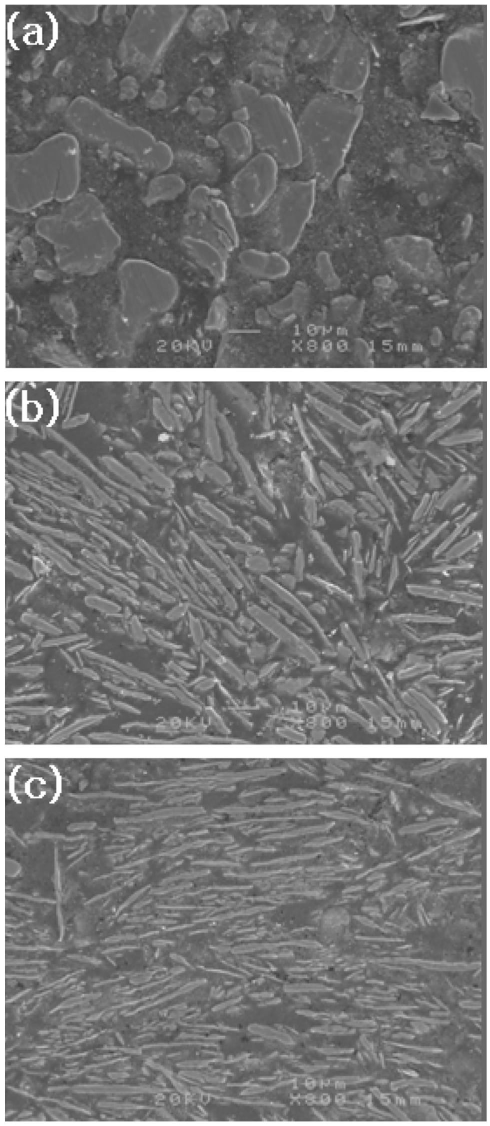

Fig. 3. Cross-sectional views of the composite sheets including nanocrystalline FeCuNbSiB alloy flakes milled for (a) 0 h, (b) 12 h, and (c) 24 h, respectively.

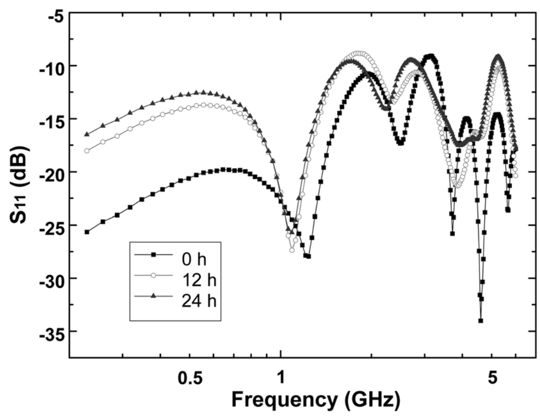

Fig. 4. Frequency dependence of the reflection parameter (S_{11}) for the composite sheets including the nanocrystalline FeCuNbSiB alloy flakes milled for 0, 12, and 24 h, respectively.

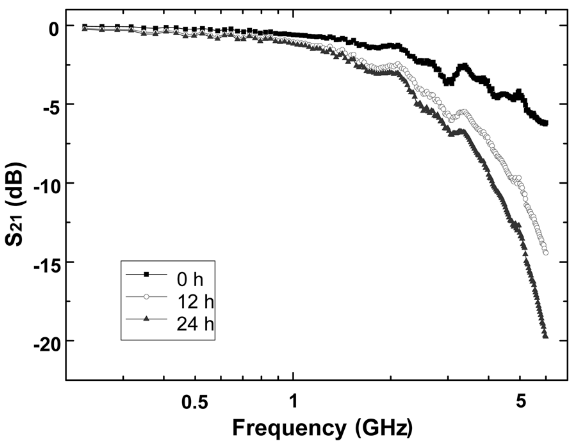

Fig. 5. Frequency dependence of the reflection parameter (S_{21}) for the composite sheets including the nanocrystalline FeCuNbSiB alloy flakes milled for 0, 12, and 24 h, respectively.

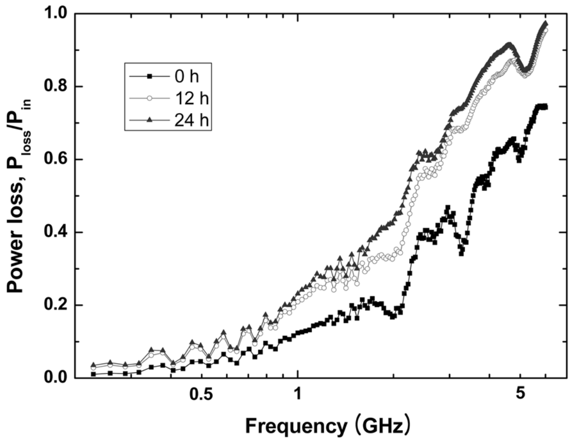

Fig. 6. Frequency dependence of the power loss (P_{loss}/P_{in}) for the composite sheets including the nanocrystalline FeCuNbSiB alloy flakes milled for 0, 12, and 24 h, respectively.

absorption was caused mainly by the significant decrease in S_{21}.

The complex permeability ($\mu = \mu' - j\mu''$) and loss factor (μ''/μ') of the composite sheets including the FeCuNbSiB alloy flakes subjected to attrition for 0, 12, and 24 h were examined to determine why the electromagnetic wave absorption property was enhanced by the decrease in flake thickness.

Figs. 7 and 8 present the milling time dependences of the real and imaginary part of complex permeability (μ' and μ''). Although there was little change in the complex permeability with milling time (i.e. flake thickness), both the μ' and μ'' values were rather high after 24 h milling compared to the cases after 0 and 12 h milling. Furthermore, the loss factor shown in Fig. 9 increased with increasing milling time at frequencies >4 GHz.

The increase in power loss in the composite sheets

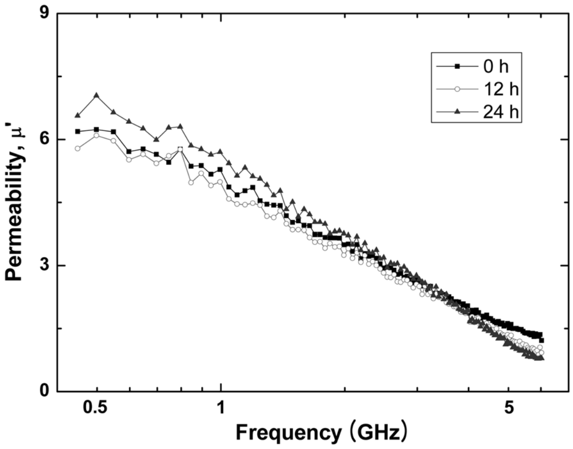

Fig. 7. Frequency dependence of the real part of the complex permeability (μ') for the composite sheets including the nanocrystalline FeCuNbSiB alloy flakes milled for 0, 12, and 24 h, respectively.

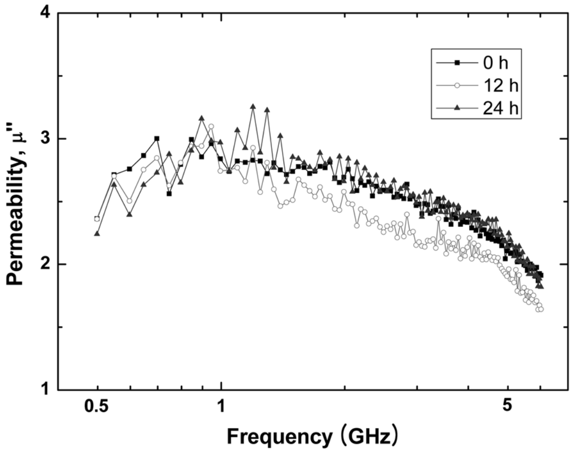

Fig. 8. Frequency dependence of the imaginary part of the complex permeability (μ'') for the composite sheets including the nanocrystalline FeCuNbSiB alloy flakes milled for 0, 12, and 24 h, respectively.

composed of thinner magnetic flakes, as shown in Fig. 6, was attributed partly to their high magnetic loss in the f = 4-6 GHz range.

However, from Figs. 6 and 9, an increase in power loss characteristics was observed over almost the entire measurement frequency range (0.2-6 GHz) after 12 and 24 h milling, while an increase in magnetic loss factor was observed in the narrow frequency range from 4 to 6 GHz only.

Therefore, different factors other than magnetic loss might be related to the larger power loss of the sheets with highly milled flakes.

Figs. 10 and 11 show the complex permittivity of the composite sheets with nanocrystalline FeCuNbSiB alloy powders milled for 0, 12, and 24 h. Both the real and

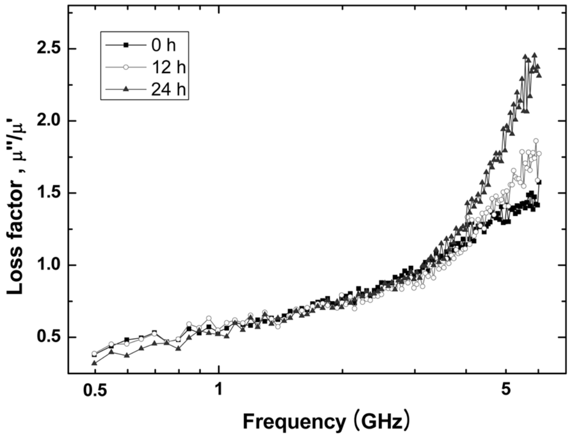

Fig. 9. Frequency dependence of the loss factor (μ''/μ') for the composite sheets including the nanocrystalline FeCuNbSiB alloy flakes milled for 0, 12, and 24 h, respectively.

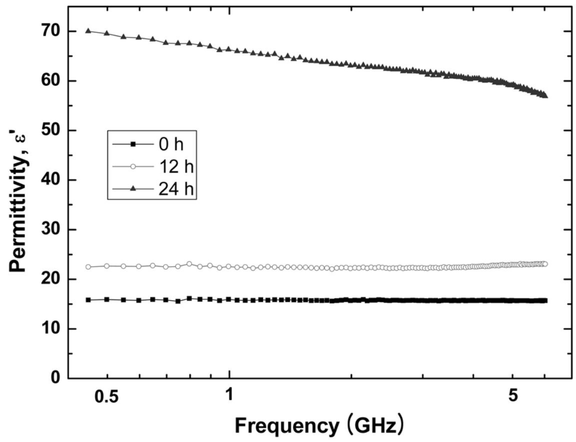

Fig. 10. Frequency dependence of the real part of the complex permittivity (ε') for the composite sheets including nanocrystalline FeCuNbSiB alloy flakes milled for 0, 12, and 24 h, respectively.

imaginary part of the permittivity (ε' and ε'') increased with increasing milling time. After 24 h milling, the ε' and ε'' values changed remarkably from 15 and 0.6 to 60-70 and 9-10, respectively.

The electrical capacitance and permittivity are expected to have different values if the lamellar structure of the conductor-dielectric materials in a composite sheet changes. The marked increase in permittivity, shown in Figs. 10 and 11, after 12 and 24 h milling was attributed to the increase in the interface of the metal/polymer per unit volume and decrease in inter-flake separation.

In Eq. (1), the magnitude of the electromagnetic wave absorption energy is dependent on the imaginary part of the permittivity ε''. From the good agreement in the frequency behavior of power loss and ε'', it can be concluded that the high power loss in the composite sheet,

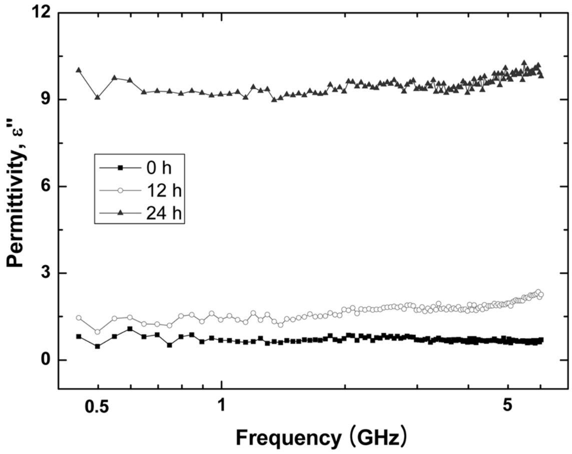

Fig. 11. Frequency dependence of the imaginary part of the complex permittivity (ε'') for the composite sheets including nanocrystalline FeCuNbSiB alloy flakes milled for 0, 12, and 24 h, respectively.

including FeCuNbSiB alloy flakes with a high aspect ratio, which was produced by long-time milling is due to mainly their high complex permittivity and resulting large dielectric loss.

4. Conclusions

This study examined the effects of a thickness reduction of magnetic alloy flakes on the electromagnetic wave absorption characteristics of nanocrystalline $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ (at.%) alloy flakes/polymer composite sheets available for the quasi-microwave band.

The thickness decreased from 18 to 1-2 μm as a result of attrition milling for 24 h. The composite sheet including the alloy flakes milled for 12 and 24 h showed considerably enhanced power loss properties over the GHz frequency range compared to the sheets with non-milled alloy flakes.

Although a considerable increase in the magnetic loss factor with the milling time in a narrow frequency range of 4-6 GHz was observed, the dependence of the complex permeability on the flake thickness was not too large.

However, the complex permittivity largely increased with increasing milling time, and good agreement was observed between the milling time and frequency dependence of the complex permittivity and power loss of the composite sheets.

Overall, the enhanced electromagnetic wave absorption characteristics in composite sheets composed of very thin FeCuNbSiB alloy flakes was attributed to their increased complex permittivity and resulting dielectric loss.

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