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High Frequency Permeability of Fe-Al-Si granular Composite Materials

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***Abstract*—High-frequency electromagnetic properties of Fe-Al-Si alloy (Sendust) granular composite materials have been studied by measuring their relative complex permeability ($\mu_r = \mu_r' - j\,\mu_r''$) and permittivity ($\varepsilon_r = \varepsilon_r' - j\,\varepsilon_r''$) spectra. The bulk Fe-Al-Si alloy shows metallic electrical conduction, and permeability decreases rapidly with frequency. On the other hand, Sendust powder shows relatively high electrical resistivity. Sendust composite material demonstrates insulating electrical properties up to at least 80 vol.% particle content. Thus the relatively high permeability in the microwave frequency range can be obtained. Frequency dispersion characteristics of permeability for the composite were analyzed by the superposition of domain wall and gyromagnetic spin resonance formula. The particle content variation of permeability can be qualitatively described by a coherent model mixing rule.**

I. INTRODUCTION

The suppression of undesired reflection and scattering waves is one of the main issues in EMC. In the designing of electromagnetic wave absorbers (EM-absorbers), the complex permeability and permittivity of absorbing materials are the important parameters.

Ferrite materials have been used for EM-absorbers; magnetic and dielectric properties of ferrites and their composite materials have been extensively studied [1]–[5]. Generally, the frequency dispersion of permeability in ferrites contains three components from the magnetizing process by alternating magnetic fields, the domain wall resonance, the rotational relaxation of magnetization and the gyromagnetic spin resonance [3],[5]. In the magnetic granular composite materials, the demagnetizing effect by the embedded magnetic particles must be taken into account for the frequency dispersion of permeability and the variation of permeability with particle content [5]. Several mixing rules such as the logarithmic law [6], the effective medium theory [7],[8] or the coherent model [9],[10] have been considered.

On the other hand, metal granular composite materials, in which ferromagnetic metal particles are embedded, have been the subject of considerable interest [11]–[15]. Since metal granular composite materials have relatively high electrical resistance, the eddy current effect can be suppressed even over the microwave frequency range. However, the high-content metal granular composite, in which the embedded metal particles are percolated, has metallic electrical conduction. As a consequence, eddy current effect appears in the permeability spectrum [11]. The electromagnetic properties of composites containing different ferromagnetic metal particles have been studied, e.g., Fe-Ni alloy (Permalloy) [11], Fe-Co alloy (Permendur) [15], and Fe-Al-Si alloy (Sendust) [13]. To improve the high frequency permeability, the oxidation treatment for particle surfaces can be used to increase the contact resistance between particles [15], [16]. Among the ferromagnetic metals, Fe-Al-Si alloy (Sendust) has relatively high electrical resistivity. This material is brittle. Complex permeability of Fe-Si-Al alloy flakes composite materials was studied over the RF and microwave frequency bands to improve the frequency dispersion of permeability for the EMC applications [13], [17].

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The objective of this study is to investigate the complex permeability and permittivity of Fe-Al-Si alloy granular composite materials containing spherical particles over the microwave frequency range. Study of the complex permeability spectra and understanding their frequency dispersion mechanism for Fe-Al-Si alloy composite materials are the main problems considered in this work. The mixing rules of the complex permeability applicable to metal granular composite materials will be also discussed.

II. Experimental

A. Sendust Particles and Sendust Composite Materials

A commercially available Sendust ($Fe_{85.56}Al_{5.69}Si_{8.75}$) alloy particles were used for granular composite materials. The composition of the Sendust particles slightly deviated from the Sendust alloy composition $Fe_{85.0}Al_{5.4}Si_{9.6}$, which exhibits the highest permeability [18]. Particle diameter was controlled below 45 μm using a 3 stack sieves with apertures within a range from 45 to 150 μm. Fig.1 shows a SEM photograph of the sieved particles. Geometry of almost all particles is spherical, and the particle size ranges from a few μm to several tens μm. Using the SEM photographs, the particle size distribution was evaluated by measuring the diameter of particles. Obtained particle size distribution of Sendust particles is shown in Fig. 2. The mean diameter of particles was determined by fitting this data to the lognormal distribution function,

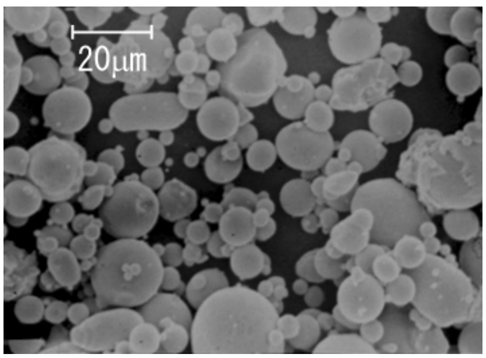

Fig. 1. SEM photograph of Sendust particles

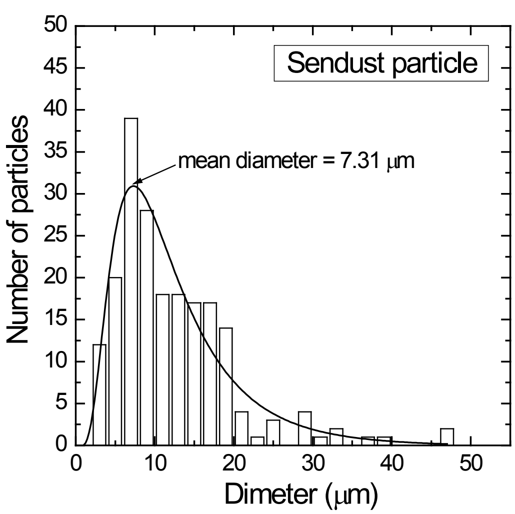

Fig. 2. Particle size distribution of Sendust particle

$$f(d) = \frac{1}{\sqrt{2\pi}\sigma d} e^{-\frac{(\log d - d_m)}{2\sigma^2}} \qquad (1)$$

where d is the diameter of particles, σ is the variance and d_m is the mean diameter. The mean particle diameter is 7.31 μm.

Sendust granular composite materials were prepared by the following process.

(1) Sendust particles were mixed with appropriate amounts of PPS (polyphenylene sulfide) resin.

(2) Pressed pellet was made using a die. The pellet in the die was heated to melt the resin and regulated at a temperature of 300 °C and pressed under the pressure of 200 kgf/cm^2.

(3) Samples were obtained by cooling the pellet to room temperature.

The particle content was estimated using the density values of Sendust (ρ_f = 7.10 g/cm^3), PPS resin (ρ_m = 1.36 g/cm^3), and Sendust composite ρ. Several Sendust composite materials having the different particle content were prepared. Obtained samples were cut into the toroidal form for the measurement of complex permeability and permittivity. Thickness of the samples was adjusted around 1 mm to avoid the dimensional resonance in permeability. Rectangle samples were also cut out for the electrical resistivity measurements.

B. Complex permeability and permittivity measurements

Relative complex permeability $\mu_r = \mu_r' - j\mu_r''$ was obtained from the input impedance of samples loaded in a coaxial line in the frequency range from 1 MHz to 100 MHz using an impedance analyzer (HP4194A) [19],[20]. Over the frequency range from 100 MHz to 6 GHz, the high-frequency complex permeability $\mu_r = \mu_r' - j\mu_r''$ and permittivity $\varepsilon_r = \varepsilon_r' - j\,\varepsilon_r''$ were measured by the transmission / reflection method using a network analyzer Agilent E5071C. The complex permeability of bulk Sendust sample over the frequency range from 100 Hz to 40 MHz was obtained by measuring the inductance and resistance differences between two toroidal coils wound around the toroidal sample and a Teflon blank. Electrical resistivity was measured by the two-probe method using the impedance analyzer from 1 kHz to 40 MHz.

III. Results and Discussion

A. Complex permeability of Sendust Alloy

Fig. 3 shows the complex permeability spectra of the Sendust alloy prepared from the Sendust particles by arc melting. Real part of the relative permeability μ_r' is about 1000 at 100 Hz and begins to decrease rapidly from about 300 Hz with increasing frequency and reaches unity at 10 MHz. Imaginary part μ_r'' shows the same frequency variation. From the d.c. electrical resistivity measurement, the resistivity value of 72.7 μΩ·cm at room temperature was obtained. Thus the decrease of permeability can be due to the eddy current effect [11]. Skin depth δ at 1 GHz estimated by the approximated formula $\delta = \sqrt{2/\omega\mu\sigma}$ was 13.6 μm, where ω, σ and μ are the angular frequency, electrical conductivity and permeability, respectively.

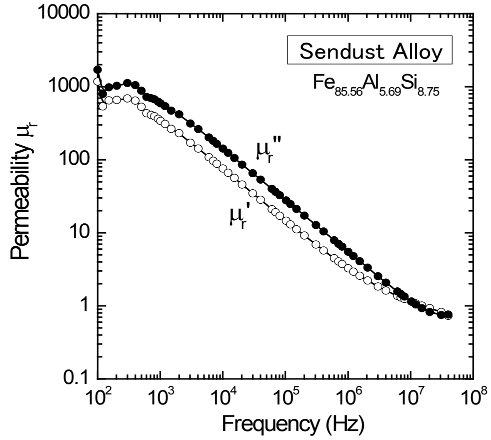

Fig. 3. Relative complex permeability spectra of Sendust alloy

B. Complex permeability of Sendust Composite Materials

The significant frequency dispersion of permeability of the Sendust composite materials, as is shown in Fig. 4, appears at higher frequencies compared to the bulk Sendust (see Fig. 3).

For the 62.9 vol.% sample, the μ_r' value is about 18 up to 30 MHz, and it decreases at frequencies above approximately 40 MHz. On the other hand, μ_r" has a maximum at about 400 MHz. As the particle content decreases, the low-frequency μ_r' decreases. The maximum frequency of μ_r" shifts to the higher frequencies and reaches 1 GHz for the particle content of 36.1 vol.%. The dispersion frequency shift can be produced by the shift of the resonance frequencies of domain wall vibration and gyromagnetic spin rotation; this feature has been observed in ferrite and ferromagnetic metal granular composite materials [5],[11],[21]. Thus the low particle content composite has larger permeability in the high frequency range above 2 GHz.

However, the permeability dispersion of Sendust composite with the high-particle content is different from that of the other metal granular composites, such as Permalloy or Permendur. In these composite materials, permeability dispersion shows the rapid decrease in high frequency range due to the eddy current effect. In Permalloy composites, percolating metal particles cause the increase of electrical conductivity, and the high frequency permeability is suppressed by the eddy current effect. The insulating characteristic of the high-content Sendust composite is attributed to the high electrical resistivity of Sendust particles which will be discussed later.

C. Magnetic Circuit Model and the Variation of Permeability with Particle Content

The frequency dispersion of the–permeability of magnetic materials can be described by the superposition of domain wall resonance, magnetization rotational relaxation, and

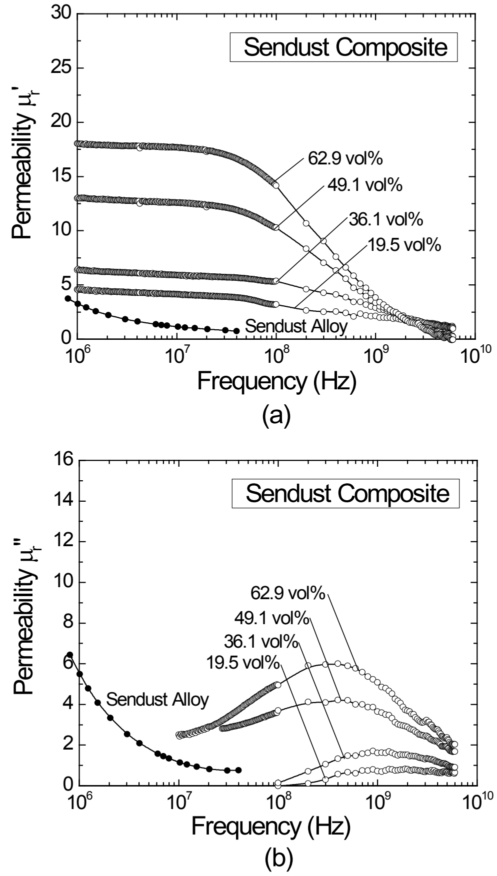

Fig. 4. Real (a) and imaginary (b) parts of relative complex permeability μ_r' and μ_r" for Sendust composite materials. Solid circle indicates the data of bulk Sendust alloy.

gyromagnetic spin resonance. In ferrite composite materials, the coherent model [9] can be applied to explain the variation of permeability, as well as the resonance or relaxation frequencies, as the functions of the ferrite particle content [5], [10]. Herein, permeability of composite materials is calculated based on the magnetic circuit model [10] as

$$\mu_r(\omega) = \frac{\mu_B\left(1+\dfrac{\delta}{D}\right)}{1+\mu_B\dfrac{\delta}{D}}, \qquad (2)$$

where, D is the average size of the ferrite particles, $\delta/2$ is the average thickness of the nonmagnetic resin layer and μ_B is the complex permeability of sintered ferrite. Thus, the permeability value of composite materials can be scaled by the gap parameter δ/D. This parameter is connected to the volume ferrite content ϕ as

$$\phi = \left(1+\frac{\delta}{D}\right)^{-3} \qquad (3)$$

Thus the variation of the permeability with particle content can be described by the mixing rule called "the coherent model",

$$\mu_r = \frac{\mu_B \phi^{-1/3}}{1-\mu_B+\mu_B\phi^{-1/3}}. \quad (4)$$

Fig. 5 shows the absolute relative permeability of Sendust composite at 1 MHz as a function of particle content. Solid circles are the measured permeability values, dash-dotted line indicates the logarithmic law curve, and the solid line is calculated from (4). Here, high density 80 vol.% sample was made using the material Sb_2O_3 as a binder by heating at 650 ^{o}C in air. The measured permeability values for all particle contents are significantly lower than the values predicted by the logarithmic mixing law. This indicates that the magnetic circuit model can qualitatively explain the demagnetizing effect in the Sendust composite, similar to ferrite composite materials [5]. Thus some other mixing theory such as the Maxwell Garnet approximation or the Bruggeman's effective medium theory can be considered to make quantitative explanation of the permeability variation in Sendust composite material [22].

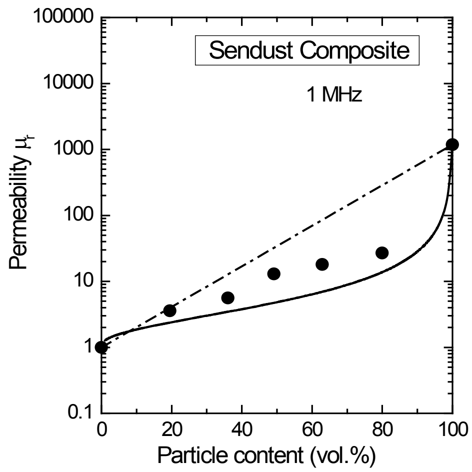

Fig. 5. Absolute permeability value at 1 MHz of Sendust composite materials as a function of particle content. The 100 vol% value is from the bulk Sendust data at 100 Hz.

D. Contribution of Domain Wall and Natural Resonance to the Permeability Spectra

The permeability spectra of Sendust composite materials was analyzed assuming the superposition of the two types of magnetic resonances, domain wall and gyromagnetic spin resonance [5]. The following permeability dispersion formula was applied

$$\mu_r = 1+\chi_d+\chi_s = 1+\frac{\omega_d^{\,2}\chi_{d0}}{\omega_d^{\,2}-\omega^2+j\omega\beta}+\frac{(\omega_s+j\omega\alpha)\omega_s\chi_{s0}}{(\omega_s+j\omega\alpha)^2-\omega^2}, \quad (5)$$

where χ_d and χ_s are the magnetic susceptibility for domain wall and gyromagnetic spin motions, respectively; $\omega_d = 2\pi f_d$ and $\omega_s = 2\pi f_s$ are the resonance angular frequencies of the domain wall and the spin components; χ_{d0} and χ_{s0} are the static magnetic susceptibilities of each component; and α and β are the damping factors. Fig. 6 shows the permeability spectra of the 62.9 vol.% Sendust composite material. The parameters of the dispersion (5) were obtained by the numerical fitting of (5) to the measured data. The estimated dispersion parameters by the numerical fitting are f_s = 2.23 GHz, f_d = 844 MHz, χ_{s0} = 5.98, χ_{d0} = 5.74, α = 2.52 and β = 6.62 × 10^9, respectively. The frequency dispersion of permeability for the Sendust composites can be described by two resonance components. High-frequency permeability is mainly attributed to the spin resonance. The relaxation characteristics in domain-wall and spin components are caused by the relatively high damping values. However, there is a slight discrepancy between the fitted curve and the measured data over the frequency range from 3 to 6 GHz. This may be attributed to the eddy current effect in the embedded metal particle. Considering the dispersed particle size, the skin depth becomes equal to the average diameter of dispersed particle at approximately 4 GHz. Though a resonance behavior can exist at frequencies above 6 GHz, the real part of permeability μ_r' does not show negative value up to 20 GHz in the Fe-Al-Si granular composite material [17].

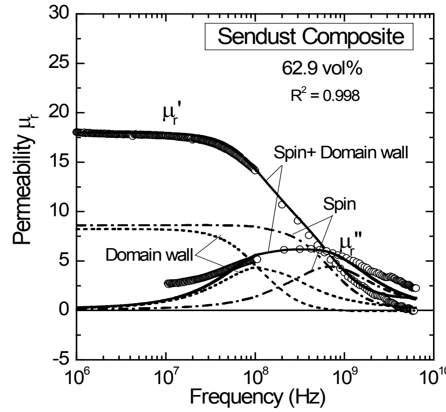

Fig. 6. Complex permeability spectra of 62.9 vol.% Sendust composite material. Solid, dotted and dashed-dotted lines are calculation curves using fitting parameters determined from equation (5). Coefficient of determination factor R^2 is 0.998.

E. Electrical properties of Sendust Composite Material

Electrical and dielectric properties of Sendust composite material was evaluated by electrical resistivity and complex permittivity. In the previous study of Fe-Ni alloy (Permealloy) composite material, metallic property is observed above 40 vol.% particle content [11]. The electrical percolation of metal particles produces relatively high electrical conductivity in the high content permalloy composite. Thus, oxidized Permalloy particles were used to achieve the insulating high particle content composite for high frequency applications [16].

Electrical conductivity σ_{ac} calculated by the resistivity data is shown in Fig. 7 as a function of frequency. Electrical conductivity of Sendust particles was measured by the two-terminal method using cylindrical electrodes [16]. The resultant conductivity value is 1.397 S/cm at 100 Hz, which is much smaller than 13755 S/cm for the bulk Sendust alloy. σ_{ac} of Sendust powder is almost constant up to 1 M Hz and decreases at the higher frequencies. The preliminary experiments for the temperature variation of σ_{ac} show that the Sendust powder behaves as a semiconductive material. Thus,

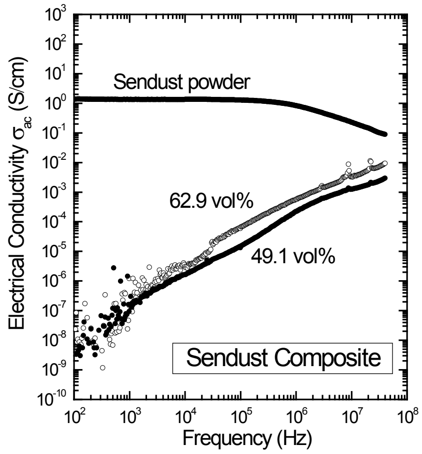

Fig. 7. The ac el ectrical conductivity of Sendust powder and composite materials as a function of frequency at room temperature.

it is considered that the decrease of σ_{ac} is caused by the oxidation of the particle surface in the atomizing process. Further, the contact resistance between particles also enhances the resistivity of the powder and composite materials.

In the Sendust composite material, the 62.9 vol.% composite shows an insulating behavior, and the conductivity value is on the order of 10^{-8} S/cm for the frequency range from 100 Hz to 1 kHz. The value σ_{ac} increases with frequency and reaches the order of 10^{-2} S/cm at 40 MHz. Since the particle content in this case is above the electrical percolation threshold [11], the contact resistance and oxidized surface layer lead to the low conductivity state. It should be noted that the 80 vol.% Sendust composite with Sb_2O_3 binder also exhibits an insulating electrical property as well. From the above results, the Sendust composite material containing surface oxidized particles has insulating electrical property at least up to about 80 vol.% particle content; the eddy current effect in the bulk composite is not necessary to consider. However, in high frequency range, the eddy current is

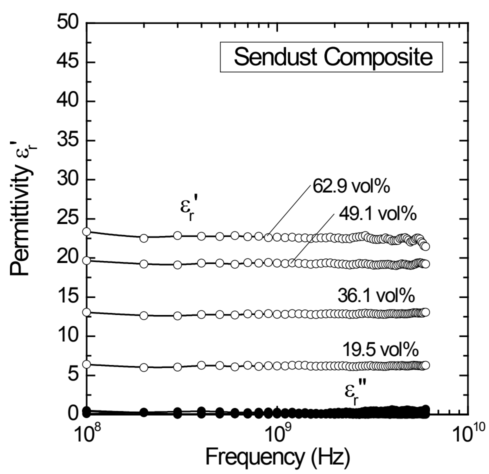

Fig. 8. The complex permittivity spectra of Sendust composite materials. The open circle and solid circle indicate the real and imaginary parts, respectively.

produced in embedded micron order particles by the intrinsic metallic property and permeability is affected by this effect.

Fig. 8 shows the complex permittivity spectra $\varepsilon_r = \varepsilon_r' - j\varepsilon_r''$ of the Sendust composite materials. The real permittivity ε_r' is constant up to 6 GHz, and the dielectric loss is almost zero in all the composite materials. The ε_r' value increases with an increasing particle content, and the variation can be described by the logarithmic law.

IV. Conclusion

The magnetic and electrical properties of Fe-Al-Si alloy composite materials up to the microwave frequency region have been studied. I Sendust composite material containing particles with a semiconductive oxidized surface layer exhibits insulating electrical behavior up to the high particle content of 80 vol.%. Frequency dispersion of permeability can be described by the domain-wall and gyromagnetic spin resonance phenomena. In the microwave range, the eddy current produced on the embedded particle surface may affect the permeability spectrum. Permeability variation with particle content can be qualitatively described by a mixing rule, the coherent model.

For further investigations, the magnetic field effect on the high frequency permeability of Sendust composite materials and the analysis of permeability variation with particle content by the other mixing rules are now in progress.

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References

[1] G. T. Rado, "Magnetic Spectra of Ferrites," *Rev. Mod. Phys.*, vol. 25, pp. 81–90, Jan. 1953.
[2] E. Schlomann, "Microwave Behavior of Partially Magnetized Ferrites," *J. Appl. Phys.*, vol. 41, pp. 204–214, Jan. 1970
[3] A. Globus, "Some Considerations about the Domain Wall Size Theory of Magnetization Mechanisms," *J. Phys.*, vol. 38, pp. C1-1–C1-15, 1977.
[4] J-L. Mattei, D. Bariou, A. Chevalier, M. L. Floc'h, "Gyroresonance in unsaturated composite bodies: Experiments and theory," *J. Appl. Phys.*, vol. 87, pp. 4975–4977, May 2000.
[5] T. Tsutaoka, "Frequency dispersion of complex permeability in Mn-Zn and Ni-Zn spinel ferrites and their composite materials," *J. Appl. Phys.*, vol. 93, pp. 2789–2796, Mar. 2003.
[6] J. B. Birks, "The measurement of the Permeability of Low-conductivity Ferromagnetic Materials at Centimetre Wavelength," *Proc. Phys. Soc*, vol. *LX3*, pp. 282–292, 1947.
[7] D. A. G. Bruggeman, "Berechung verschiedener physikalischer Konstanten von heterogenen Substanzen," *Annalen der Physik*, vol. 5, pp. 636–679, 1936.
[8] D. Bariou, P. Queffelec, P. Gelin, M. L. Floc'h, "Extension of the Effective Medium Approximation for Determination of the Permeability Tensor of Unsaturated Polycrystalline Ferrites," *IEEE Trans. Mag.*, vol. 37, pp. 3885–3891, Nov. 2001.
[9] M. T. Johnson and E. C. Visser, "A Coherent Model for the Complex Permeability in Polycrystalline Ferrites," *IEEE Trans. Mag.*, vol. 26, pp. 1987–1989, Sept. 1990.
[10] T. Nakamura, T. Tsutaoka, K. Hatakeyama, "Frequency dispersion of permeability in ferrite composite materials," *J. Magn. Magn. Mat.*, vol. 138, pp. 319–328, 1994.

[11] T. Kasagi, T. Tsutaoka, K. Hatakeyama, “Particle Size Effect on the Complex Permeability for Permalloy Composite Materials,” *IEEE Trans Mag.*, vol. 35, pp. 3424–3426, Sep. 1999.
[12] S. Yoshida, M. Sato, E. Sugawara, Y. Shimada, “Permeability and electromagnetic-interference characteristics of Fe-Si-Al alloy flakes-polymer composite,” *J. Appl. Phys.*, vol. 85, pp. 4636–4638, Apr. 1999.
[13] C. Brosseau, P. Talbot, “Effective magnetic permeability of Ni and Co micro- and nanoparticles embedded in a ZnO matrix,” *J. Appl. Phys.*, vol. 97, pp. 104325-1–104325-11, 2005.
[14] Y. Deng, L. Zhao, B. Shen, L. Liu, W. Hu, “Microwave characterization of submicrometer-sized nickel hollow sphere composites,” *J. Appl. Phys.*, vol. 100, pp. 014304-1–014304-5, 2005.
[15] T. Kasagi, T. Tsutaoka, K. Hatakeyama, “Complex Permeability Spectra of Permendur Composite Materials,” *J. Phys.: Conf. Series*, vol. 200, pp. 082012-1–082012-4, 2009.
[16] T. Kasagi, T. Tsutaoka, K. Hatakeyama, “Negative permeability spectra in Permalloy granular composite materials,” *Appl. Phys. Lett.*, vol. 88, pp. 172502-1–172502-3, 2006.
[17] Y. W. Yun, S. W. Kim, G. Y. Kim, Y. B. Kim, Y. C. Yun, K. S. Lee, “Electromagnetic shielding properties of soft magnetic metal and ferrite composites for application to suppress noise in a radio frequency range,” *J. Electroceramics.*, vol. 17, pp. 467–469, 2006.
[18] H. Masumoto, T. Yamamoto, “On a New Alloy “Sendust” and its Magnetic and Electric Properties,” *J. Japan. Inst. Metals*, vol. 1, pp. 127–135, 1937 (in japanese).
[19] T. Tsutaoka, T. Nakamura, K. Hatakeyama, “Frequency dispersion and temperature variation of complex permeability of Ni-Zn ferrite composite materials,” *J. Appl. Phys.*, vol. 78, pp. 3983–3991, Sept .1995.
[20] W. B. Weir, “Automatic measurement of Complex Dielectric Constant and Permeability at Microwave Frequencies,” in *Proc. IEEE*, 1974, paper 10.1109, pp. 33–36.
[21] T. Kasagi, S. Suenaga, T. Tsutaoka, K. Hatakeyama, “High frequency permeability of ferromagentic metal composite materials,” *J. Magn. Magn. Mat.*, vol. 310, pp. 2566–2568, 2007.
[22] A. N. Lagarkov, K. N. Rozanov, "High-frequency behavior of magnetic composites", *J. Magn. Magn. Mat.*, vol. 321, pp. 2082–2092, 2009.