12 Dec 2012

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Recommended Citation

T. Tsutaoka et al., "Electromagnetic Properties Of Metal Granular Composite Materials For EMC Applications," *IEEE International Symposium on Electromagnetic Compatibility*, pp. 411 - 415, article no. 6351812, Institute of Electrical and Electronics Engineers, Dec 2012.
The definitive version is available at https://doi.org/10.1109/ISEMC.2012.6351812

Electromagnetic Properties of Metal Granular Composite Materials for EMC Applications

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Abstract—**Electromagnetic properties of copper and $Fe_{55}Ni_{45}$ alloy (Permalloy) composite materials have been studied by measuring the relative complex permeability ($\mu_r = \mu_r' - j\mu_r''$), and permittivity ($\varepsilon_r = \varepsilon_r' - j\varepsilon_r''$) spectra over the microwave range, as well as the a.c. electrical conductivity σ_{ac} spectra. The variation of σ_{ac} and ε_r with particle content shows the insulator-metal transition due to the percolation effect. The effective cluster model can be applied to the percolation effect in permittivity. The permeability spectra in Permalloy composites, which contain the percolated particles, can be affected by the eddy current effect. The permeability relaxation frequency of Permalloy composites shifts to the higher frequencies by the oxidation of embedded particle surfaces.**

I. Introduction

The suppression of undesired reflection and scattering waves, as well as reduction of currents on conducting surfaces, is one of the main issues in electromagnetic compatibility and immunity (EMC and EMI). Various kinds of electromagnetic wave absorbers (EM-absorbers) or shielding materials are used for solving EMC/EMI problems. In the designing of EM-absorbers or shielding materials, the complex permeability and permittivity spectra, as well as the electrical conductivity of the magnetic, dielectric, and metallic materials are the important characteristics. The magnetic or dielectric loss in insulating materials can be used to dissipate the electromagnetic energy in the EM-absorbers, and the combination of high electrical conductivity and high permeability is effective to shield the EM waves.

As the material for EMC/EMI, metal granular composite materials, in which magnetic or non-magnetic metal particles are embedded, have been the subject of considerable interest [1]-[4]. Since metal granular composite materials have relatively high electrical resistance, the eddy current, or skin effects, can be suppressed even in the microwave frequency range. However, the high-content metal granular composite material may show metallic electrical conduction due to the percolation effect of embedded particles [5]. Thus, the control of the metallic property in metal granular composites is desired for designing the EM wave shielding materials.

The electromagnetic properties of ferromagnetic metal granular composite materials, containing different types of particles, have been studied, *e.g.*, Fe-Ni alloy (Permalloy) [6], Fe-Co alloy (Permendur) [7], and Fe-Al-Si alloy (Sendust) [8], [9]. To improve the high-frequency permeability dispersion, the oxidation treatment for particle surfaces can be used to increase the contact resistance between particles [7], [10].

The objective of this study is to investigate the electromagnetic properties of metal granular composite materials by considering the percolation effect in the electrical conductivity and permittivity. Also, the effect of the surface oxidation of embedded particles on the magnetic permeability and electric permittivity spectra in Permalloy composite materials has been studied.

II. Sample Preparation and Measurements

A commercially available Cu and Permalloy ($Fe_{55}Ni_{45}$) particles were used for metal granular composite materials.

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Geometry of particles is spherical and the mean particle diameters d_{av} are 10 μm for Cu and 2.5 μm for Permalloy powders, respectively. Permalloy particles were also heat treated in air using an electric furnace at 300 °C in order to make oxidized surfaces.

Granular composite materials were prepared by mixing metal particle powders with polyphenylene sulphide (PPS) resin powder, melting the resin at 300 °C and pressing the mixture at a pressure of 31.83 MPa in the cooling process down to room temperature. The samples obtained were cut into a toroidal form with the inner diameter of 3 mm, the outer diameter of 7 mm. The thickness of the samples of about 1 mm was controlled to avoid the dimensional resonance of the electromagnetic wave in the coaxial line. Rectangle samples were also cut out for the electrical resistivity measurements. The particle content was estimated using the density values of fillers, PPS resin, and the metal granular composites.

Relative complex permeability $\mu_r = \mu_r' - j\mu_r''$ and permittivity $\varepsilon_r = \varepsilon_r' - j\varepsilon_r''$ spectra in the frequency range from 1 MHz to 40 MHz were obtained using an impedance analyzer (HP4194A) by measuring the input impedance of samples loaded in a coaxial line [11]. Over the frequency range from 100 MHz to 10 GHz, the high-frequency complex permeability μ_r and permittivity ε_r were measured by the standard 7/3-mm coaxial air line techniques [12], [13] using a network analyzer Agilent E5071C. The complex permeability of the bulk Permalloy sample in 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 ρ_{ac} was measured by the two-probe method using the impedance analyzer from 1 kHz to 40 MHz and the electrical conductivity $\sigma_{ac} = 1/\rho_{ac}$ was obtained.

III. Results and Discussion

A. AC Conductivity and Percolation Effect in Copper and Permalloy Composite Materials

Fig. 1 shows the a.c. conductivity σ_{ac} spectra of Cu (a) and Permalloy (b) composite materials. In the low particle content from 2 to 10 vol.%, Cu composites show low electrical conductivity about 10^{-7} to 10^{-5} S/cm at 1 kHz and σ_{ac} increases with increasing frequency. Thus low particle content composites indicate the insulating property. On the other hand, 20 vol.% and above Cu composites show a relatively high electrical conductivity of 2 to 5 S/cm, indicating the metallic property. No frequency dispersion is observed up to 10 MHz. This insulator-metal transition in electrical conductivity is attributed to the percolation effect of embedded particles; the variation of the conductivity with particle content in non-magnetic metal granular composites has been discussed by the shell structure model [1] or the effective medium theory [5].

In Permalloy composite materials, a metallic electrical conduction is observed for 41 vol.% and above particle contents, as shown in Fig.1 (b). The σ_{ac} spectrum of metal Permalloy is also shown in this figure; the σ_{ac} at 1 kHz is 5.9 $\times 10^4$ S/cm. A linear increase of σ_{ac} with frequency is observed from about 100 kHz in the insulating 39 vol.% composite. These results indicate that the percolation threshold in Permalloy composites is higher than that in Cu composites. Since the percolation threshold is determined by the geometry of the composite which includes the agglomeration or the surface condition of the particles etc., the difference between Cu and Parmalloy composites can be

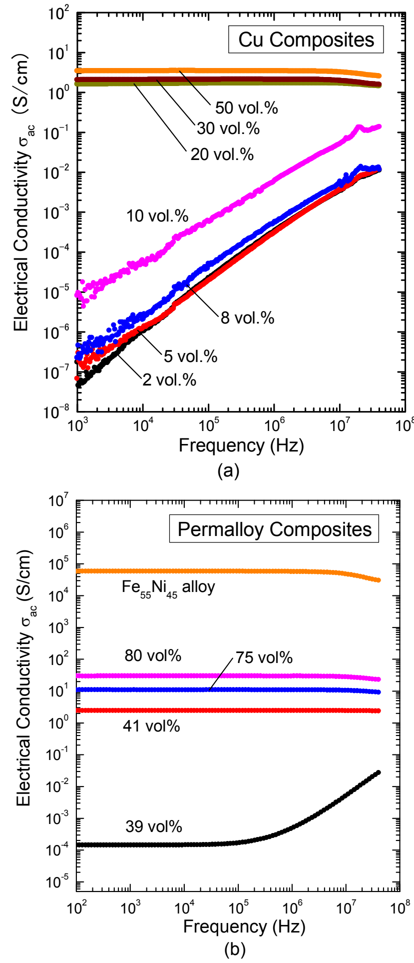

Fig. 1. Electrical conductivity spectra of copper (a) and Permalloy (b) granular composite materials.

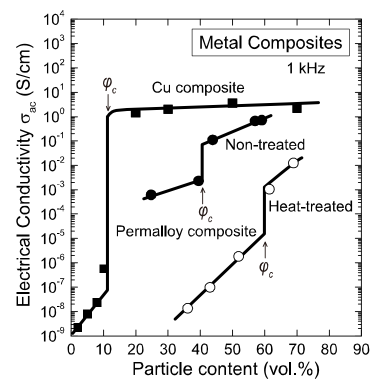

Fig. 2. Electrical conductivity σ_{ac} at 1 kHz for Cu and Permalloy composite materials as a function of particle content.

caused by the difference of the particle size distribution or the geometry of particles. Further, it is known that the morphology of a polymer also affects percolation; it may build chains or nets of one type of inclusions easier than of the other [14]. Hence the percolation threshold can be changed in the other host resin composites. It should be noted that the electrical conductivity of PPS resin is in the order of 10^{-8} S/cm.

The variation of a.c. conductivity at 1 kHz with particle content is summarized in Fig. 2 for Cu and Permalloy composite materials. An electrical jump in the σ_{ac} versus particle content curves was observed in all composites indicating the insulator-metal transition. The estimated percolation threshold volume fractions φ_c are 0.10, 0.40 and 0.61 for Cu composite, non-treated and heat-treated Permalloy composites, respectively. The increase of the percolation threshold φ_c in Permalloy composites can be attributed to the enhancement of the contact resistance between embedded particles by the surface oxidation; thus, the insulating property of composite structures can be obtained in relatively high particle contents by the heat-treatment of embedded particles.

B. Complex Permittivity of Metal Granular Composite Materials

The frequency dispersion curves of relative complex permittivity of the Cu (a) and Permalloy (b) composite materials are shown in Fig. 3. In the host PPS resin and low particle content Cu composites from1.8 to 7.6 vol.%, the real part ε_r' is constant up to 40 MHz and increases with increasing particle content. Simultaneously, the imaginary part ε_r" is almost zero in the all contents (Fig. 3 (a)). On the other hand, in the 10.1 vol.% and above Cu composites, the imaginary part ε_r" becomes huge and decreases rapidly with increasing frequency. The variation of the electrical permittivity with frequency in the high conductive metal can be described by the following formula,

$$\varepsilon_r = -j\frac{\sigma}{\varepsilon_0\omega}, \qquad (1)$$

where ω is the angular frequency of electric field and σ is the dc electrical conductivity. Thus the ω^{-1} dependence of ε_r" in the 10.1 and 20 vol.% samples indicates the metallic property in this particle content.

On the other hand, Permalloy composite materials show the different change in permittivity spectra. Up to the 20 vol.% particle content, the real part ε_r' shows almost the same property as Cu composites; ε_r' is almost constant up to 6 GHz and dielectric loss is very small. The ε_r' value increases with particle content and relatively high imaginary part ε_r" appears at 30 vol.%. Finally, a metallic permittivity dispersion is observed at 57.1 vol.%, which is above the percolation threshold. Therefore, dielectric and metallic properties coexist in the intermediate particle content range in Permalloy composites.

Doyles and Jacobs investigated the change of dielectric to metallic properties in the suspension of conducting spheres using the Effective Cluster Model (ECM) based on the Clausius-Mossotti relation [15] and evaluated the dielectric

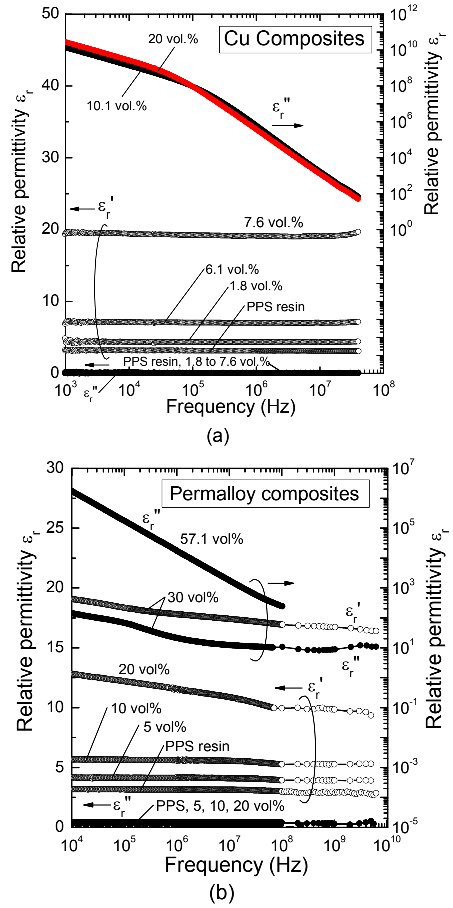

Fig. 3. Relative complex permittivity spectra of Cu (a) and Permalloy (b) composite materials.

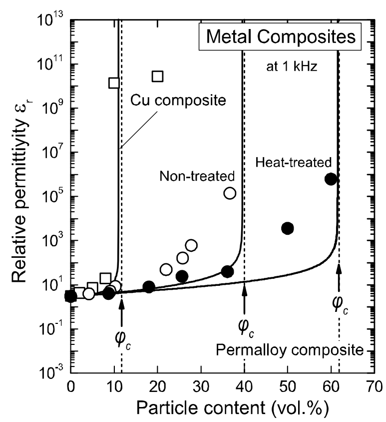

Fig. 4. Relative permittivity ε_r at 1 kHz for metal composite materials as a function of particle content.

enhancement in the metal granular composite [16]. They assumed that a part of dispersed particles make clusters with increasing particle content, and the fraction of isolated particles decreases. At the percolation threshold, the cluster size runs up to whole area in the composite. To describe the variation of permittivity with particle content, the following formula was derived [16].

$$\frac{\varepsilon_r - 1}{\varepsilon_r + 2} = \varphi\left[1 + \frac{\varphi}{\varphi_c}\left(\frac{1}{\varphi_c} - 1\right)\right] \qquad (2)$$

Fig. 4 shows the absolute value of relative permittivity at 1 kHz for Cu and Permalloy composites as a function of particle content. Open squares indicate the values of the Cu composite, solid and open circles are those of the Permalloy composites. The relative permittivity shows a rapid increase near the percolation threshold φ_c. Solid lines indicate the curves calculated using (2). From these data, the dielectric enhancement in Cu and Permalloy composites caused by the percolation effect can be described by the ECM.

C. Magnetic Permeability of Permalloy Composite Materials

The complex permeability spectra of bulk Permalloy and its composite materials are shown in Fig. 5: (a) real parts μ_r', and (b) imaginary parts μ_r''. The real part μ_r' of bulk Permalloy is about 300 at 100 Hz and decreases rapidly as frequency increases, it becomes almost unity at about 1 MHz. The imaginary part μ_r'' has the same order magnitude and shows a maximum at around 400 Hz. On the other hand, the μ_r' value of Permalloy composite materials is almost constant up to several tens MHz, and then it decreases with frequency. The imaginary part μ_r'' has a broad maximum. The frequency dispersion of permeability can be described by the domain wall and gyromagnetic spin resonance [9]. In a metal ferromagnetic, eddy currents cause the decrease of the penetration depth of the electromagnetic field; and magnetic permeability decreases with frequency. For high conductive metal, the skin depth δ can be approximated by

$$\delta = \sqrt{\frac{2}{\omega\mu\sigma}} \qquad (3)$$

where ω, σ and μ are the angular frequency ($\omega = 2\pi f$), electrical conductivity and permeability, respectively. The estimated skin depth (3) of bulk Permalloy using measured permeability and conductivity values is 3.2 mm at 10 kHz. The dashed line in Fig. 5 (a) indicates the $f^{-1/2}$ behavior; the frequency dependence of μ_r' follows (3). Thus the permeability dispersion of bulk Permalloy can be described by the skin effect. On the other hand, the skin depth of the 57.1 vol.% composite, which was roughly estimated assuming the conductivity value of 4.0 S/cm and μ_r = 18.1 is 5.9 mm at 10 GHz; and the skin depth below 10 GHz is larger than the sample size. However, since the conductivity of 57.1 vol.% composite is much smaller than that of the bulk permalloy, the approximation (3) may not be applicable for the Permalloy composites. In fact, the μ_r' of the 57.1 vol.% composite shows a rapid decrease above 20 MHz as shown in Fig.5 (a), eddy current effect cannot be ignored in high particle content composites.

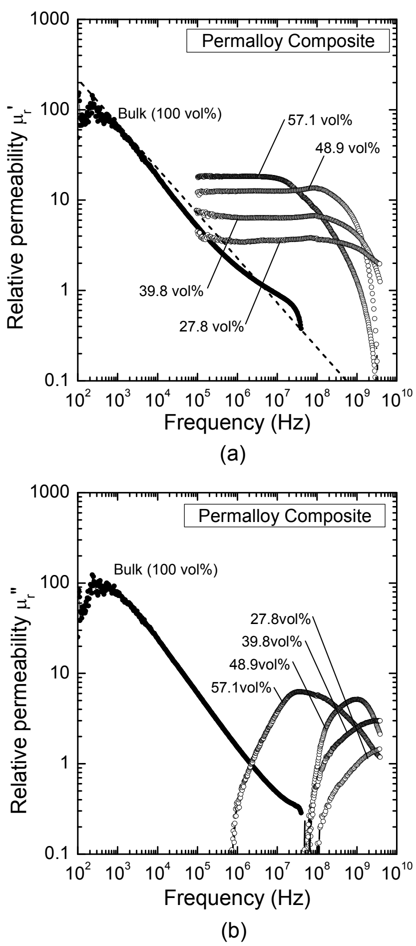

Fig. 5. Relative complex permeability spectra of metal Permalloy (bulk) and its composite materials: (a) real part, (b) imaginary part.

D. Effect of the Heat Treatment of Embedded Particles on the Permeability and Permittivity Spectra

The complex permeability spectra of 60 vol.% Permalloy composite containing the heat-treated particles, as well as of the composite with 57.1 vol.% of untreated particles, are shown in Fig. 6. The 60.0 vol.% composite exhibits the permeability dispersion above several hundred MHz, which indicates that the skin effect was suppressed. Therefore, the high-frequency permeability of Permalloy composite

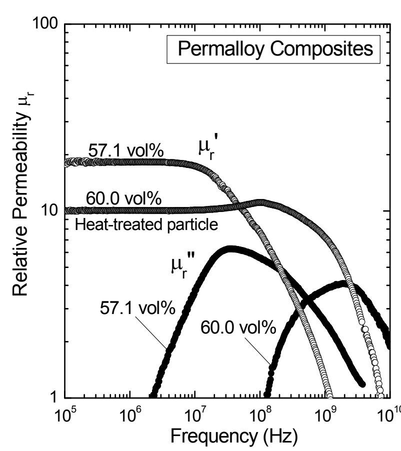

Fig. 6. Relative complex permeability spectra of Permalloy composite materials containing non-treated and heat treated particles.

materials can be improved by the surface treatment of embedded particles.

Fig. 7 shows the frequency dispersion of the real part of permittivity ε_r' for the heat-treated 60 vol.% composite as well as the imaginary part ε_r" for the non-treated 57.1 vol.% one as a function of frequency. The inset shows the complex permittivity spectra of Permalloy composites containing high

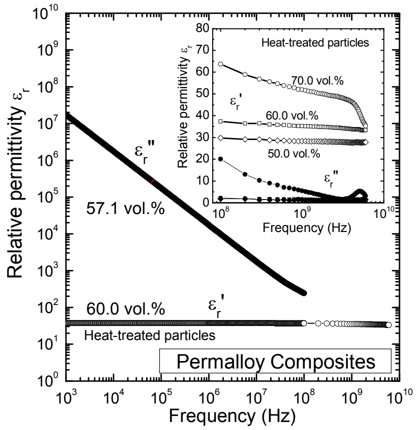

Fig. 7. Relative complex permittivity spectra of Permalloy composite materials containing non-treated and heat treated particles. The inset shows the relative permittivity spectra of the heat-treated particle composites at several particle contents.

volume fraction of heat-treated particles. Permittivity frequency dispersion characteristic changes drastically by the heat-treatment of the embedded particles, and as a result, the insulating property is maintained up to high particle content of at least 70 vol.%.

IV. Conclusion

The electromagnetic properties of Cu and Permalloy composite materials have been studied over a broad frequency range from tens Hz up to about 10 GHz. The variation of the electrical conductivity and permittivity with particle content indicate the insulator-metal transition due to the percolation effect of embedded metal particles. The percolation threshold of volume fraction φ_c of Permalloy composite materials can be increased by the surface treatment of embedded particles. The effective cluster model can be applied to explain the percolation effect in permittivity.

High-frequency magnetic permeability spectra of the high particle content Permalloy composites can be affected by the skin effect produced by the eddy currents. Surface oxidation of the embedded metal particles shifts the percolation threshold to higher particle content. Thus the high-frequency permeability can be improved by the heat-treatment of metal particles in the composite materials. It is important that the composites with heat-treated particles remain in an insulating phase at higher contents of ferromagnetic inclusions. This allows for applying such materials with improved magnetic and absorptive properties to solving practical EMC problems, where absorbing materials should be non-conducting in principle, *e.g.*, on shields of cables, on tops of noise-producing chips or heatsinks [17].

Acknowledgments

A part of this research was supported by Grant-in Aid from the Japanese Ministry of Education, Science, Sports, and Culture No. 21360340. M. Koledintseva also acknowledges the support by the U.S. National Science Foundation under Grant No. 0855878.

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