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# Negative permeability spectra in Permalloy granular composite materials

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Complex permeability spectra of Permalloy granular composite materials have been studied in the microwave frequency range. The heat-treated Permalloy particles in the air at several hundreds of °C have a high surface electrical resistance; the eddy current effect in the high frequency permeability spectra can be suppressed in the composite structure containing the percolated particles. A negative permeability has been obtained above 5 GHz due to the natural magnetic resonance in the 70 vol % particle content composite material. In this content, electrical permittivity spectra show a nonmetallic characteristic. This permeability dispersion can be applied for the left-handed media. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2198113]

The left-handed metamaterial, which simultaneously has negative permeability and permittivity in the microwave frequency range, has been the subject of considerable interest in these days. The peculiar propagation of electromagnetic waves such as the negative refraction of electromagnetic waves occurs in this medium.[1] A metamaterial using a periodic structure composed of the split ring resonator (a negative permeability) and the metal wire array (a negative permittivity) has been proposed in order to obtain the left-handed media (LHM),[2] and the negative refraction of the microwave has been observed in this artificial medium.[3] On the other hand, a metallic granular composite material composed of nanometer-sized particles has been theoretically investigated as a candidate for the LHM.[4] If there exists the composite material which has negative permeability and permittivity as an intrinsic magnetic and dielectric properties, small sized microwave devices such as high potential microwave antenna[5] can be developed using these left-handed properties. We have been studying the complex permeability spectra of ferrites, magnetic metals, and their composite materials considering the application for the electromagnetic compatibility (EMC) devices such as electromagnetic wave absorbers or shielding materials.[6,7] In some ferrite materials, the negative permeability spectra can be obtained as an intrinsic magnetic property, i.e., the ferromagnetic resonance of magnetic materials.[8] Further, the possibility of a left-handed material has been investigated using the frequency dispersion of permeability in yttrium iron garnet (YIG) under external magnetic field combined with the permittivity spectrum of the metal wire array composite.[9]

Though the magnetic metal such as Permalloy has a large permeability in the low frequency region, permeability rapidly decreases with increasing frequency due to the effect of eddy current.[7] Meanwhile low content Permalloy composite materials have a relatively high permeability in the high frequency region because of the high electrical resistivity. However, for the high particle content Permalloy composites above about 50 vol %, the metallic property takes place due to the percolation of embedded particles; the Ohmic contact between particles brings about the eddy current effect in the high frequency permeability. In this work, the complex permeability and permittivity spectra of Permalloy composite materials containing surface treated granular particles have been studied in the microwave frequency range for preparing the material which has the negative permeability spectra.

A commercially available $Ni_{45}Fe_{55}$ powder was used for Permalloy composite materials. Particle shape and diameter were examined by a scanning electron microscope (SEM). The particle shape is spherical and the mean particle diameter $d_m$ is 2.53 $\mu$m. Permalloy particles were heat treated in air using an electric furnace at the temperature range from 100 to 600 °C in order to make the oxidized surface. Permalloy composite materials were prepared by mixing Permalloy powder with polyphenylene sulfide (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. Obtained samples were cut into a toroidal form (inner diameter is 3 mm, outer diameter is 7 mm) and a controlled thickness of about 1 mm in order to avoid the dimensional resonance of the electromagnetic wave in a coaxial line.

The powder x-ray diffraction measurement was performed to investigate the heat-treated effect. Magnetization curve was measured using a vibrating sample magnetometer. The apparent ac electrical resistivity $\rho_{ac}$ of compacted Permalloy particles was measured by a two-terminal method using a coaxial electrode and an impedance analyzer in the frequency range from 100 Hz to 40 MHz. The schematic diagram of a coaxial electrode is shown in the inset of Fig. 2. Permalloy particles were set in the coaxial electrode in the air and pressed on the top of the Teflon plug with the pressure of 15 MPa. The value of $\rho_{ac}$ was determined using the following formula:

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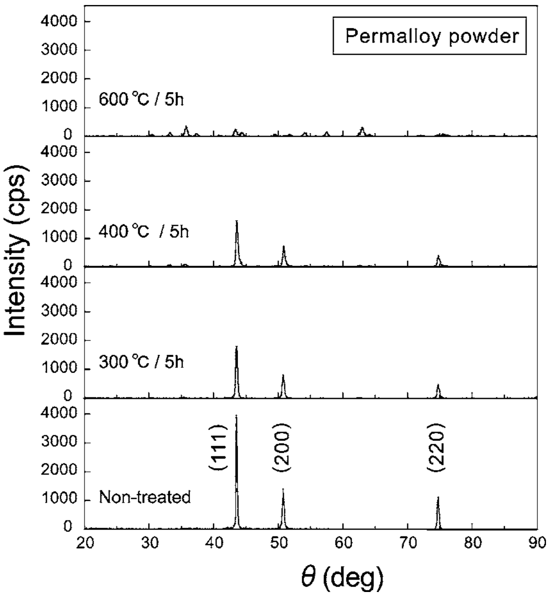

FIG. 1. Powder x-ray diffraction profiles for the Permalloy which are nontreated and heat treated at 300, 400, and 600 °C for 5 h.

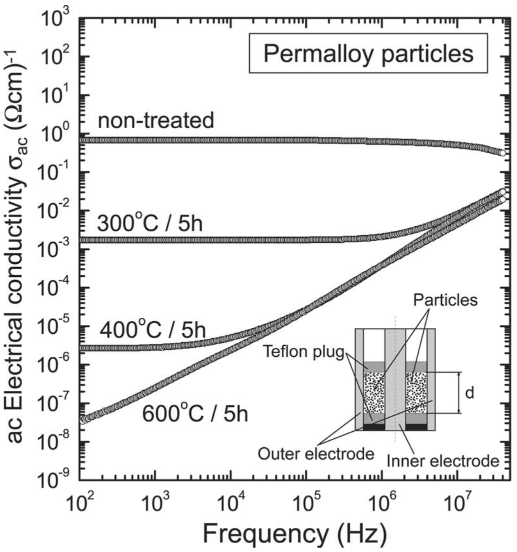

FIG. 2. The ac electrical resistivity for the Permalloy particles as a function of frequency, which are nontreated and heat treated at 300, 400, and 600 °C for 5 h.

$$\rho_{ac} = \frac{V}{I}\frac{2\pi d}{\ln(r_2/r_1)}, \tag{1}$$

where $d$ is the sample length, $I$, $V$, and $r_1$ and $r_2$ are the applied current, the measured voltage, and the inner and outer radii of the coaxial electrode, respectively. The relative complex permeability ($\mu^* = \mu' - i\mu''$) and permittivity ($\varepsilon^* = \varepsilon' - i\varepsilon''$) of composite materials were measured by a coaxial line technique in the frequency range from 100 MHz to 10 GHz using a network analyzer.

Figure 1 shows the x-ray diffraction profiles of Permalloy powders, which are nontreated and heat treated in air at 300, 400, and 600 °C for 5 h. For the nontreated Permalloy powder, the x-ray diffraction profile shows the characteristic reflections of the Ni–Fe alloy (fcc structure) from the (111), (200), and (220) planes. When the heat-treating temperature increases, the intensity of these peaks decreases and at 600 °C, many extra peaks appear. This indicates that the heat treatment up to 500 °C does not affect the main crystal structure. The apparent ac electrical conductivity $\sigma_{ac}$ of compacted Permalloy particles is shown in Fig. 2 as a function of frequency. For the nontreated particles, the $\sigma_{ac}$ is about $0.8\ (\Omega\,\mathrm{cm})^{-1}$ up to 40 MHz. In the heat-treated particles at 600 °C, the $\rho_{ac}$ indicates about $3.0 \times 10^{-8}\ (\Omega\,\mathrm{cm})^{-1}$ at 100 Hz and increases with increasing frequency. The electrical conduction mechanism of the compacted particles can be attributed to the variable range hopping through the oxide layer in percolating clusters; the frequency variation of $\sigma_{ac}$ can be explained by the power-law exponent in percolating clusters.[10–12] Considering the x-ray analysis results, it can be concluded that the Permalloy particle surface is oxidized and contact electrical resistance between particles increases by the heat treatment.

Magnetization curves for the nontreated and the heat-treated particles in air at 300, 400, and 600 °C for 5 h are shown in Fig. 3. The magnetization of nontreated particles saturates above of about 6 kOe and shows the value of about 150 emu/g. On the other hand, for the heat-treated particles at 300 °C, the saturation magnetization is about 142 emu/g. The saturation magnetization decreases with increasing the heat-treating temperature. It is considered that the decrease of the saturation magnetization of the Permalloy particle is owing to the decrease of the volume which contributes to the magnetization in the particle. From these electrical conductivity and magnetization measurement results, the heat-treated particles at 300 °C were used for preparing the Permalloy composite materials since they have enough electrical resistivity to suppress the eddy current effect and the small reduction of magnetization.

Figure 4 (a) shows the complex permeability spectra of Permalloy composite materials containing the heat-treated particles at 300 °C in the frequency range from 100 MHz to 10 GHz. The real part $\mu'$ of 70 vol % particle content composite is about 17 at 100 MHz and decreases with increasing frequency and shows a negative value above 5 GHz; the minimum value of $\mu'$ is about −1. On the other hand, the imaginary part $\mu''$ shows a maximum around 2 GHz. Generally, the frequency dispersion of permeability can be expressed by the superposition of the two types of magnetic resonance induced by the domain wall motion and

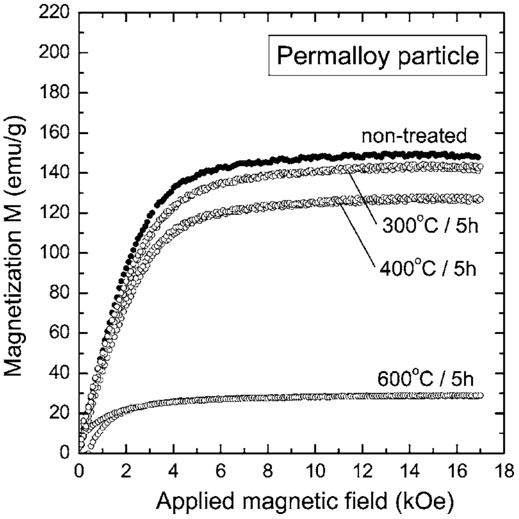

FIG. 3. Magnetization curves for the Permalloy particles which are nontreated and heat treated at 300, 400, and 600 °C for 5 h.

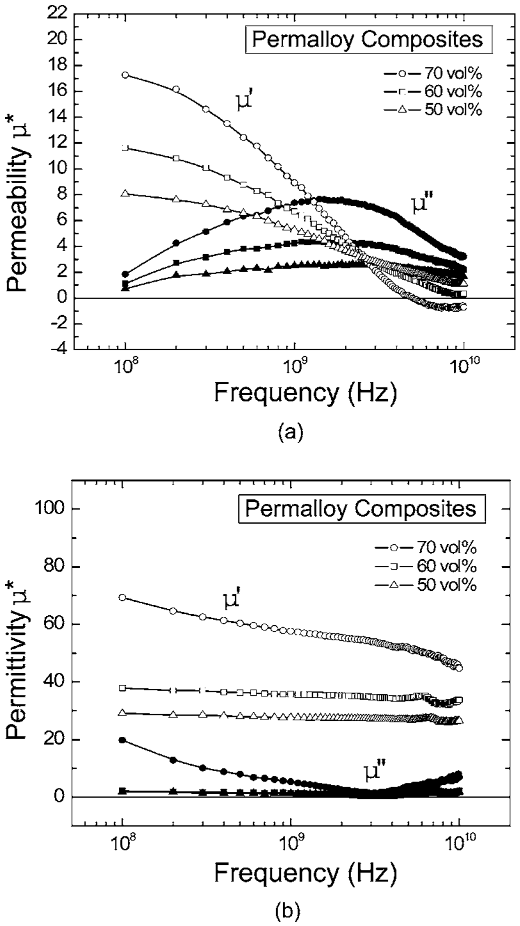

FIG. 4. Complex permeability (a) and permittivity (b) spectra for the Permalloy composite materials containing heat-treated particles at 300 °C for 5 h.

the gyromagnetic spin rotation (natural resonance). The relaxation-type frequency dispersion of permeability is observed in the absence of the external magnetic field due to high damping coefficient of the resonances in ferrite composite materials.[13] By the numerical fitting of permeability data to a simple resonance formula consisting of these two resonance components, the contribution of the domain wall and magnetic spin to the permeability spectra has been estimated in the spinel ferrite and Permalloy composite materials.[13,14] In this estimation, it was obtained that the damping of gyromagnetic spin resonance, which mainly contributes to permeability in the high frequency range, is smaller in Permalloy composites than that in spinel ferrite composites. In addition, the spin resonance frequency of Permalloy composites is higher than that in spinel ferrite composites. Thus the resonance-type frequency dispersion of permeability accompanying the $-\mu'$ can be obtained in the insulating state in Permalloy composite materials without the eddy current effect. With the decrease of Permalloy content, the permeability value decreases in the measured frequency range and the frequency at which $\mu''$ shows a maximum slightly shifts higher frequency. This frequency shift is mainly attributed to the resonance frequency shift of the spin component by adding the demagnetizing field, which is induced in Permalloy particles by the alternating magnetic field, to the internal magnetic anisotropy field.[15]

Complex permittivity spectra for Permalloy composite materials with heat-treated particles in the frequency range from 100 MHz to 10 GHz are shown in Fig. 4(b). In the 50 and 60 vol % composites, the $\varepsilon'$ is almost constant and the $\varepsilon''$ is almost zero up to 10 GHz; in the 70 vol % composite, the permittivity spectra show a small frequency dispersion in the measurement frequency range. This indicates that these composites have insulating property. In the spherical particle granular composite material, the particle content of 60 vol % is higher than the percolation limit of the embedded particles;[16] the mechanical contact among the embedded particles is established in this composite material. In the case of the nontreated Permalloy composite, Ohmic contact of the percolated particles brings about the metallic conduction of the composite and the $\varepsilon''$ shows the frequency dispersion of inversely proportional to the frequency.[14] However, in the heat-treated particle composite materials, non-Ohmic contact among the particles makes the high electrical resistivity and insulating properties can be maintained even in the high particle content. Accordingly, the eddy current in the composite is suppressed and natural resonance owing to the internal magnetic anisotropy field in the Permalloy particles can be seen in the permeability spectra. Furthermore, a large dielectric constant value more than 60 is observed in the 70 vol % composite materials, as shown in Fig. 4(b). Since the $\varepsilon'$ value of the host resin is 3 and no frequency dispersion is observed up to 6 GHz, the enhancement of the $\varepsilon'$ value can be attributed to the dielectric polarization in the embedded metallic particles. This type of enhancement is observed in the artificial dielectric materials.[17] The small frequency dispersion of permittivity in the 70 vol % composite material may be originated by the small metallic part in the composite or the tunneling current between particles.[18] The investigations of the left-handed material using the composite structure of Permalloy composites and thin metal wire array are now in progress.

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