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The electric properties of low-magnetic-loss magnetic composites containing Zn–Ni–Fe particles

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Abstract

Recently, magnetic composites consisting of magnetic particles dispersed in a polymer matrix have been widely discussed for miniaturizing high-frequency electronic components such as antennae. Previously, we investigated the influence of the manufacturing process on the homogeneous dispersion of magnetic particles in the polymer and on the magnetic properties of the magnetic composites. In order to miniaturize electronic components, it is crucial to be able to independently control the permeability and permittivity in magnetic composites. This paper investigates the anisotropy and frequency dependence of the dielectric properties of magnetic composites fabricated from 20 vol% $Zn_5Ni_{75}Fe_{20}$ flaked particles. The permittivity of magnetic composites fabricated from $Zn_5Ni_{75}Fe_{20}$ flaked particles is anisotropic: at 1 GHz, the relative permittivities parallel and perpendicular to the plane of the specimens are 27.2 and 16.9, respectively. The permittivity varied little between frequencies of 50 MHz and 10 GHz.

1. Introduction

The rapidly growing demand for multifunctional and compact mobile communication devices such as mobile phones requires further miniaturization and higher density mounting of electronic components. The use of magnetic composites with high permeabilities and low losses is anticipated to be one way to meet these requirements. Recently, a method for preparing magnetic composites that involves adding an electrical insulator to a ferromagnetic powder has been investigated to reduce the magnetic losses at high frequencies [1–4].

When designing electronic components it is important to know not only the magnetic characteristics of the magnetic composite but also its dielectric characteristics, because wavelength shortening and the impedance of electronic components depend on $\sqrt{(\mu\varepsilon)}$ and $\sqrt{(\mu/\varepsilon)}$, respectively.

The dielectric characteristics of magnetic composites containing spherical, soft-magnetic particles such as NiFe and Fe_3O_4 have been reported in several papers [5–7]. Gokturk *et al* investigated the dielectric properties of a composite consisting of a thermoplastic elastomer containing 40 vol% nickel–iron alloy powder. This magnetic composite had a relative permittivity of 25 at 5 kHz. For frequencies in the range from 5 kHz to 1 MHz, the dielectric constant of the composite was insensitive to the frequency [5]. Nedkov *et al* reported the dielectric properties of a magnetic composite containing Fe_3O_4; it had a relative permittivity of 6 at 7 GHz. The complex permittivity of this composite had no measurable frequency dependence between 7 and 17 GHz [6]. Zhen *et al* mixed 15 vol% FeNi alloy nanoparticles that had been prepared by thermal hydrogen reduction with 85 vol% wax. The resulting magnetic composites had a real part of the permittivity of 6 in the frequency range of 2–18 GHz; however, no frequency dependence was observed [7].

Theoretical approaches for estimating the permittivity of heterogeneous mixtures consisting metal particles dispersed in a dielectric material have been reported. Lal and Parshad tested previously developed expressions for spherical metallic inclusions (e.g., the Bruggeman's equation [8],

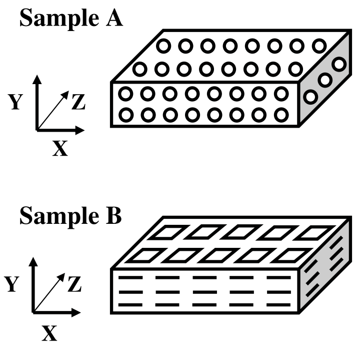

Figure 1. Schematic diagram of samples A and B indicating the direction of axes.

De Loor's equation [9] and Van Beek's equation [10]), and they developed a new theoretical expression that accounts for the shape of the dispersed metal particles. Their calculations predict that mixtures consisting of non-spherical metal particles should have higher permittivities than spherical metal particles [11]. Kubo *et al* performed calculations using commercial electromagnetic wave analysis and found that artificial dielectrics containing flaked-metal strips have high anisotropies and permittivities [12]. However, these two studies did not contain any experimental investigations.

We have investigated the magnetic properties of a composite consisting of Zn–Ni–Fe ($Zn_5Ni_{75}Fe_{20}$) flaked particles dispersed in a polymer which is an electrical insulator. We fabricated a magnetic composite with a high permeability and low magnetic losses at high frequencies by crushing the agglomerated particles so that most of the particles were deformed into flaked particles [13]. We also investigated the influence of the fabrication process on the homogeneous distribution of flaked particles in a polymer. By improving the fabrication process, the magnetic losses of the composite could reduced due to its fine microstructure and the dispersibility of flaked particles in the polymer could be enhanced [14]. Our developed materials can be expected to be applied in electronic components because of their low magnetic loss. However, we have not yet investigated the dielectric properties of magnetic composites containing Zn–Ni–Fe flaked particles.

This paper reports the anisotropy and frequency dependence of the dielectric properties of a magnetic composite containing Zn–Ni–Fe flaked particles.

2. Experimental details

2.1. Raw materials

Zn–Ni–Fe alloy particles were prepared by reducing nickel chloride hexahydrate ($NiCl_2{\cdot}6H_2O$), iron chloride tetrahydrate ($FeCl_2{\cdot}4H_2O$) and zinc nitrate hexahydrate ($Zn(NO_3){\cdot}6H_2O$) in an aqueous solution. The chemical compositions used were 5 mass% Zn, 75 mass% Ni and 20 mass% Fe. The particles were spherical in shape and had a median size of 250 nm. The Zn–Ni–Fe particles had a coercive force of 9.04 kA m^{-1} and a saturation magnetization of 76.4 A kg^{-1}.

2.2. Preparation of magnetic composites

The mixing solution consisted of xylene and cyclopentanone with cyclo-olefin polymer (permittivity and dielectric loss factor at 1 GHz are 2.6 and 0.0099, respectively) used as the solvent. To investigate the effect of particle shape on the anisotropy and frequency dependence of the dielectric properties, the following samples were prepared.

(1) $Zn_5Ni_{75}Fe_{20}$ particles were added to the solvent. A slurry of spherical particles was prepared using an ultrasonic homogenizer for 60 min (sample A).
(2) $Zn_5Ni_{75}Fe_{20}$ particles and 200 μm-diameter zirconia spheres (used as the grinding medium) were added to the solvent. A slurry of flaked particles was prepared using a high-speed rotation/revolution mixer at a stirring rate of approximately 1300 m s^{-1} for 150 min (sample B).

After removing the grinding medium, the slurries were mixed with a thermosetting polymer until the volume content of $Zn_5Ni_{75}Fe_{20}$ particles became 20 vol%. An approximately 50 μm thick film was fabricated on the polyester film by the conventional doctor blade [15] method at a transfer rate of 100 mm min^{-1}. The blade clearance was controlled to be 800 μm. The formed film was dried at 323 K in the atmosphere for 60 min. Approximately 2 mm thick magnetic composites were prepared by laminating the films by hot-pressing them at 433 K under a uniaxial pressure of 0.1 MPa for 40 min in a vacuum of 0.01 MPa.

2.3. Magnetic composite measurements

The microstructures of the magnetic composites were observed using a scanning electron microscope (SEM). Figure 1 shows a schematic of the samples indicating the directions of the axes. The X-axis is parallel to the long axis of the sheet, while the Y-axis is normal the sheet. To investigate the influence of particle shape on the dielectric properties, the dielectric properties along the Y-axis of the samples (height: 1.5 mm; width: 20 mm; length: 20 mm) were measured by the parallel-plate capacitor method using an impedance analyzer (4291A, Hewlett-Packard) for frequencies in the range from 50 to 1000 MHz. The dielectric properties along the X-axis in sample B were also measured to confirm that the permittivity is anisotropic in sample B. Furthermore, to investigate the frequency dependence at frequencies over 1 GHz, the dielectric properties along the X-axis in the samples (height: 0.05 mm; width: 2 mm; length: 85 mm) were measured by the cavity resonator method based on a perturbation theory by using a vector network analyzer (8791ES, Agilent) at frequencies between 1 and 10 GHz.

3. Results and discussion

3.1. Characteristics of magnetic composites

Figure 2 and table 1 show the microstructures and magnetic properties of both samples, respectively. The microstructure of sample A consists of spherical particles, while that of sample B consists of flaked particles. The long axes of the

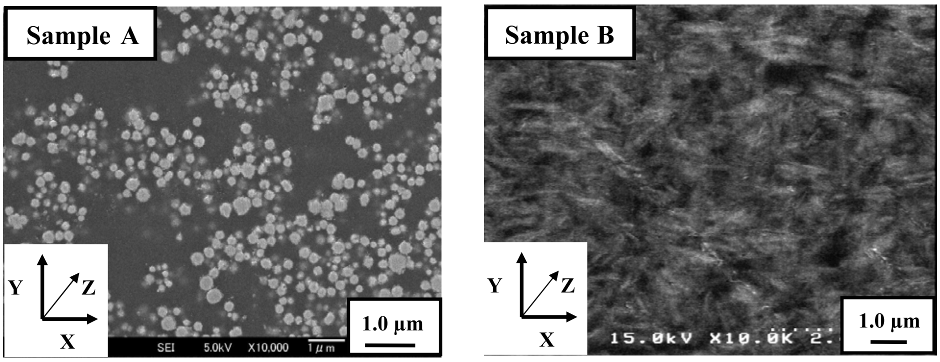

Figure 2. Cross-sectional SEM images of samples A and B.

Table 1. Characterization of magnetic composite materials (20 vol%).

Sample	Microstructure characterization		Magnetic properties @ 1 GHz	
	Average grain size (μm)	Aspect ratio	Permeability	Loss factor
A	0.25	1	2.54	0.128
B	0.59	5	3.24	0.063

flaked particles in sample B are aligned parallel to the X-axis; this orientation was caused by the shearing stress of the doctor blade that was used to fabricate the films. The average grain size and aspect ratio are respectively 0.25 μm and 1 for sample A and they are respectively 0.59 μm and 5 for sample B. The increase in the ratio of the diameter to the thickness of the particles in sample B is due to the spherical particles being deformed into flaked particles by the shear stress among the zirconia spheres during preparation of the slurry of flaked particles. The magnetic properties listed in table 1 were measured by the parallel line method. Sample B has a higher permeability and a lower magnetic loss factor than sample A. The excellent magnetic properties of sample B are considered to be due to the low demagnetizing factor and the homogeneous dispersion of the Zn–Ni–Fe particles. Generally, the permeability is decreased by the demagnetizing field. The demagnetizing field is proportional to the demagnetizing factor depending on the aspect ratio of the magnetic particles. Though the demagnetizing factor is equal to 1/3 for spherical particles, the flaked particles are smaller than 1/3 in the direction of the long axis (X axis) [16].

3.2. *Dielectric properties of the magnetic composites*

3.2.1. Influence of particle shape on dielectric properties. Figure 3 shows the dielectric properties along the Y-axis in sample A (electric field configuration of the Y-axis) in the range 50–1000 MHz. The relative permittivity and dielectric loss factor at 500 MHz are 5.5 and 0.015, respectively. The dielectric properties along the Y-axis in sample B (electric field configuration of the Y-axis) are shown in figure 4. The relative permittivity and dielectric loss factor at 500 MHz are 16.9 and 0.023, respectively. Sample B has a higher relative permittivity along the Y-axis than sample A. For magnetic composites in which magnetic particles are dispersed in a polymer, a capacitive effect operates between the magnetic particles in the polymer. Thus, the permittivity of a magnetic composite is strongly related to the distance between the particles; specifically, as the distance between the particles is reduced, the relative permittivity increases. Figure 2 shows the interparticle distance is smaller in sample B than in sample A.

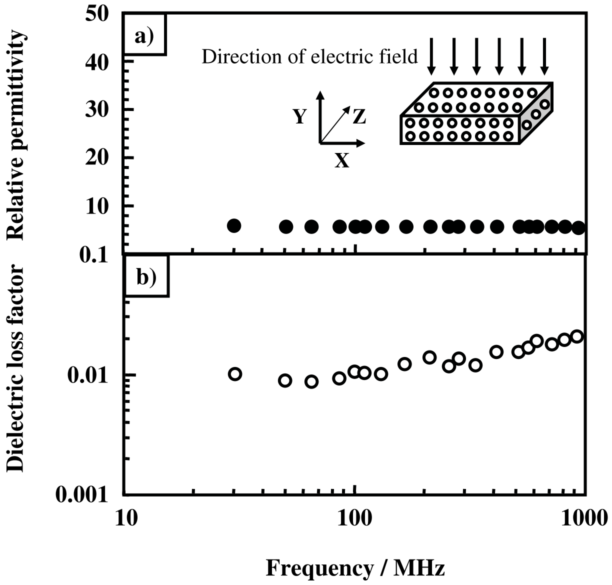

Figure 3. (a) Relative permittivity and (b) dielectric loss factor of sample A along the Y-axis as a function of frequency.

The dielectric properties along the X-axis in sample B (electric field configuration of the X-axis) are shown in figure 5. Typical values for the relative permittivity and the dielectric loss factor at 500 MHz are 27.2 and 0.016, respectively. The relative permittivity along the X-axis is higher than that along the Y-axis in sample B. This anisotropy of the permittivity in sample B was observed for frequencies in the range 50–1000 MHz. According to [12], the interparticle distance decreases when the aspect ratio of the metallic particles becomes larger, thereby increasing the capacitance between the particles. This capacitance strongly affects the relative permittivity. Moreover, the polarization parallel to the

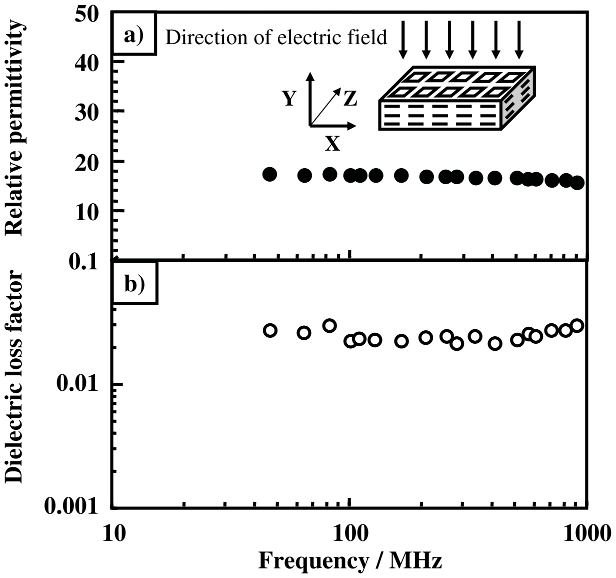

Figure 4. (a) Relative permittivity and (b) dielectric loss factor of sample B along the Y-axis as a function of frequency.

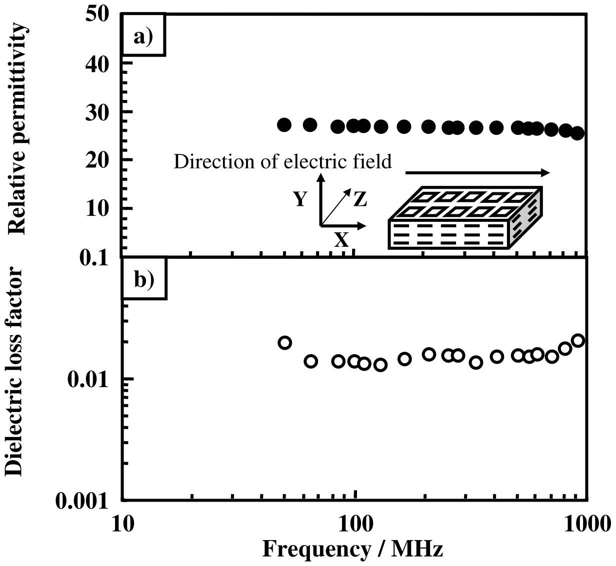

Figure 5. (a) Relative permittivity and (b) dielectric loss factor of sample B along the X-axis as a function of frequency.

long axis of a flat particle is larger than that parallel to the short axis. This effect explains the observed permittivity anisotropy in magnetic composites that consist of flaked particles, such as sample B.

3.2.2. Frequency dependence of dielectric properties. Figure 6 shows the dielectric properties along the X-axis in sample A (electric field configuration of the X-axis) in the frequency range from 1 to 10 GHz. The relative permittivity and dielectric loss factor at 5 GHz are 4.8 and 0.015, respectively. Figure 6 is consistent with the dielectric properties shown in figure 3. No difference in the permittivities along the X and Y axes was observed. Figures 3 and 6 reveal that there is no frequency dependence in the relative permittivity in the range from 50 MHz to 10 GHz. By contrast, the dielectric loss factor increases slightly when the frequency increases for frequencies under 1 GHz. However, this tendency of the dielectric loss factor was slightly exaggerated due to the specification limit of the impedance analyzer.

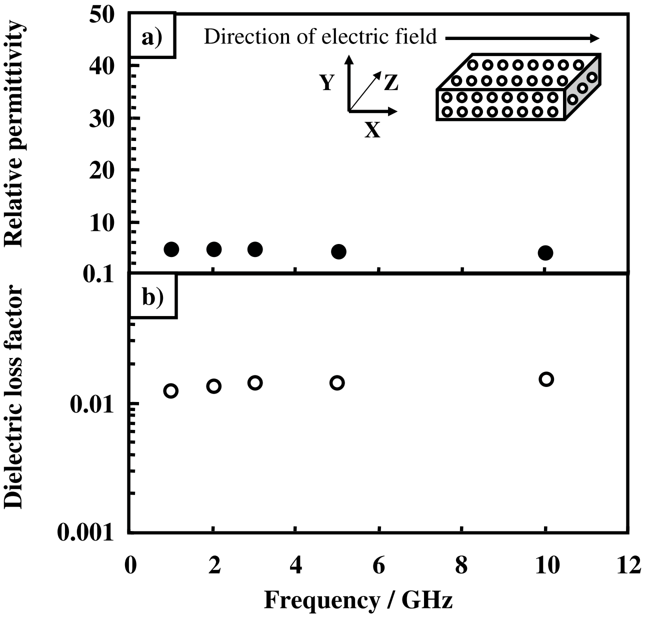

Figure 6. (a) Relative permittivity and (b) dielectric loss factor of sample A along the X-axis as a function of frequency.

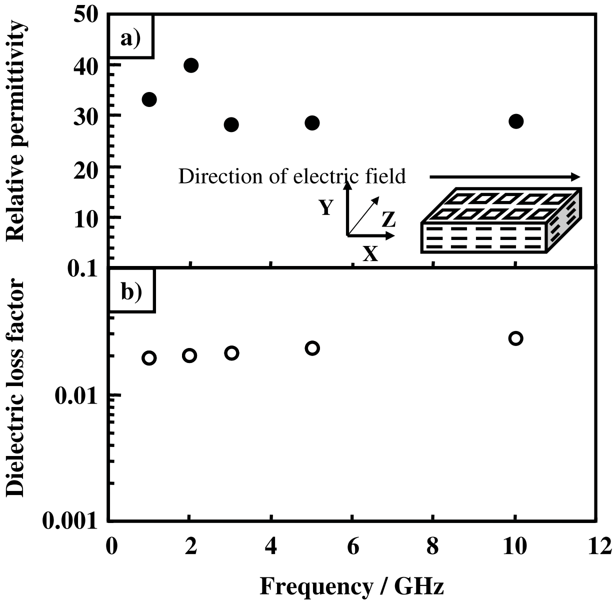

Figure 7. (a) Relative permittivity and (b) dielectric loss factor of sample B along the X-axis as a function of frequency.

The dielectric properties along the X-axis in sample B (electric field configuration of the X-axis) are shown in figure 7. The relative permittivity and dielectric loss factor at 5 GHz are 28.3 and 0.024, respectively. Similar to sample A, no frequency dependence of the relative permittivity was observed in sample B.

4. Conclusions

Magnetic composites containing $Zn_5Ni_{75}Fe_{20}$ flaked particles were found to have an anisotropic permittivity and the permittivity was found to be frequency dependent at high frequencies. The permittivity parallel to the long axis of the specimens is higher than that perpendicular to the plane of the specimens. This result is consistent with previous findings obtained using commercial electromagnetic wave analysis software for an artificial dielectric containing metallic strips. The relative permittivity varied little with frequency.

It is important for both the permittivity and permeability to be anisotropic when designing electronic components fabricated from magnetic composites consisting of $Zn_5Ni_{75}Fe_{20}$ particles because it is desirable for electronic components with a high impedance in order to increase the permeability and reduce the permittivity, and the permittivity and permeability can be controlled by the particle shape and its direction.

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