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Polymer composite sheet and BaCoTiFe$_{10}$O$_{19}$ thin films may have the potential for use at GHz frequencies and as anisotropic-electromagnetic wave suppression materials.

ABSTRACT

We prepared a high-quality magnetic BaCoTiFe$_{10}$O$_{19}$ ferrite-based composite sheet and BaCoTiFe$_{10}$O$_{19}$ thin films exhibiting in-plane magnetic anisotropy. In-plane magnetic anisotropy was realized by controlling particle orientation to achieve high-resonance frequency values. The dynamic permeability of the samples in the GHz-frequency region was evaluated by the short-circuited microstrip line method up to 30 GHz. Controlling the propagation of electromagnetic waves with GHz frequencies is extremely difficult. Nevertheless, the ferromagnetic resonance phenomena were observed, and the obtained sheet showed a resonance frequency value of approximately 4 GHz. However, the intrinsic ferromagnetic resonance behaviors were not elucidated for the thin films. The obtained BaCoTiFe$_{10}$O$_{19}$ powder/polymer composite sheet and BaCoTiFe$_{10}$O$_{19}$ thin films may have the potential for use at GHz frequencies and as anisotropic-electromagnetic wave suppression materials.

Keywords: Magnetoplumbite-structured Ferrite, Dynamic Permeability Behaviors, High-frequency Electromagnetic Waves, Short-circuited Microstrip Line Method

1. Introduction

We have been intensively working on a creation of magnetic materials based on novel principles using the interaction between magnetism and electromagnetic waves in last five years. In these studies, we attempted to precisely control the material structures to optimize the physical properties of the materials. In this study, we report the dynamic magnetic properties of the materials in the GHz-frequency region induced by ferromagnetic resonance (FMR) under the irradiation of GHz-frequency electromagnetic waves. These characteristics are important as permeability and FMR determine the applicable and upper limit of frequency bands of magnetic materials. For example, FMR should be in the MHz region for a magnetic-based electromagnetic wave shield for the use of wireless power transmission circuits, whereas magnetic sheets with FMR in the GHz region can be applied as electromagnetic wave absorbers for mobile phones.

Ferrites, which are iron oxide-based materials, are promising for high-frequency devices owing to their wide FMR in MHz to GHz-frequency region. The surface resistivity of ferrites is sufficiently high, suggesting that the influence of eddy currents is negligible under the irradiation of the sample by high-frequency electromagnetic waves. This is one of the greatest advantages for ferrites applied to high-frequency technologies. Magnetoplumbite-structured ferrites with a chemical formula of $A$Fe$_{12}$O$_{19}$ ($A$ = alkali or alkaline-

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earth metal) are expected to have the $f_e$ value of ~50 GHz.\textsuperscript{6-12} One of the magnetoplumbite-structured ferrites, i.e., an Sm-doped single-crystal SrFe$_{12}$O$_{19}$, demonstrated an $f_e$ value of 53 GHz.\textsuperscript{13} Due to the difficulties in sufficiently controlling the measurements in GHz-frequency electromagnetic waves, the FMR phenomena and permeability behaviors of magnetic materials in this frequency region are not yet well established, though the technological use of GHz-frequency electromagnetic waves is essential.

In this study, we evaluate the dynamic permeability of magnetoplumbite-structured ferrites under the irradiation of electromagnetic waves in the GHz-frequency region using the short-circuited microstrip line (MSL) method.\textsuperscript{12,14,15} We aimed to understand how the structure of magnetoplumbite-structured ferrites affects the dynamic magnetic behaviors in the GHz region. To clarify this issue, we selected BaCoTiFe$_{12}$O$_{19}$, which is one of the magnetoplumbite-structured ferrites. To obtain fundamental dynamic permeability behaviors, we studied a BaCoTiFe$_{12}$O$_{19}$ powder/polymer composite sheet sample as a “reference.” Afterward, we studied the dynamic permeability behaviors of BaCoTiFe$_{12}$O$_{19}$ thin films as they can more flexibly alter the structure leading to magnetic anisotropy, compared with the powder/polymer composite sheet sample.

2. Strong Correlation between the Structure, Magnetic Anisotropy, and High-frequency Magnetic Behaviors

From the high-frequency electromagnetic wave, a high-frequency magnetic field ($H_f$) is generated as well as a high-frequency alternating current ($I_f$).

In magnetic thin films-sheet-shaped samples, a strong correlation was observed between the structure of the sample, magnetic anisotropy, and high-frequency magnetic behaviors, such as FMR, permeability behaviors, and $f_e$. The $f_e$ value of the samples is considerably affected by the “direction” of magnetic anisotropy. Takeda has reported that the $f_e$ value of the samples varies in accordance with the direction of magnetic anisotropy.\textsuperscript{36} When the sample exhibits in-plane magnetic anisotropy ($H_{a//}$), and $H_f$ is disposed perpendicular to the magnetic domain walls ($W$) of the sample, $f_e$ can be described by the following formula:

$$f_e = \gamma H_{a//} \left[ H_{a//}, H_f \perp W \right]$$

where $\gamma$ denotes the gyromagnetic ratio, 2.8 MHz/Oe. However, if the magnetic sample possesses $H_{a//}$, the configuration of $H_f$ and $W$ forms that are parallel, $f_e$ can be expressed as

$$f_e = \gamma \sqrt{(H_{a//} + 2a\pi M_0)(H_{a//} + 4a\pi M_0)} \left[ H_{a//}, H_f \parallel W \right]$$

where $4a\pi M_0$ denotes the saturation magnetization, and $a$ denotes the number defined as $0 \leq a \leq 1$, which depends on the size of magnetic domains. Moreover, when magnetic anisotropy is in the out-of-plane direction of the surface of magnetic thin films/sheets ($H_{a//}$), and a perpendicular disposition is formed between $H_f$ and $W$, $f_e$ can be calculated as

$$f_e = \gamma H_{a//} \left[ H_{a//}, H_f \perp W \right]$$

When $H_{a//}$ with a parallel configuration is formed between $H_f$ and $W$, $f_e$ is described by

$$f_e = \gamma \sqrt{H_{a//} + 2\pi a M_0} \left[ H_{a//}, H_f \parallel W \right]$$

Of note, Eqs. (1)-(4) can be applied only for thin films and/or sheet-shaped magnetic samples.

From the perspective of mathematics, we can expect the highest $f_e$ values for the case in Eq. (2) among all proposed equations.\textsuperscript{16} Therefore, magnetic thin films/sheets with the in-plane magnetic anisotropy, $H_{a//}$, forming the parallel disposition of $H_f$ and $W$ are ideal to achieve the high $f_e$ value. However, it is mostly impossible to artificially control the configuration between $H_f$ and $W$. In this study, we only focused on the BaCoTiFe$_{12}$O$_{19}$ powder/polymer composite sheets and BaCoTiFe$_{12}$O$_{19}$ thin films exhibiting $H_{a//}$ characteristics.

3. Experimental

3.1 Preparation of the BaCoTiFe$_{12}$O$_{19}$ powder/polymer composite sheet

First of all, a BaCoTiFe$_{12}$O$_{19}$ ceramic powder was synthesized by the solid-state reaction method using stoichiometric quantities of Fe$_3$O$_4$ (99.9 %), BaCO$_3$ (99.95 %), CoO (99.7 %), and TiO$_2$ (99.99 %). These starting materials were mixed and calcined in air at 750 °C for 12 h. The calcined powder was ground and pelletized with the diameter of 10 mm, applying a mechanical pressure of 10 MPa. Finally, the pelletized sample was annealed in air at 1200 °C for 12 h. The obtained BaCoTiFe$_{12}$O$_{19}$ powder was mixed with polymers to form the BaCoTiFe$_{12}$O$_{19}$ powder/polymer composite sheet.

After the final annealing, the BaCoTiFe$_{12}$O$_{19}$ ceramic powder was carefully ground to form a homogeneous size/shape distribution. Then, 30 g of the BaCoTiFe$_{12}$O$_{19}$ ceramic powder, 14.2 g of a polymer as a binder, 0.64 g of a curing agent, and 10 g of a diluted were mixed to obtain the BaCoTiFe$_{12}$O$_{19}$ powder/polymer composite sheet. During the preparation of the BaCoTiFe$_{12}$O$_{19}$ powder/polymer composite sheet, an external magnetic field was applied to the sheet sample using two permanent magnets facing the same poles of each other with a gap distance of 40 mm between the magnets. The permanent magnets were used to control the orientation of each BaCoTiFe$_{12}$O$_{19}$ particle embedded in a matrix of the sheet to lead $H_{a//}$.

3.2 Preparation of BaCoTiFe$_{12}$O$_{19}$ thin films by the metal–organic decomposition (MOD) method

We prepared BaCoTiFe$_{12}$O$_{19}$ thin films on Si (100) substrates (20 mm × 20 mm × 525 ± 25 µm) using the MOD method. Prior to the film preparation, the substrate was cleaned with isopropyl alcohol and then ultrapure water under sonication for 10 min for each process. A total of 120 µL of the MOD solution consisting of the mixture of Ba, Co, Ti, and Fe ions (a customized solution from Kojundo Chemical Laboratory Co., Ltd.) was deposited onto the substrate. Then, the MOD solution was spread using a spin coater (MS-B100, Mikasa Co., Ltd.) with the initial rotation speed of 250 rpm for 20 s, followed by 3000 rpm for 30 s, to form a liquid film on the substrate. The sample was calcined using a hot plate in air at 100 °C for 10 min. This was the first calcination. The second calcination was performed after the first calcination at 325 °C or 350 °C for 15 min. The formation of liquid film and the first/second calcinations were regarded as one sequence. We repeated the sequence 10 times to achieve the sufficient thickness of the films, which resulted in sufficient magnetic properties. Then, the sample was annealed in a furnace in air by varying the annealing temperature from 800 °C to 900 °C for 1 h. For annealing, a thin film sample was inserted into the furnace immediately after the temperature of the furnace reached at the aiming annealing temperature: the thin film sample was rapidly heated from room temperature to the desired annealing temperature. The calcination and annealing temperatures were based on the results of thermogravimetry (TG) and differential thermal analysis (TDA) as a function of temperature of the MOD solution.

3.3 Fundamental characterizations

Phase purity was evaluated by X-ray diffraction (XRD, SmartLab, Rigaku Corporation). The magnetic field-dependent magnetization behaviors were measured in the range of ±10 kOe at room temperature using a vibrating-sample magnetometer (VSM, C7–10A, Toei Industry Co., Ltd.). Using field-emission scanning electron microscopy (FE-SEM, JSM–7000F, JEOL Ltd.), the microscopic structures of the sample were observed.
3.4 Dynamic permeability
To measure the dynamic permeability in the GHz-frequency region, the short-circuited MSL\textsuperscript{12,14,15} method was employed in this study. For this method, the BaCoTiFe\textsubscript{10}O\textsubscript{19} powder/polymer composite sheet and BaCoTiFe\textsubscript{10}O\textsubscript{19} thin films were cut into rectangular pieces and placed at the short end of MSL. \(I_{rf}\) propagates along the MSL, such that \(H_{rf}\) is generated perpendicular to \(I_{rf}\). In this method, we regarded the overall experimental setup as an electric circuit model\textsuperscript{12,14,15}. An output signal from the measurements in this method is one of the scattering matrixes of an electric circuit, \(Y\), where \(1, 2, \ldots, n\) denotes the admittance, \(Y_{1}, Y_{2}, \ldots, Y_{n}\), of the sample using the following equation:

\[
Y_i = \frac{1}{j\omega C_0 L_0} + j\omega C_0, \\
Y_j = \frac{1}{j\omega L_0} + j\omega C_0.
\]

where \(\mu\) denotes effective permeability. Then, a strong static external magnetic field (\(H_{ext}\)) was applied parallel to the longitudinal direction of the rectangle-shaped sample. Consequently, the relationship of the direction between \(I_{rf}\) and \(H_{rf}\) is perpendicular. Under the application of \(H_{ext}\), the electric-circuit model of the experimental setup was changed; thus, \(Y_{1}\) was also changed to \(Y_{2}\):\textsuperscript{12,14,15}

\[
Y_2 = \frac{1}{j\omega L_0} + j\omega C_0.
\]

By comparing \(Y_1\) and \(Y_2\), we can determine \(\mu_e\). Finally, we obtained permeability, \(\mu\), of the sample using the following equation:

\[
\mu = \frac{\mu_e - 1}{\eta_m} + 1,
\]

where \(\eta_m\) denotes the coupling coefficient of the circuit. For the details of the method and calculation procedures, see Refs. \textsuperscript{12, 14, 15}.

4. Results and Discussion
4.1 BaCoTiFe\textsubscript{10}O\textsubscript{19} powder
Figure 1a presents the most representative XRD pattern of the ceramic BaCoTiFe\textsubscript{10}O\textsubscript{19} powder sample synthesized by the final annealing temperature of 1200°C. We confirmed essentially high-phase purity of polycrystalline-magnetoplumbite-structured BaCoTiFe\textsubscript{10}O\textsubscript{19} except some unknown peaks marked with asterisks.

The magnetic hysteresis loops of the corresponding sample to Fig. 1a is presented in Fig. 1b. In Fig. 1b, the magnetic behaviors of the synthesized BaFe\textsubscript{12}O\textsubscript{19} powder, which is the mother compound of BaCoTiFe\textsubscript{10}O\textsubscript{19} powder, are also plotted as a reference. The magnetic properties of the BaCoTiFe\textsubscript{10}O\textsubscript{19} powder exhibit typical soft magnetic behaviors with a low coercivity (\(H_c\)) value, \(\sim 186\) Oe. The saturation magnetization value (\(M_s\)) is approximately 59 emu/g, which is consistent with the values reported by another group.\textsuperscript{17} For BaFe\textsubscript{12}O\textsubscript{19}, the \(H_c\) value is much higher than BaCoTiFe\textsubscript{10}O\textsubscript{19}, which is approximately 2.3 kOe. Meanwhile, the magnetization value of BaFe\textsubscript{12}O\textsubscript{19} at 10kOe is \(\sim 53\) emu/g, though magnetization seems to be unsaturated. By substituting the Fe site with Co and Ti, we achieved low \(H_c\), resulting in a soft magnetic material.\textsuperscript{15} Soft magnetism is essential for magnetic materials utilized at high-frequency technologies. However, low \(M_s\) value is one of the drawbacks of magnetic materials for the application of high-frequency bands as absolute-permeability values tend to be deteriorated.

4.2 BaCoTiFe\textsubscript{10}O\textsubscript{19} powder/polymer composite sheet
The physical appearance of the prepared BaCoTiFe\textsubscript{10}O\textsubscript{19} powder/polymer composite sheet is presented in Fig. 2a. The average thickness of this sheet is 158 \(\mu\)m. The FE-SEM image of the sheet (Fig. 2b) indicates that fine particles of approximately less than 10 \(\mu\)m are embedded in the polymer matrix. Some BaCoTiFe\textsubscript{10}O\textsubscript{19} particles in the image seem to be hexagonally shaped, i.e., the ab-plane of the particles is observed in the image (Fig. 2b). This is attributed to the magnetoplumbite structure, i.e., the hexagonal structure of BaCoTiFe\textsubscript{10}O\textsubscript{19}.

The magnetic hysteresis loops of the BaCoTiFe\textsubscript{10}O\textsubscript{19} powder/polymer composite sheet were measured by applying an external magnetic field with in-plane (//) and out-of-plane (\(\perp\)) configurations to the surface of the sheet, respectively (Fig. 2c). The BaCoTiFe\textsubscript{10}O\textsubscript{19} ceramic powder exhibited a typical soft magnetic behavior (Fig. 1b), and the BaCoTiFe\textsubscript{10}O\textsubscript{19} powder/polymer composite sheet exhibited the same characteristics. The \(H_c\) values were 203 Oe for in-plane and 229 Oe for out-of-plane directions, respectively. Concerning this sheet, an apparent configuration dependence of magnetic hysteresis loops was observed, suggesting a magnetic anisotropy of the sheet. From hysteresis loops, the magnetic easy axis is inferred to lie in the in-plane direction of the sheet. The in-plane magnetic anisotropy of the sheet is what we tried to achieve.
Strong correlation between the structure, magnetic anisotropy, and high-frequency magnetic behaviors section). To control the orientation of BaCoTiFe\textsubscript{10}O\textsubscript{19} particles by permanent magnets is one of the reasons for the establishment of in-plane magnetic anisotropy (see “Preparation of the BaCoTiFe\textsubscript{10}O\textsubscript{19} powder/polymer composite sheet” section).

4.3 Dynamic permeability of the BaCoTiFe\textsubscript{10}O\textsubscript{19} powder/polymer composite sheet

The complex permeability of the BaCoTiFe\textsubscript{10}O\textsubscript{19}-powder/polymer composite sheet was measured using the short-circuited MSL method\textsuperscript{12,14,15} (Fig. 3). The real (\(\mu^\prime\)) and imaginary (\(\mu^\prime\prime\)) parts of permeability are plotted as a function of the electromagnetic wave frequency. Note that this figure is plotted with relative permeability values for the Y-axis. In the MSL measurements, the maximum frequency of electromagnetic wave irradiated on the sample was 30 GHz. With an increase in the frequency, a gradual decrease was observed in the \(\mu^\prime\) – \(f\) behavior, whereas a broadened peak was observed in the \(\mu^\prime\prime\) – \(f\) profile. \(f_r\) can be defined as the frequency at the maximum position of the \(\mu^\prime\prime\) – \(f\) curve, so that the \(f_r\) value was approximated at 4 GHz from Fig. 3. This suggests that the present sheet should be applicable under the high-frequency electromagnetic wave environments up to \(\sim 4\) GHz. At this frequency, the \(\mu^\prime\) and \(\mu^\prime\prime\) values of this sheet were 1.1 and \(2.2 \times 10^{-1}\), respectively. It should not be ignored that the \(\mu^\prime\) and \(\mu^\prime\prime\) values are low, which is caused by the drawback of low magnetization value, as discussed in Fig. 1b.

The absence of maximum in the \(\mu^\prime\) – \(f\) behavior and the broadened peak in the \(\mu^\prime\prime\) – \(f\) profile imply that the orientation of the BaCoTiFe\textsubscript{10}O\textsubscript{19} particles dispersed in the composite sheet would not be perfect. Furthermore, each BaCoTiFe\textsubscript{10}O\textsubscript{19} particle does not have a perfectly hexagonal shape (Fig. 2b), thus leading to a different magnetic characteristic from particle to particle in the BaCoTiFe\textsubscript{10}O\textsubscript{19} powder/polymer composite sheet. The size, shape, and homogeneity of sample constituents considerably affect dynamic permeability.\textsuperscript{16,18} Specifically, permeability deteriorates when the sample consists of inhomogeneous constituents. The absence of maximum in the \(\mu^\prime\) – \(f\) behavior at \(f_r\) and the broadened-\(\mu^\prime\prime\) – \(f\) profile are caused by these reasons.
4.4 BaCoTiFe_{10}O_{19} thin films

The XRD patterns of BaCoTiFe_{10}O_{19} thin films prepared using the MOD method are presented in Fig. 4a. For these films, the first calcination was performed at 100 °C for 10 min, followed by the second calcination at 325 °C for 15 min and annealing at various temperatures for 1 h. Note that, in the figure, the annealing temperatures are labeled. The formation of liquid film and the first/second calcinations were repeated 10 times for all films. The typical surface and cross-section images of BaCoTiFe_{10}O_{19} thin films are shown in (b).

![Figure 4](image)

**Figure 4.** (a) Phase purities of BaCoTiFe_{10}O_{19} thin films prepared through the MOD method annealed at various temperatures. For these films, the first calcination was performed at 100 °C for 10 min, followed by the second calcination at 325 °C for 15 min and annealing at various temperatures for 1 h. The annealing temperatures are labeled. The formation of liquid film and the first/second calcinations were repeated 10 times for all films. The typical surface and cross-section images of BaCoTiFe_{10}O_{19} thin films are shown in (b).

![Figure 5](image)

**Figure 5.** Magnetic characteristics of BaCoTiFe_{10}O_{19} thin films formed using the MOD method. (a) The hysteresis loops for the film annealed at 900 °C for 1 h. In-plane (\(//\)) hysteresis is indicated by the thick line, whereas out-of-plane (\(\perp\)) hysteresis is indicated by the thin line. The comparison of magnetic behaviors is presented in (b) for selected thin films annealed at 800 °C, 860 °C, and 900 °C for 1 h. (c) The most crucial magnetic parameters obtained from magnetic hysteresis loops, \(M_s\) and in-plane \(H_c\) values, as a function of annealing temperature.

4.4.1 BaCoTiFe_{10}O_{19} thin films

The XRD patterns of BaCoTiFe_{10}O_{19} thin films prepared using the MOD method are presented in Fig. 4a. For these thin films, second calcination was performed at 325 °C for 15 min. We performed a final heat treatment of thin films from 800 °C to 900 °C for 1 h, but we show some selected specimens in Fig. 4a. The peak intensities are lower than that of the BaCoTiFe_{10}O_{19} ceramic powder (Fig. 1a). However, essentially pure-phased polycrystalline-magnetoplumbite-structured BaCoTiFe_{10}O_{19} thin films were formed in the range of 860 °C to 900 °C. Note that XRD peaks marked with asterisks, which originated from a Si substrate of thin-film samples, are detected in the patterns. Figure 4b shows the most representative microstructures, the surface and cross section, of the thin film. Although the casting of the MOD solution and second calcinations were repeated 10 times, surprisingly, a highly dense film was formed. The adherence of the upper layers of the film seems to be sufficient; thus, homogeneous magnetic properties throughout the overall film layers are expected. The inset shows the fracture surface of the thin film, which consists of fine particles. The inside of the film also exhibits high density. The thickness of the film evaluated by SEM cross-sectional observations was ~480 nm on average.

![Figure 5a](image)

**Figure 5a** exhibits the magnetic hysteresis loops of the film annealed at 900 °C for 1 h. A clear in-plane magnetic anisotropy behavior was observed with the \(M_s\) value of ~302 emu/cm^3. For this sample, the squareness ratio defined by \(\frac{M_r}{M_s}\) (\(M_r\) represents remanence magnetization) is \(4.5 \times 10^{-1}\) for in-plane, whereas it is \(2.2 \times 10^{-1}\) for out of plane. On the basis of the squareness ratio, it is confirmed that this film also exhibits in-plane magnetic anisotropy. We emphasize that we achieved magnetic anisotropy without forming any underlayers of BaCoTiFe_{10}O_{19} thin films for a control of the orientation of BaCoTiFe_{10}O_{19} particles. The \(M_s\) value for the mother compound, BaFe_{12}O_{19} thin film, was ~350 emu/cm^3. A reduction in the \(M_s\) value for the present thin film is due to the Co and Ti substitutions at Fe sites. Moreover, the reported \(M_s\) value of SrCoTiFe_{10}O_{19} film, with a composition similar to that of the obtained BaCoTiFe_{10}O_{19} thin film, was ~350 emu/cm^3. The higher \(M_s\) value for SrCoTiFe_{10}O_{19} than that of the present thin film is caused by the presence of Sr instead of Ba; thus, the obtained \(M_s\) value in our thin film would be reasonable. A slight increase in \(H_c\) is observed in Fig. 5a, i.e., 342 Oe for in-plane and 445 Oe for out-of-plane directions. These values are larger than those of the BaCoTiFe_{10}O_{19} ceramic powder (Fig. 1b) and BaCoTiFe_{10}O_{19}-powder/polymer composite sheet (Fig. 2c). An increase in the...
coercivity is due to the size reduction of each BaCoTiFe$_{10}$O$_{19}$ particle, as observed in Fig. 4b.

The magnetic properties of thin films are sensitive to the annealing temperature. Figure 5b shows the in-plane magnetic hysteresis curves of the samples annealed at 800 °C, 860 °C, and 900 °C. There is a tendency of reduction in magnetic anisotropy with a decrease in the annealing temperature. Specifically, magnetically isotropic behaviors were concluded for thin films annealed at lower temperatures. The films annealed in the range of 800 °C–840 °C exhibited similar magnetic hysteresis curves between in-plane and out-of-plane configurations. An apparent in-plane magnetic anisotropy was observed for the films annealed at 860 °C–900 °C under our experimental conditions. As previously mentioned, we aimed to prepare thin films with in-plane magnetic anisotropy to achieve high $f_r$. Thus, we considered that an optimum annealing temperature was in the range of 860 °C–900 °C. Figure 5c presents changes in $M_I$ and in-plane $H_C$ values plotted against the annealing temperature. The films with high $M_I$ but low in-plane $H_C$ are available above the annealing temperature of 860 °C. For the thin film annealed at 900 °C, the highest $M_I$ but low $H_C$ values were obtained, which is the most identical magnetic characteristics for the use of high-frequency bands.

4.5 Dynamic permeability of BaCoTiFe$_{10}$O$_{19}$ thin films

Similar to the case of the BaCoTiFe$_{10}$O$_{19}$ powder/polymer composite sheet, we evaluated the dynamic permeability of the BaCoTiFe$_{10}$O$_{19}$ thin films. For the measurements, we cut thin films into rectangular pieces with the size of 2 mm $\times$ 10 mm $\times$ $d$ (various thicknesses). For the thin films, the FMR signal must be quite weak owing to the small volume fraction of magnetization. In addition, the specific characterization of small magnetization values for magneto-plumbite-structured ferrites generates a weak FMR signal.

By considering these difficulties, we thinned Si substrates to attain $d = 500$, which is the original thickness of the substrate, 400, 150, and 100 µm, respectively, to subtract the influence of the substrate from the FMR signal. Figures 6a–6d present the results of the dynamic permeability of BaCoTiFe$_{10}$O$_{19}$ thin films from the short-circuited MSL method. In Fig. 6a, a sharp maximum can be observed in the vicinity of 28 GHz in the $\mu'' - f$ profile. The peak maximum at mostly the same frequency is also observed in the $\mu'' - f$ behavior in Fig. 6b. However, the corresponding peak disappears in Figs. 6c ($d = 150$ µm) and 6d ($d = 100$ µm). Taking these results into account, the sharp maximum in the $\mu'' - f$ curves should not be the intrinsic FMR peaks, but generated by the resonance of Si substrates. Even when the thickness of the Si substrate was thinned to 100 µm, we did not observe the FMR peak in the $\mu'' - f$ and $\mu'' - f$ profiles.

This result was obtained possibly owing to the low signal to noise ratio of the measurement from the BaCoTiFe$_{10}$O$_{19}$ thin films. To overcome this issue, a possible solution is to increase the volume fraction of magnetization of the BaCoTiFe$_{10}$O$_{19}$ thin films. An increase in film thickness could be one of the solutions.

5. Conclusions

In conclusion, we evaluated the dynamic permeability of the BaCoTiFe$_{10}$O$_{19}$ powder/polymer composite sheet and BaCoTiFe$_{10}$O$_{19}$ thin films using the short-circuited MSL method. We developed the design of the materials; substitutes, composition of the material, control of magnetoplumbite anisotropy through the control of the particle orientations, and sample-preparation procedures are our original.

For the BaCoTiFe$_{10}$O$_{19}$ powder/polymer composite sheet, we successfully attained in-plane magnetic anisotropy with superior soft magnetic properties by applying external magnetic fields using permanent magnets during the sheet preparation. The $\mu'' - f$ and $\mu'' - f$ profiles for the sheet were found to be broad; however, $f_r$ was determined to be approximately 4 GHz. By considering these results, the “anisotropic suppression” or “anisotropic absorption” of electromagnetic waves with ~4 GHz is expected for this sheet.

Thus far, the MOD method has not been widely studied; it can be employed to successfully prepare magnetoplumbite-structured ferrites. To our knowledge, the BaFe$_{12}$O$_{19}$ thin films, which are the mother compounds of the BaCoTiFe$_{10}$O$_{19}$ thin films, are the only previous example prepared using the MOD method. In the present study, we successfully formed single-phase BaCoTiFe$_{10}$O$_{19}$ thin films for the first time using the MOD method by annealing the film above 860 °C for 1 h without any thin-film underlayers. The MOD method allows us to prepare magnetoplumbite-structured ferrite films at ambient-air environments without using vacuum systems. In this method, short- and low-heat treatments are effective for film preparation. The film exhibited soft magnetism with in-plane magnetic anisotropy. Unfortunately, the dynamic permeability behaviors of thin films were not available due to low magnetization signals. Further study on the dynamic permeability of BaCoTiFe$_{10}$O$_{19}$ thin films is required; however, BaCoTiFe$_{10}$O$_{19}$ thin films could be promising as anisotropic-electromagnetic wave suppression materials.

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Figure 6. Electromagnetic wave frequency dependence of permeability for BaCoTiFe$_{10}$O$_{19}$ thin films evaluated by the short-circuited MSL method. The thickness of Si substrate ($d$) was processed. $d$ = (a) 500, (b) 400, (c) 150, and (d) 100 µm. All measured samples were prepared from the same sample batch.
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