Significantly Improved Absorption Properties at Microwave Bands for Multi-layer Hexaferrite Thick Film Composites

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Abstract— Z-type hexaferrite, $Ba_3Co_2Fe_{24}O_{41}$, films without any substrate and the multi-layer hexaferrite-film (MLHF) composites have been fabricated using an modified infiltration technology. The microstructure, static and high-frequency magnetic properties and electromagnetic attenuation characteristics have been studied. The hexaferrite films without any substrate can reach a high volume concentration of $p = 0.45$ and has a certain degree of flexibility to be suitable for application. More importantly, the microwave magnetic properties are greatly improved; $\mu'_0 = 6$ and $\mu''_{\max} = 4.4$ increase by 67% and 175%, respectively, as compared to general particle-filler composites. Due to the significantly enhanced permeability and almost the same low permittivity, the absorption properties are greatly improved: return loss $RL \leq -10$ dB from 2.1 to 14.6 GHz, bandwidth $W_P = 150\%$ and thickness $t = 0.3$ cm. The bandwidth achieves 75% of theoretically maximum value.

1. INTRODUCTION

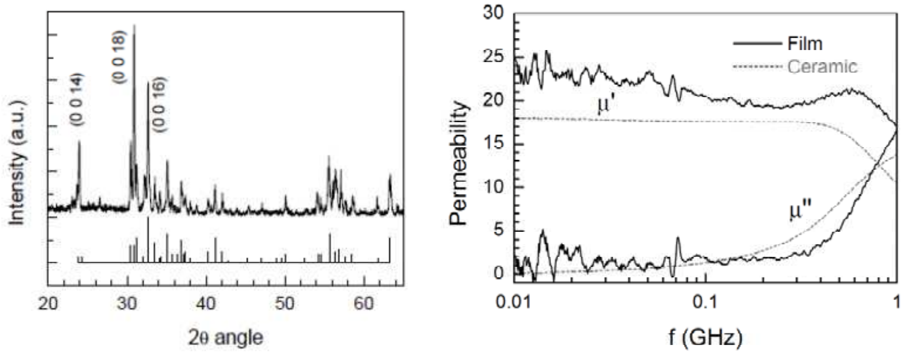
With the developments of electronic, telecommunication and radar technologies, the absorption and shielding of electromagnetic (EM) radiation has attracted much attention. When EM waves are irradiated into magnetic materials, a natural resonance (ferromagnetic resonance without an applied magnetic field) occurs, thus achieving the attenuation of EM waves. The EM materials generally consist of magnetic fillers and silicone, known as the EM composites. The fillers play an important role in determining the absorption properties of composites. However, for usual composites filled with metallic or ferrite particles, the permeability is rather small; the initial permeability μ'_0 is about 3–4 for composites with volume concentration p between 0.35 and 0.5 [1–4].

One way to enhance permeability is used hexaferrite flakes or films, instead of particles, as fillers in composites. In general, ferrite films were prepared using PLD (pulsed laser of deposition), spin coating, spin spray platting or tape casting methods [5]. For most of the general methods, the substrate is inevitably used in preparation of films. However, the substrates have the volume concentration of films in composites very small, thus limiting the increase of permeability. If films as fillers in composites for EM applications, one of preconditions is to remove the substrates from samples. In this work, a modified infiltration method has been developed to fabricate hexaferrite films without any substrate and, using the films, the multi-layer hexaferrite film (MLHF) composites have been prepared. Their high-frequency magnetic properties and microwave absorption performance are significantly improved.

2. EXPERIMENTS

The Z-type hexaferrite, $Ba_3Co_2Fe_{24}O_{41}$, membranes was fabricated using a modified infiltration technology [6]. The 0.4 g hexaferrite powders, fabricated using general ceramic method, were dispersed in distilled water. After that, the water was filtered out from the suspension through membrane filter paper. The suitable amount of PVA (polyvinyl alcohol) solution was dispensed uniformly on top of the hexaferrite powders. Using a pump, the PVA solution was infiltrated into the voids between the powders. After the PVA solution was dried, the membranes with the diameter of 3.5 cm, consisting of hexaferrite particles and filter paper, were obtained. Follow, the filter paper was torn from the membrane to obtain the pure hexaferrite membranes. The membranes was cut into rings followed by sintering at 1250°C for 4 h. Thickness of the rings are estimated to be 50–80 μm using a screw micrometer and also from the measured mass and volume of the films. Finally, fluid-like silicone was coated on each rings followed by pressing them together in a metal mould and thus preparing multi-layer hexaferrite film (MLHF) composites. Using the method, two MLHF composites are fabricated with almost the same volume concentration of about 45% and different dimensions of hexaferrite film rings. One is the outer and inner diameters of 1.42 and 0.62 cm and the other is 0.7 and 0.3 cm, respectively.

XRD was performed on the hexaferrite film using a ULTIMA IV diffractometer with Cu K_α radiation. Magnetization curves $M(H)$ and M-H loops were measured with applied fields of 0–20 kOe, and between −20 to +20 kOe, respectively, at room temperature using an EV9 VSM.



(a) (b)

Figure 1: (a) XRD patterns and (b) complex permeability spectrum for Z-type hexaferrite films.

Complex permeability and permittivity were measured from 0.1 to 9 GHz and from 5 to 18 GHz using Agilent VNA (Vector Network Analyzer) HP5230A with 14 mm and 7 mm coaxial line for the two MLHF composites. Also, the pure hexaferrite films were measured from 0.01 to 1 GHz using Agilent E4991A RF impedance/materials analyzer with open-short-load calibration. The measured fixtures are 16451A.

3. RESULTS AND DISCUSSION

3.1. Structure and Magnetic Properties of Hexaferrite Films

XRD pattern confirms that the film is single pase with the Z-type hexaferrite structure, as shown in Figure 1(a). All XRD lines are identical with the standard positions, based on JCPDS (Joint Committee on Powder Diffraction Standards) International Center for Diffraction Data (#19-0097), as shown in the bottom of the figure. Also, it is noted that, the intensity of XRD lines is significantly enhanced for the crystallographic planes of (0 0 14) at 23.6° and (0 0 18) at 30.8°, as compared to the standard intensity. This implies that considerable number of grains has preferential orientations of c-axis [0 0 l] to surface of the film.

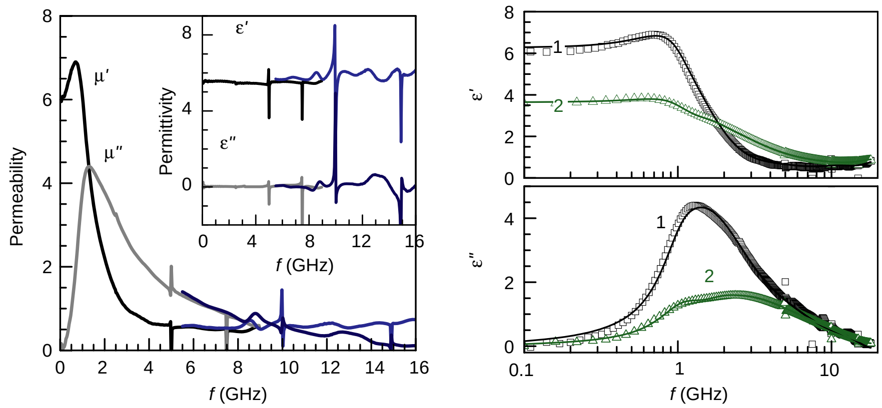
From the measured magnetization curves and M-H loops, the specific saturation magnetization M_s is 48.5 and 53.5 emu/g and the coercivity H_c is 9.1 and 3.1 Oe, respectively, for the Z-type hexaferrite powder sample and corresponding hexaferrite films. As compared to the former, M_s of the film increases by 10%, which has its origin in relatively lower density for the films. From the value of M_s, the density of the film approaches to 90% of the ceramic sample.

Figure 1(b) shows the complex permeability of the hexaferrite film. For comparison, permeability of the corresponding ceramic hexaferrite is also plotted in the figure. Initial (or static) permeability, μ'_0, defined as the value at 10 MHz, is 23 and the maximum imaginary μ'' at 1 GHz is 17 for the hexaferrite films. As compared to 18 and 14 for the corresponding ceramic hexaferrite, μ'_0 increases by 20% and μ'' by 30%. It is known that the Z-type hexaferrites are easy c-plane anisotropy and thus the magnetic moments are located in the c-plane of grains. XRD also indicates that most of grains tend to an c-plane arrangement in film plane. The magnetic moments are preferentially located in the plane, instead of random arrangements in the whole space like ceramic sample. Consequently, along the ac magnetic field direction, the averaged magnetization $\langle M \rangle$ is larger for films than for ceramic samples, due to two dimension distribution of magnetic moments for the former and three dimension distribution for the latter. According to $\mu = (\langle M \rangle / H)_{H \to 0} + 1$, larger $\langle M \rangle$ leads to larger μ'_0.

3.2. High-frequency Magnetic Properties of Multi-layer Hexaferrite-film Composite

Figure 2(a) shows the measured complex permeability and permittivity of MLHF composite at frequency f from 0.1 to 16 GHz. $\epsilon'(f)$ and $\epsilon'(f)$ are almost independent of frequency, as shown in the insert. The static real permeability μ'_0 is 6.0, the maximum imaginary μ'' is 4.4 and the corresponding resonance frequency f_R is 1.2 GHz.

μ'_0 and $\mu''_{\max}$ are only 3.6 and 1.6, respectively, for composites filled with general particle-filler, plotted together in Figure 2(b). As compared to the composite, μ'_0 increases by 67 and 175%,



(a) (b)

Figure 2: (a) Complex permeability and (inset) permittivity for MLHF composite and (b) real and imaginary permeability, where the open symbols and solid lines are the measured and curve-fitted results, respectively, 1 and 2 denote the MLHF and particle-filler composites, respectively.

respectively, for the MLHF composites. According to the Maxwell-Garnett law [7],

$$\mu_0' = \frac{(\mu_b - 1)\, p}{(\mu_b - 1)\,(1-p)N_\mathrm{d} + 1} + 1 \tag{1}$$

not only the value of μ_0' for composites is determined by μ_b, the initial permeability of fillers, but also the effective demagnetizing factor N_d. N_d is associated with the shape of fillers and the distribution of fillers in composites. The particle fillers can be approximately considered to be spherical in shape, thus $N_\mathrm{d}{\sim}0.3$. From $\mu_b = 18$ and Eq. (1), μ_0' is estimated as 3.4, which is very close to the experimental value of 3.6. On the other hand, for MLHF composites, if effective N_d is assumed to be 0.05, the calculated μ_0' is 6.3, which is well consistent with the measured value. Therefore, the significantly enhanced μ_0' is attributed to the decreased N_d for the MLHF composite.

3.3. Absorption Properties

The EM reflection property of material is typically characterized in terms of the power reflection of a plane wave reflected from an infinite slab of the material that is backed by a metallic plate. The reflectivity or return loss of composites, generally produced for normal incidence, is expressed in decibels RL (dB) [8]

$$RL\,(\mathrm{dB}) = 20\ \log_{10}\left|\frac{Z_{in} - Z_0}{Z_{in} + Z_0}\right| \tag{2}$$

and

$$Z_{in}/Z_0 = \sqrt{\frac{\mu}{\epsilon}}\tanh\left[j\frac{2\pi f t}{c}\sqrt{\mu\epsilon}\right] \tag{3}$$

where Z_{in} is the impedance of the composites backed by a ground plane, Z_0 is the intrinsic impedance of free space, c is the velocity of light in free space, t is the thickness of composites, f is the frequency of the incident EM wave, $\mu = \mu' - j\mu''$ is the complex permeability and $\epsilon = \epsilon' - j\epsilon''$ is the complex permittivity. Here, the used permeability and permittivity are the curve-fitted and linear-fitted data, respectively, in order to remove the fluctuations of calculated RL, due to measurement errors, especially at 5, 10 and 15 GHz. The sharp peaks of permittivity and permeability at these frequencies are not the intrinsic properties of materials, and instead have the origin in the measurement and calculation methods, namely Nicolson-Ross-Weir algorithm [9]. The fitted permeability spectra are shown in the solid lines in Figure 2(b), where the symbols are the measured data for MLHF and particle-filler composites.

Based on (2) and (3), the return loss RL, as a function of frequency f, are calculated at different thicknesses (t = 0.2–0.8 cm). Some representative results are shown in Figure 3(a) for the MLHF composites. From the figures, some important absorption parameters: return loss RL, the corresponding percentage bandwidth W_P and thickness t, can be achieved. The percentage bandwidth is defined as $W_P = (f_\mathrm{up} - f_\mathrm{low})/f_0$, where f_up and f_low are the frequency upper- and lower-limits of the frequency bands, respectively, and f_0 is the center frequency of the bands. For

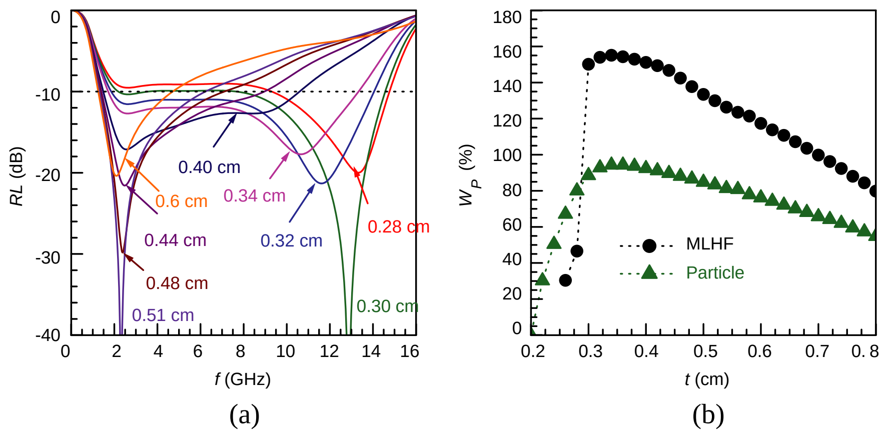


Figure 3: (a) Calculated absorption curves RL-f with different thickness t and (b) the dependence of bandwidth W_P on t, where 1 and 2 denote the MLHF and particle-filler composites, respectively.

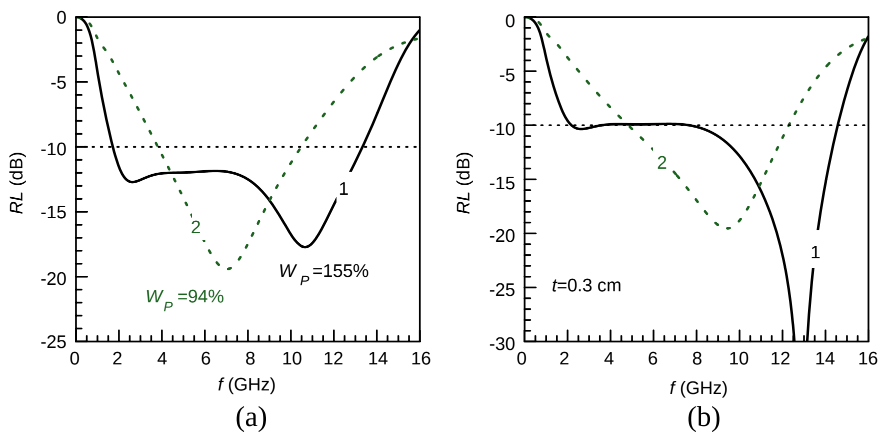


Figure 4: The absorption curves RL-f, (a) with maximum bandwidth W_P and (b) at the same thickness t=0.3 cm, where 1 and 2 denote the MLHF and particle-filler composites, respectively.

example, at thickness $t = 0.3$ cm, the frequency bands are from 2.07 to 14.5 GHz for $RL \leq -10$ dB and the corresponding W_P is 150% from 14.5/2.07 for the MLHF-filler composite. Using the method, the dependence of W_P on the thickness t can be obtained, as shown in Figure 3(b) for the MLHF and particle-filler composites.

From Figure 3(b), at the thickness range of 0.3–0.8 cm, W_P is significantly expanded for the MLHF-filler composite, as compared to the particle-filler composites. Figure 4(a) shows the absorption curves RL-f for the maximum bandwidth W_P for the two composites. The maximum W_P is observed at $t = 0.34$ cm. At the thickness, the absorption bands are from 1.7 to 13.6 GHz for $RL \leq -10$ dB and the maximum W_P achieves to 155% for the MLHF composite, while, for the particle-filler composites, the bands cover over 3.8–10.5 GHz and the corresponding W_P is only 95%. Further, at smaller thickness, such as $t = 0.3$ cm, the MLHF composite can also reach a broad absorption from 2.1 to 14.6 GHz with $W_P = 150\%$. However, at the thickness, only $W_P = 88\%$ is obtained from 4.8 to 12.3 GHz for the particle-filler composite. It is noted that the theoretically maximum percentage bandwidth is 200% for EM materials [10]. Now, for the MLHF composite with $p = 0.45$, the bandwidth W_P achieves above 75% of the theoretical value and thickness is only 0.3–0.34 cm. Therefore, the MLHF-filler composite is powerful and potential candidate for EM composites with low return loss, broad bandwidth and small thickness in S, C and X microwave bands.

4. CONCLUSION

Z-type hexaferrite, $Ba_3Co_2Fe_{24}O_{41}$, hexafilms without any substrate and the corresponding MLHF composites have been fabricated using an modified infiltration technology. The hexaferrite films without any substrate can reach a high volume concentration of $p = 0.45$ and has a certain degree of flexibility to be suitable for application. More importantly, the microwave magnetic properties are greatly improved; $\mu'_0 = 6.0$ and $\mu''_{\max} = 4.4$ increase by 67% and 175%, respectively, as compared to general particle-filler composites. Due to the significantly enhanced permeability and almost the same low permittivity, the absorption properties are greatly improved: return loss $RL \sim -10\,\text{dB}$ from 2.1 to 14.6 GHz, bandwidth $W_P = 150\%$ and thickness $t = 0.3\,\text{cm}$. The bandwidth achieves 75% of theoretically maximum percentage bandwidth.

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