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Advanced Structural and Functional Materials for Protection

High-frequency Properties and Electromagnetic Wave Attenuation for Hexaferrite Composites

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Abstract

Static, high-frequency magnetic properties and EM attenuation characteristics have been studied for Ti substituted Z-type hexaferrites, $\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ with $x=0-0.6$, and ferrite/silicone composites. The results show that Ti substitution is able to enhance μ' and μ'' and improve EM attenuation with expanded relative bandwidth W_R and decreased thickness of composites. The $\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ composite with $x=0.3$ has best EM attenuation properties with low reflectivity and broad bandwidth at C and X bands (4-12 GHz). W_R of 3.7 for $R < -10$ dB and thickness of only 0.30 cm are achieved.

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Keywords: EM attenuation, High-frequency properties, Ti substituted Z-type hexaferrites

1. Introduction

With the developments of electronic, telecommunication and radar technologies, electromagnetic interference (EMI) at microwave is of growing importance. The attenuation or shielding of electromagnetic radiation has attracted much attention [1-3]. To prevent from or reduce EMI, electromagnetic (EM) attenuation materials are often used. When an EM wave is irradiated into magnetic materials, magnetic resonance occurs and EM energy is greatly attenuated. To achieve broad bandwidth with low reflectivity, wave impedance $(\mu/\epsilon)^{1/2}$ should be close to unity for the impedance matching between materials and free space, and the imaginary permeability μ'' should be large enough to attenuate the incident EM wave inside the materials.

The magnetic particles (fillers) for use as EM attenuation composites are usually divided into two types, namely ferrite particles and metallic particles. As compared to metallic particles, ferrite particles have many important characteristics, such as low permittivity, high resonance frequency, high resistivity, low density and good chemical stability.

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In general, there are three kinds of ferrites: spinel, garnet and hexaferrite (or barium ferrites). According to their anisotropy, the hexaferrites are classified two types, namely c-axis and c-plane anisotropy. The correlation between high-frequency (permeability μ and resonance frequency f_R) and static magnetic properties ($4\pi M_s$ and anisotropy fields) is expressed as [4]

$$\mu'_0 = \frac{8\pi M_s}{3H_a} \quad \text{and} \quad f_R = \frac{\gamma}{2\pi} H_a \quad (1)$$

$$\mu'_0 = \frac{2\pi M_s}{H_\phi} \quad \text{and} \quad f_R = \frac{\gamma}{2\pi} \sqrt{H_\theta \cdot H_\phi} \quad (2)$$

for hexaferrites with c-axis and c-plane anisotropy, respectively, where $\gamma/2\pi$ is the gyromagnetic factor, $4\pi M_s$ is the saturation magnetization, H_a is the anisotropy field for c-axis anisotropy, H_θ and H_ϕ are the out-of-plane and in-plane anisotropic fields for c-plane anisotropy, respectively. For c-axis anisotropy, H_a is very large, about tens kOe. According Eq. (1), although large H_a leads to high resonance, it gives rise to small permeability μ . On the other hand, hexaferrites with c-plane anisotropy are soft magnetic materials due to small H_ϕ . According to equation (2), small H_ϕ leads to large μ' , and f_R can be modified by H_θ . Therefore, we can simultaneously obtain both large μ and appropriate f_R . These are great advantages for hexaferrites with c-plane anisotropy.

Most hexaferrites are hard magnetic materials. However, Co2Z hexaferrite, $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$, is soft magnetic material due to its c-plane anisotropy. In this work, Ti substituted Co_2Z hexaferrites, $\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ with $x=0-0.6$ and their composites were prepared, and the effect of Ti substitution on magnetic and EM attenuation properties was studied.

2. Experiments

$\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ with $x=0, 0.15, 0.3, 0.45$ and 0.6 was synthesized using the conventional ceramic technique. The aligned samples were prepared by mixing the fine powders with silicone for studying the anisotropy properties. Composites are prepared by mixing $\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ powders and silicone. The composites are annular in shape with the outer diameters of 7 mm, inner diameters of 3 mm, and thicknesses of about 2 mm. The volume concentration of the powders is 50 %.

X-ray diffraction (XRD) was performed on the powder and aligned samples using a ULTIMA IV diffractometer with $\text{Cu K}\alpha$ 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 for all sintered samples using an EV9 VSM. The complex permittivity and permeability of the hexaferrite/silicone composites (or hexaferrite composites) from 0.1 to 18 GHz were measured using Agilent VNA (Vector Network Analyzer HP5230A) with a reflection-through-line calibration, based on the transmission/reflection method.

3. Results and discussion

3.1. Crystal structure and magnetic properties

XRD patterns identify that all $\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ with $x=0-0.6$ are single phase with Z-type hexagonal structure. The lattice parameters a and c are listed in Table 1. Saturation magnetization $4\pi M_s$ and intrinsic coercivity H_c are obtained from the measured magnetization curves and $M-H$ loops, respectively. With Ti substitution, $4\pi M_s$ first increases to the maximum at $x=0.3$ and then decreases. H_c can be considered as constant, about 7 Oe, up to $x=0.45$, and rapidly increases to 16 Oe at $x=0.6$. The out-of-plane anisotropy fields H_θ gradually increases from 12.5 to 17 kOe, as x varies from 0 to 0.6. The magnetic parameters are also listed in Table 1.

Complex permittivity, ϵ' and ϵ'' , as shown in Fig.1 (a), exhibits two important characteristics for $\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ composites with $x=0$ to 0.6. (1) ϵ' and ϵ'' are almost frequency independent for low Ti substitution of $x=0-0.3$. However, for high substitution of $x=0.45$ and 0.60, the frequency dependence of ϵ' and ϵ'' are observed. At $f > 4$ GHz, they are approximately considered as constant; however, at low frequency, they rapidly increase. For example, ϵ' is 7.1 at 4 GHz, while it increases to about 10 at 0.1 GHz, for the composites with $x=0$. (2) Both ϵ' and ϵ'' increase with Ti substitution in the whole frequency range. At low frequency, the increase is significant. For example, at 1 GHz, they increase from 6.4 to about 8.0, and from 0.2 and 1.1, respectively, as x varies from 0 to 0.6.

Table 1: Lattice parameters, a and c , saturation magnetization $4\pi M_s$, coercivity H_c and out-of-plane anisotropy H_0

x	0	0.15	0.3	0.45	0.6
a (nm)	0.5883(3)	0.5885(2)	0.5887(2)	0.5889(3)	0.5887(3)
c (nm)	5.251(3)	5.252(3)	5.256(3)	5.259(2)	5.264(4)
$4\pi M_s$ (kGs)	2.82	2.94	3.32	3.22	2.58
H_c (Oe)	7.2	5.6	7.6	7.3	16
H_0 (kOe)	12.5	14.1	15.3	16.2	17.0

3.2. High-frequency permittivity and permeability

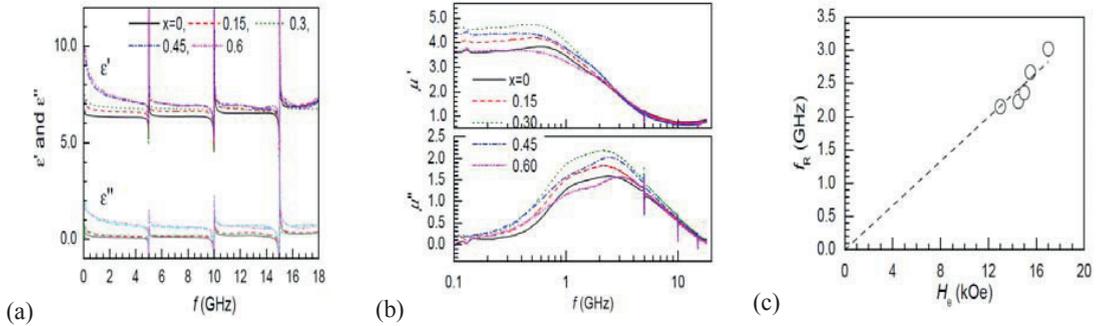


Figure 1: High-frequency properties: (a) complex permittivity ϵ' and ϵ'' , and (b) complex permeability, μ' and μ'' for composites with $x=0-0.6$, and (c) the proportional relationship between resonance frequency f_R and anisotropy field H_0

When a Ti^{4+} ion is substituted for an Fe^{3+} ion, the Fe ion will be transfer from trivalent to bivalent for the requirement of valence balance. The two characteristics of ϵ' and ϵ'' can be attributed to the formation of Fe^{2+} ions due to Ti^{4+} substitution. The presence of Fe^{2+} ions and the hopping between Fe^{2+} and Fe^{3+} ions can enhance the electronic polarizations in ferrite grains, thus increasing ϵ' [5]. Due to much higher resistivity of grain boundary than that of grains, more hopping electrons are piled up at the grain boundary at lower frequency, thus increasing polarization. Consequently, the permittivity rapidly increases with decreasing frequency. Meanwhile, a conductive loss due to the hopping is also one of the major origins in large dielectric losses ϵ'' [6].

Complex permeability, $\mu'(f)$ and $\mu''(f)$, are shown in Fig.1 (b) for $Ba_3Co_2Fe_{24-x}Ti_xO_{41}$ composites with $x=0$ to 0.6. The quasi-static permeability μ'_0 is defined as the averaged real permeability near 0.1 GHz, μ''_{max} is the maximum imaginary permeability in $\mu''(f)-f$ curves, and the resonance frequency f_R is defined as the corresponding frequency for μ''_{max} .

From Fig. 1 (b), two resonance peaks are found on the imaginary permeability spectra, $\mu''(f)$. The resonance at relatively low frequency of about 1 GHz is attributed to wall resonance and the resonance at relatively high frequency of 2-3 GHz to natural resonance [4]. The two resonance peaks are significantly distinguished with Ti substitution x . Now, in the two resonance frequencies, the natural resonance frequency f_R , i.e. the resonance at higher frequency, is considered. With Ti substitution x , f_R is shifted to higher frequency. If f_R is plotted, as a function of the out-of-plane anisotropy field H_0 , a good proportional relationship between f_R and H_0 is found, as shown in Fig.1 (c). The proportional relationship can be understood by [7]

$$f_R = \frac{\gamma}{2\pi} \sqrt{H_\theta \cdot H_\phi} = \frac{\gamma}{2\pi} \beta \cdot H_\theta \quad (3)$$

where β is the ratio of H_θ and H_ϕ (in-plane anisotropy field). Therefore, the increase in f_R can be attributed to the increase in H_θ with Ti substitution.

From Fig.1 (b), without Ti substitution, $\mu'_0=3.6$ and $\mu''_{\max}=1.6$. With Ti substitution x , μ'_0 and μ''_{\max} first increase to the maximum of 4.5 and 2.2 at $x=0.3$ and then decrease to 3.6 and 1.6 at $x=0.6$, respectively. The characteristic can be explained based on the results of curve-fitting to the permeability spectra for ceramic $\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ [8]. The permeability is comprised of gyromagnetic permeability from natural and wall permeability from wall resonance for Z-type hexaferrites. With Ti substitution x , for the former, μ'_0 gradually decreases due to increased H_0 , while, for the latter, μ'_0 increases up to $x=0.45$ and then decreases at $x=0.6$. Because of the competition between two permeability, the total permeability μ'_0 first increases and then decreases and has the maximum at $x=0.3$.

3.3. EM attenuation

The reflectivity (R) or return loss of EM wave in the composites is determined, for the case of a metal-backed single layer, by

$$R = 20 \log_{10} \left| \frac{Z_{in} - 1}{Z_{in} + 1} \right| \quad \text{and} \quad Z_{in} = \sqrt{\frac{\mu}{\varepsilon}} \tanh\left(j \frac{2\pi ft}{c} \sqrt{\mu \cdot \varepsilon}\right) \quad (4)$$

where Z_{in} is the normalized impedance of the composite with respect to the impedance in free space for a normally incident plane wave, c is the velocity of light, t is the thickness of composite, f is the frequency of the incident EM wave, $\mu = \mu' - j\mu''$ is the complex permeability and $\varepsilon = \varepsilon' - j\varepsilon''$ is the complex permittivity. The relative bandwidth is defined as $W = f_{up}/f_{low}$, where f_{up} and f_{low} are the upper- and lower-frequency limits, respectively, for $R < -10$ dB.

Based on Eq.(4), the dependence of reflectivity R on f can be calculated over the whole frequency range for a given thickness t , as shown in Fig. 2 (a) for $\text{Ba}_3\text{Co}_2\text{Fe}_{24-x}\text{Ti}_x\text{O}_{41}$ composite with $x=0.3$ as an example. From the figure, the relative bandwidth W_R for $R < -10$ dB can be obtained. For example, at $t=0.28$ cm, the bandwidth covers over 4-14 GHz and $W_R=3.5$ is achieved. Thus, the maximum W_R ($W_{R,\max}$) and the corresponding thickness t_0 can be obtained for each composite, as shown in Table 2. With Ti substitution, $W_{R,\max}$ increases from 2.9 to the maximum 3.7 at $x=0.3$ and then decreases to 3.1 at $x=0.6$ and t_0 decreases from 0.36 cm to the smallest 0.3 cm at $x=0.3$ and then increases to 3.2 cm at $x=0.45$ and 0.6. After Ti substitution, both bandwidth and thickness of composites are significantly improved.

Table 2: EM attenuation parameters, where f_{low} and f_{up} are the lower- and upper –limit of bandwidth, respectively, $W_{R,\max}$ is the maximum relative bandwidth and t_0 is the optimum thickness for the maximum W_R

x	0	0.15	0.3	0.45	0.6
f_{low} (GHz)	3.5	3.4	3.5	3.5	3.8
f_{up} (GHz)	10.1	10.2	12.9	12.1	11.7
$W_{R,\max}$	2.9	3.0	3.7	3.5	3.1
t_0 (cm)	0.36	0.34	0.30	0.32	0.32

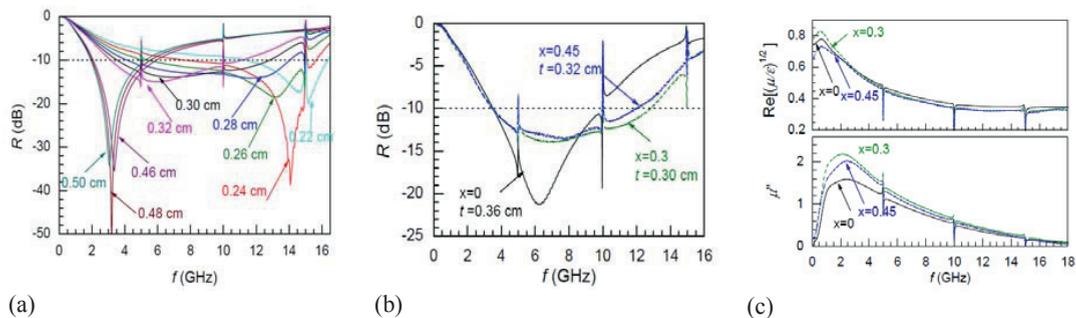


Figure 2: EM attenuation properties: (a) chart of R - f with various thicknesses t for the composite with $x=0.3$, (b) the maximum W_R with corresponding t , and (c) $\text{Re}[(\mu/\varepsilon)^{1/2}]$ and $\mu''(f)$ for composites with $x=0, 0.3$ and 0.45 .

Figure 2 (b) shows the chart of R against f at the optimum thickness t_o for composites with $x=0, 0.3$ and 0.45 . The lower-limit of bandwidth f_{low} is almost the same, about 3.5 GHz for all composites. However, the upper-limit f_{up} is shifted to higher frequency, 12.9 and 12.1 GHz, respectively, for $x=0.3$ and 0.45 , as compared to 10.1 GHz for composite without Ti substitution.

It is found that $Ba_3Co_2Fe_{24-x}Ti_xO_{41}$ composites with $x=0.3$ have the optimum attenuation with W_R of 3.7 at thickness of 0.3 cm. For comparison, at the same thickness, M -type hexaferrite composites have a frequency band from 5 to 10 GHz for $R < -10$ dB and the corresponding W_R of 2.0 [2], and W -type hexaferrite composites can achieve a frequency band from 3.5 to 13.8 GHz for $R < -10$ dB and W_R of 3.9 [9].

To achieve broad bandwidth with low reflectivity, $(\mu/\epsilon)^{1/2}$ should be close to unity for the impedance matching between materials and free space, and $\mu''(f)$ should be large enough to attenuate the EM wave inside materials. The values of $(\mu/\epsilon)^{1/2}$ and $\mu''(f)$ are plotted in the whole frequency range (0.1-16.5 GHz) for composites with $x=0, 0.3$ and 0.45 , as shown in Fig. 2 (c). In the frequency band of 3-16.5 GHz, $(\mu/\epsilon)^{1/2}$ is almost the same for all composites. However, as compared to the composite without Ti substitution, $\mu''(f)$ is significantly enhanced for composites with $x=0.3$ and 0.45 . As a result, after Ti substitution, attenuation properties of the composites are improved.

4. Conclusions

A small amount of Ti Substitution is able to improve the static and high-frequency magnetic properties of $Ba_3Co_2Fe_{24-x}Ti_xO_{41}$ and the corresponding composites. With Ti substitution, $4\pi M_s$, μ'_0 and μ''_{max} first increases to the maximum at $x=0.3$, and then decreases.

Due to enhanced $\mu''(f)$ and almost the same $(\mu/\epsilon)^{1/2}$, the $Ba_3Co_2Fe_{24-x}Ti_xO_{41}$ composite with $x=0.3$ has best EM attenuation properties with low reflectivity and broad bandwidth at C and X bands (4-12 GHz). W_R of 3.7 for $R < -10$ dB and thickness of only 0.30 cm are achieved.

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