

# Magnetic and microwave absorbing properties of Ti and Co substituted M-hexaferrites in Ka-band frequencies (26.5~40 GHz)

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**Abstract** Magnetic and microwave absorbing properties of M-hexaferrites have been investigated in Ka-band frequencies (26.5~40 GHz) with substitution of  $Ti^{+4}$  and  $Co^{+2}$  ions for  $Fe^{+3}$  sites. The powders of composition  $BaFe_{12-2x}Ti_xCo_xO_{19}$  were prepared by conventional ceramic processing, where the substitution ratio  $x$  was varied from 0.0 to 2.0. With the calcined powders, polycrystalline bulk and composite sheet samples were prepared for the evaluation of magnetic and microwave absorbing properties. VSM measurements indicate that the saturation magnetization and coercivity of the M-hexaferrites decrease with the substitution of Ti and Co, which is attributed to the reduction in crystal anisotropy field by changing of easy-axis of magnetization from c-axis to basal plane. Ferromagnetic resonance frequency with the peak magnetic loss is decreased sensitively with the substitution of Ti and Co, which is well consistent with ferromagnetic resonance theory. Theoretical estimation of matching frequency and matching thickness for zero wave reflection is made using the solution map of impedance matching. A wide bandwidth of microwave absorption is predicted in the composite samples containing the ferrite powders of  $x=0.5$  in Ka-band frequencies.

**Keywords** Microwave absorbers · M-hexaferrites · Ka-band frequency · Ti-Co substitution

## 1 Introduction

Recent advances in electronic and telecommunication technology requires the control of the EMI (Electro-

Magnetic Interference) problems in the high-frequency region of millimeter wave. Ka-band (26.5~40 GHz) is a lower region of those high-frequency spectrum and now is used for satellite broadcasting and wireless local area network. The spinel ferrites (Ni–Zn or Mn–Zn ferrites) are widely used as the noise absorbing materials at low frequency and their magnetic and dielectric properties in correlation with microwave absorbance have been extensively investigated for this purpose [1–3]. However, the use of spinel ferrites as the EMI suppressors should be limited below a frequency defined by Snoek's limit because of the cubic magnetocrystalline anisotropy [4]. To overcome this problem, the hexagonal ferrites with high magneto-crystalline anisotropy can be considered as the absorbent material in the high-frequency band of millimeter wave.

Barium hexaferrites are anisotropic iron oxides which can represent high values of permeability and thus one of the promising materials for microwave devices and absorbers in gigahertz frequency range. These are classified into many types depending on their chemical formulas and crystal structures: M-type ( $BaFe_{12}O_{19}$ ), W-type ( $BaMe_2Fe_{16}O_{27}$ ), X-type ( $Ba_2Me_2Fe_{16}O_{27}$ ), Y-type ( $Ba_2Me_2Fe_{12}O_{22}$ ), Z-type ( $Ba_3Me_2Fe_{24}O_{41}$ ), and U-type ( $Ba_4Me_2Fe_{36}O_{60}$ ) where Me is the transition metal element [4, 5]. Among these, Y- and Z-types with planar magnetic anisotropy in basal plane have been investigated widely for the high-frequency applications [6–8]. Microwave absorbing properties of  $Co_2U$  ( $Ba_4Co_2Fe_{36}O_{60}$ ) hexaferrites in composite sheet form has also been reported [9].

M-type hexaferrite is a well known material having a strong uniaxial anisotropy and, therefore, used as a starting material for permanent magnets. However, substituted M-hexaferrites, where  $Fe^{3+}$  cations are substituted by  $A^{4+}/B^{2+}$  pair while maintaining the average 3+ valance

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states, changes the magneto-crystalline anisotropy from the hexagonal c-axis to a-b plane. The most studied sample is the substitution of  $\text{Fe}^{3+}$  by a  $\text{Ti}^{4+}/\text{Co}^{2+}$  pair ( $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$ ), which presents the magnetic anisotropy change at  $x=1.1\sim 1.3$  [10, 11]. Similar types of substitution  $\text{Ir}^{4+}/\text{Co}^{2+}$  and  $\text{Ru}^{4+}/\text{Co}^{2+}$  bring about the planar magnetic anisotropy at a smaller substitution rate ( $x=0.6$  and  $x=0.4$ , respectively) [12, 13]. The substituted M-hexaferrites are of technological interest in microwave applications because ferromagnetic resonance frequency and thus the absorbing frequency band can be controlled by the substitution rates [14, 15]. However, there are no reports on microwave absorbing properties of the substituted M-hexaferrites in millimeter wave frequency range.

In this study, magnetic and microwave absorbing properties of M-hexaferrites have been investigated in Ka-band frequencies (26.5~40 GHz) with the substitution of  $\text{Ti}^{4+}$  and  $\text{Co}^{2+}$  into  $\text{Fe}^{3+}$  sites. The static magnetic properties (saturated magnetization, coercive force) are investigated for the indirect information about the magneto-crystalline anisotropy (uniaxial or planar). Complex permeability and permittivity are measured in Ka-band frequency range using a rectangular waveguide method. Theoretical prediction of microwave absorbing properties (matching frequency and bandwidth, matching thickness) is conducted from the measured complex permeability and permittivity.

## 2 Experimental procedures

The powders and sintered specimens of  $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$  composition were prepared by conventional ceramic processing technique, where the substitution ratio  $x$  was varied from 0 to 2.0. Powder mixtures were calcined for 2 h in air at the temperature of 1200°C. The powder X-ray diffraction patterns indicated the completely reacted M-type barium ferrite. With the calcined powders, composite sheet samples were prepared using silicone rubber as a matrix material. The mixing ratio of ferrite powder to silicone rubber was 4 by weight. The sintered specimens were also prepared at a slightly higher temperature of 1250°C for the study of static magnetic properties.

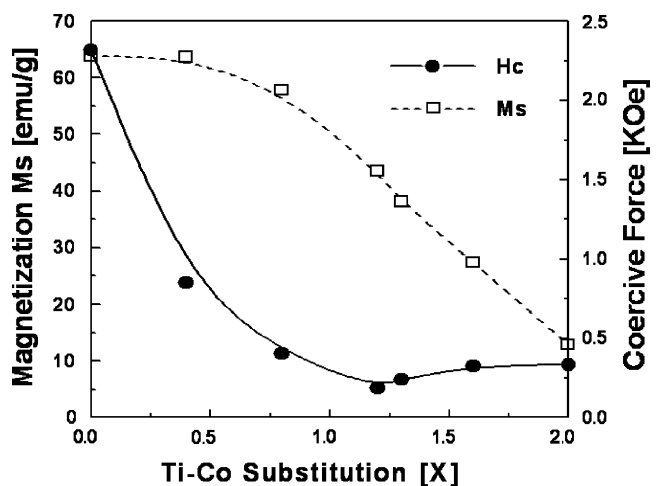
The saturated magnetization ( $M_s$ ) and the coercive force ( $H_c$ ) were measured in the sintered specimens by vibration sample magnetometer (VSM). The complex permeability and dielectric constant were determined from the measured reflected and transmitted scattering parameters ( $S_{11}$  and  $S_{21}$ , respectively) by using HP 8722D network analyzer and WR-28 rectangular waveguide whose dimension is height=3.51 mm and width=7.13 mm. Measurements were conducted in Ka-band frequencies (26.5~40 GHz).

## 3 Results and discussion

### A. Magnetic Properties

Figure 1 shows the variation of magnetic properties ( $M_s$ ,  $H_c$ ) with the substitution of  $\text{Ti}^{4+}$  and  $\text{Co}^{2+}$  for  $\text{Fe}^{3+}$  sublattices in sintered specimens of  $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$  composition. The saturated magnetization ( $M_s$ ) decreases nearly linearly with the substitution of Ti and Co.  $M_s$  value of pure barium ferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) was 65 emu/g, which is almost the same value reported earlier [11]. At the substitution ratio of  $x=2.0$ ,  $M_s$  is reduced to 12 emu/g. The result is attributed to the reduction in molecular magnetic moment by replacing the  $\text{Fe}^{3+}$  ions by nonmagnetic  $\text{Ti}^{4+}$  ions and magnetically weak  $\text{Co}^{2+}$  ions.

The reduction in coercive force ( $H_c$ ) with Ti and Co substitution is more apparent. The coercive force of pure barium ferrite is very high (about 2300 Oe) due to its strong uniaxial anisotropy along the c-axis of the magnetoplumbite structure. With the substitution of Ti and Co, the coercive force is decreased rapidly to 185 Oe at  $x=1.2$ . The result is interpreted to be due to the reduction in crystal anisotropy field by changing of easy-axis of magnetization from c-axis to basal plane. It was reported that magnetic anisotropy becomes planar on basal plane at the substitution rate of  $x=1.1\sim 1.3$  in the Ti–Co substituted M-hexaferrites [10, 11]. Further addition of Ti and Co above 1.2 gives rise to a slight increase in coercive force ( $H_c=335$  Oe at  $x=2.0$ ), which is due to the increase in in-plane anisotropy [8]. The similar result of planar anisotropy was observed in the Ir–Co or Ru–Co substituted M-hexaferrites [12, 13].



**Fig. 1** The variation of saturated magnetization ( $M_s$ ) and coercive force ( $H_c$ ) with the substitution of  $\text{Ti}^{4+}$  and  $\text{Co}^{2+}$  into  $\text{Fe}^{3+}$  sites in M-type hexaferrites

### B. High Frequency Properties (Complex Permeability and Permittivity)

Figure 2 shows the frequency dispersion of complex permeability ( $\mu_r = \mu_r' - j\mu_r''$ ) and permittivity ( $\epsilon_r = \epsilon_r' - j\epsilon_r''$ ) determined in the composite sheet samples containing the  $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$  powders as absorbent fillers. For the specimen of pure M-hexaferrite ( $x=0$ ), the real part of permeability ( $\mu_r'$ ) is approximately 1.1 and imaginary part ( $\mu_r''$ ) is negligibly small as shown in Fig. 2(a). Because of strong uniaxial anisotropy (large coercive force), ferromagnetic resonance is not observed in the Ka-band frequencies. It is believed that the resonance occurs at a higher frequency above 40 GHz.

In the specimens with lower coercive force, ferromagnetic resonance is observed in the frequency region of Ka-band as shown in Fig. 2(b) and Fig. 2(c). The resonance frequency was determined to be 37.5 GHz and 29.5 GHz for the specimen of  $x=0.3$  and  $x=0.5$ , respectively. With weakening of uniaxial anisotropy, the resonance frequency moves to a lower frequency region. For the specimen of highly substituted with Ti and Co ( $x=0.8$ ), no magnetic resonance is observed in Ka-band frequency as shown in Fig. 2(d). It is believed that the ferromagnetic resonance

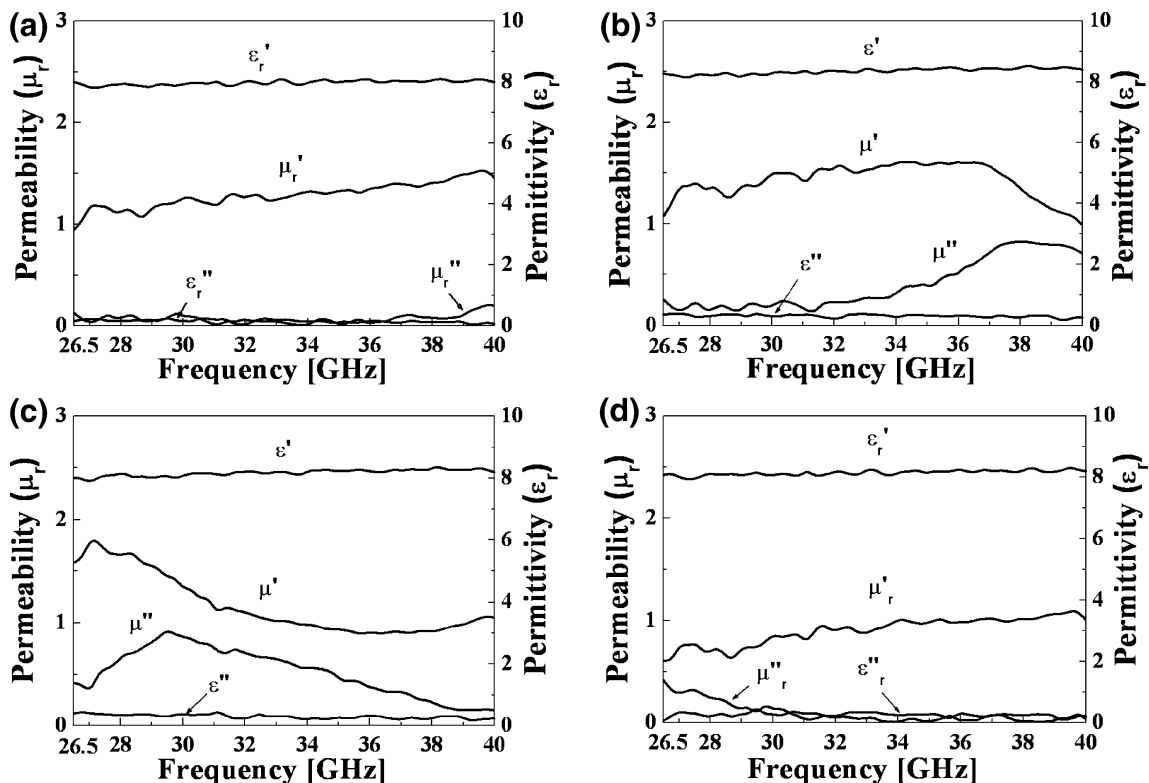
occurs at a lower frequency below 26.5 GHz due to its small anisotropy field.

According to the ferromagnetic resonance theory [16], the resonance frequency ( $f_r$ ) in hexagonal crystal is closely related with the magnetic properties which is given by (1)

$$f_r(\mu_i - 1) = \frac{1}{3\pi\mu_0} \gamma M_s \left( \frac{1}{2} \sqrt{\frac{H_{A1}}{H_{A2}}} + \frac{1}{2} \sqrt{\frac{H_{A2}}{H_{A1}}} \right) \quad (1)$$

where  $\mu_i$  is initial permeability,  $\mu_0$  is permeability of vacuum and  $\gamma$  is gyromagnetic constant.  $H_{A1}$  is the anisotropy field for the magnetization rotation in the easy plane and  $H_{A2}$  is that for rotation out of the plane. In random polycrystals, it is difficult to determine the  $H_{A1}$  and  $H_{A2}$ . If the coercivity can be taken as a degree of the anisotropy field, the resonance frequency is considered to be proportional to the saturated magnetization and the coercivity. The present result is quite well consistent with the ferromagnetic resonance theory.

The real part of permittivity ( $\epsilon_r'$ ) of the composite specimens is about 8 and irrespective of the substitution ratio of Ti and Co. The dielectric loss ( $\epsilon_r''$ ) is negligibly small for all the specimens.



**Fig. 2** Complex permeability and permittivity of composite sheet samples containing  $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$  powders: (a)  $x=0$ , (b)  $x=0.3$ , (c)  $x=0.5$ , (d)  $x=0.8$

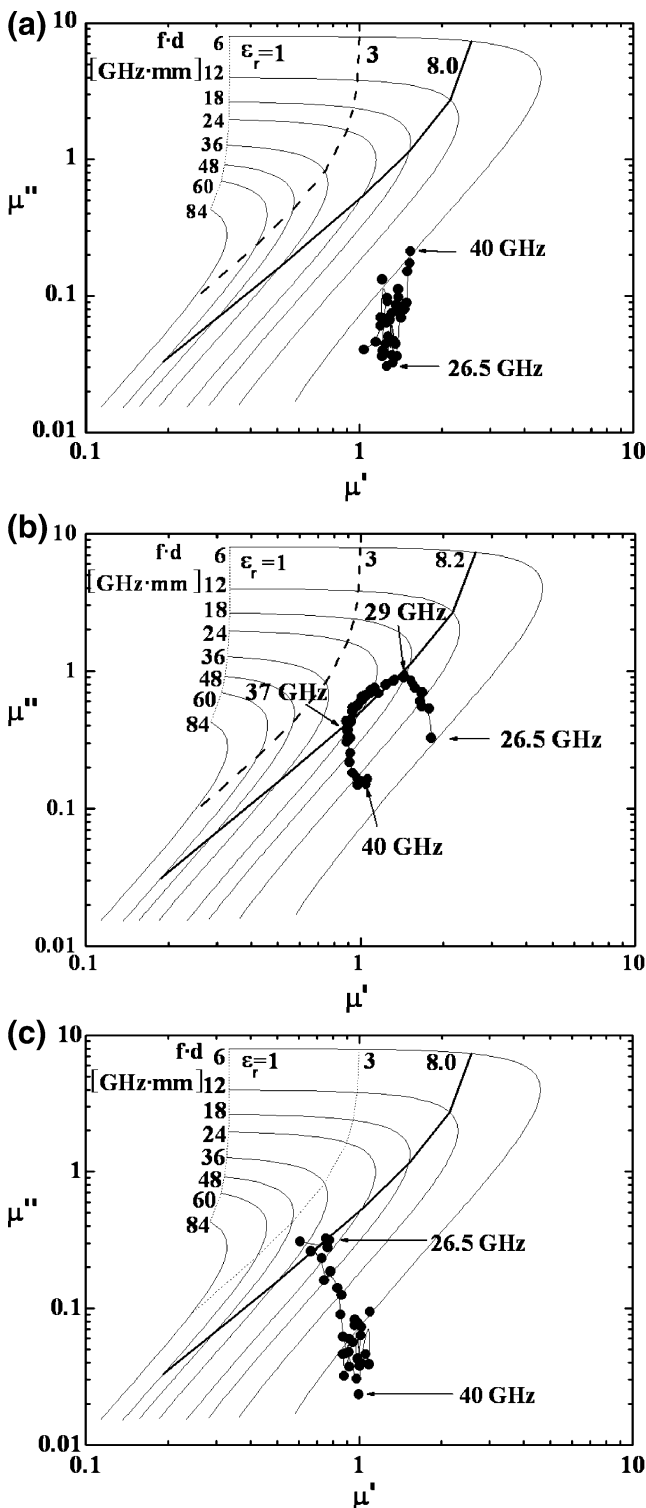


Fig. 3 Contour of complex permeability ( $\mu_r = \mu_r' - j\mu_r''$ ) of the composite specimens with  $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$  powders in the solution map of impedance matching: (a)  $x=0$ , (b)  $x=0.5$ , (c)  $x=0.8$

C. Microwave Absorbing Properties

For a microwave absorbing layer terminated by metal plate, the normalized input impedance at the absorber surface ( $Z_{in}$ ) is given by (2),

$$Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\epsilon_r}} \tanh \left[ \frac{j2\pi d}{\lambda} \sqrt{\epsilon_r \mu_r} \right] \tag{2}$$

where  $Z_0$  is wave impedance of free space ( $377\Omega$ ),  $\lambda$  is wavelength in free space, and  $d$  is layer thickness. Since the reflection coefficient ( $\Gamma$ ) is proportional to the difference between  $Z_{in}$  and  $Z_0$  as expressed in (3),

$$\Gamma = \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \tag{3}$$

reflection loss can be calculated from the measured material parameters ( $\mu_r$  and  $\epsilon_r$ ) as a function of frequency and thickness. A proper combination of  $\mu_r$  and  $\epsilon_r$  satisfying  $Z_{in}=Z_0$  at the given frequency and thickness produces the zero reflection condition (impedance matching).

Figure 3 shows the contour of impedance matching solution for zero reflection in the frequency region of Ka-band. The frequency dispersion value of  $\mu_r' - \mu_r''$  and dielectric constant  $\epsilon_r$  (about 8) of the composite specimens are plotted in the maps. The matching frequency ( $f$ ) and matching thickness ( $d$ ) can then be determined from the crossing point of complex permeability and dielectric constant, which was described in detail in the previous study [2]. For the specimen of  $x=0$  and  $x=0.8$ , magnetic loss is too small to satisfy the impedance matching in the frequency range of Ka-band as shown in Fig. 3(a) and Fig. 3(c). For the specimen of  $x=0.5$ , the impedance matching is predicted over a relatively broad bandwidth (29–37 GHz) with a matching thickness of 0.65 mm as shown in Fig. 3(b).

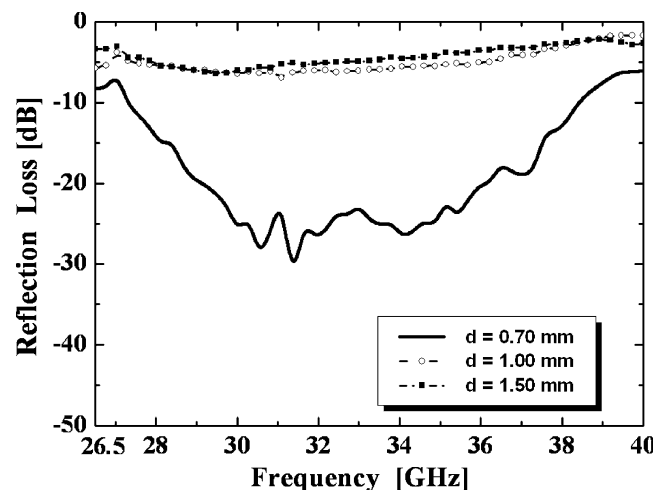


Fig. 4 Thickness dependence of reflection loss determined in the composite specimens of  $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$  powders ( $x=0.5$ )

Figure 4 shows the reflection loss determined in the specimens of  $x=0.5$ . A broad bandwidth of microwave absorption (29–37 GHz with respect to  $-20$  dB reflection loss) is predicted at the thickness of 0.7 mm. The M-type barium ferrites precisely substituted Ti and Co ions can be proposed as good absorbing materials for electromagnetic radiation in Ka-band frequencies.

#### 4 Conclusion

This study has demonstrated that Ti and Co substituted M-hexaferrites ( $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$ ) can be utilized as good microwave absorbers used at high frequencies of Ka-band (26.5~40 GHz), and the operating frequency can be controlled by substitution rate of  $\text{Ti}^{+4}$  and  $\text{Co}^{+2}$  ions for  $\text{Fe}^{+3}$  sites. The composite sheet samples containing the ferrite powders of  $x=0.5$  showed the broad bandwidth of microwave absorbance (29–37 GHz with respect to  $-20$  dB reflection loss). It was also found that the frequency dispersion of complex permeability has a strong relationship with the substitution ratio of Ti and Co. The more the Ti and Co substitution, the lower the ferromagnetic resonance frequency, which is due to weakening of crystal anisotropy field by changing of easy-axis magnetization from the c-axis to the basal plane of hexagonal cell.

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