

Magnetic and Microwave Absorbing Properties of Ti- and Co- Substituted Barium Ferrite ($\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$)

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The M-type barium ferrite ($\text{BaFe}_{12}\text{O}_{19}$) is well known magnetic material to be used as a permanent magnet due to its strong uniaxial anisotropy. The substitution of nonmagnetic Ti^{4+} and magnetically weak Co^{2+} ion for Fe^{3+} sublattices reduces the uniaxial anisotropy and those compounds open a new application field of noise suppressor at high frequencies. In this study, the magnetic and microwave absorbing properties are investigated in Ti- and Co- substituted barium ferrites ($\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$). The saturated magnetization decreases linearly with the substitution of Ti and Co. The rapid drop in coercive force is observed with Ti and Co substitution upto $x=1.2$. The magnetic permeability spectrum shows the natural magnetic resonance in the specimens with small coercive force and large attenuation of microwave is predicted in those specimens at high frequencies (above 4 GHz).

1. Introduction

Recent advances in electric and electronic equipments require the control of the EMI (Electro-Magnetic Interference) problems. The spinel ferrites (Ni-Zn or Mn-Zn ferrites) are widely used as a noise suppressor at low frequency and their basic magnetic and electric properties in correlation with microwave absorbance have been extensively investigated for this purpose [1, 2]. However, the use of spinel ferrites as the EMI suppressor should be limited below a frequency defined by Snoek limit because of the cubic magnetocrystalline anisotropy. As an example, the Snoek of Ni-Zn ferrite with relative permeability of about 10 limit is only 0.56 GHz [3]. To overcome this limit, the hexagonal ferrites with planar magnetocrystalline anisotropy should be considered.

M-type barium ferrite ($\text{BaFe}_{12}\text{O}_{19}$) is a well known material having a very strong uniaxial anisotropy and, therefore, used as a starting material for permanent magnets. However, substituting x ($\text{Ti}_x\text{-Co}_x$) for $2x$ Fe cations reduces its uniaxial anisotropy, which becomes planar on the basal plane at the substitution ratio of about $x = 1.3$ [4, 5]. The Ti-Co substituted M-hexaferrites, therefore, is a promising material for the microwave absorber at high frequencies (above 1 GHz) and worth to be investigated.

In this study, the magnetic and microwave absorbing properties of the Ti-Co substituted M-type barium ferrites ($\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$) are investigated. The static magnetic properties (saturated magnetization, coercive force) are investigated for an indirect information about the magnetocrystalline anisotropy (uniaxial or planar). From the complex

permeability spectrum, qualitative explanation is given about the relationship between magnetic resonance and static properties. A large microwave attenuation is predicted at higher frequencies than the Snoek limit of the spinel ferrites.

2. Experimental Procedures

The specimens were prepared by ceramic processing technique. The powders of $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$ composition, $x = 0.0-2.0$, were prepared by milling the calcined powder in an attritor. The calcining temperature was 1200 °C. The powder X-ray diffraction patterns indicated the completely reacted M-type barium ferrite. The powder compacts in toroidal form with 3 mm in inner diameter and 9 mm in outer diameter were sintered at 1250 °C for 2 hrs under flowing oxygen gas.

The saturated magnetization (M_s) and the coercive force (H_c) were measured by vibration sample magnetometer. The applied field for saturation was 17 KOe. 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 in the frequency range of 50 MHz-18 GHz.

3. Results and Discussion

3.1. Magnetic Properties

Fig. 1 shows the variation of magnetic properties (M_s , H_c) with the substitution of Ti^{4+} and Co^{2+} for Fe^{3+} sublattices. The saturated magnetization (M_s) decreases nearly linearly

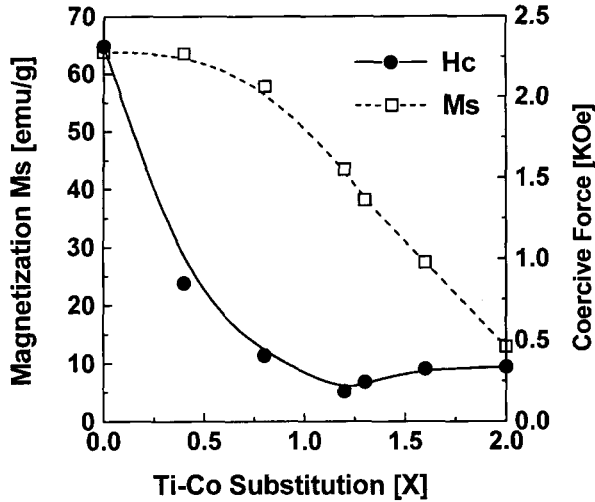


Fig. 1. The variation of saturated magnetization (M_s) and coercive force (H_c) with the substitution of Ti and Co in the M-type barium ferrite.

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 [6]. At the substitution ratio of $x=2.0$, the M_s is reduced to 12 emu/g. The result is attributed to the reduction in molecular magnetic moment by replacing the Fe^{+3} ions (magnetic moment of 5 Bohr magneton) by non-magnetic Ti^{+4} ions and magnetically weak Co^{+2} ions (3.75 Bohr magneton). The saturated magnetization is, therefore, decreased gradually with the addition of Ti and Co.

The reduction in coercive force (H_c) with Ti and Co substitution is more evident as shown in Fig. 1. The coercive force of pure barium ferrite ($x=0.0$) is very high (about

2300 Oe) which is due to strong uniaxial anisotropy along the c-axis of the magnetoplumbite structure. With the substitution of Ti and Co, the coercive force reduces rapidly to 185 Oe at $x=1.2$. Further addition of Ti and Co above 1.2 gives rise to a slight increase in coercive force ($H_c=335$ Oe at $x=2.0$).

The reduction in coercive force with Ti and Co substitution is attributed to the weakening of uniaxial magnetocrystalline anisotropy. It was reported that in the Ti-Co substituted M-hexaferrites, the magnetic anisotropy becomes planar on basal plane for the substitution ratio $x=1.3$ [4]. The present result on coercive force is quite well consistent with the previous study. The similar result of planar anisotropy was observed in the Ir-Co or Ir-Zn substituted M-hexaferrites [7].

3.2. High Frequency Properties (Complex Permeability and Permittivity)

Fig. 2 shows the frequency dispersion of complex permeability and permittivity of $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$. For the specimen of pure M-hexaferrite ($x=0.0$), the real part of permeability (μ_r') is approximately 1.1 and imaginary part (μ_r'') is negligibly small as shown in Fig. 2(a). Because of strong uniaxial anisotropy (large coercive force), no magnetic resonance is observed in this specimen.

However, in the specimens with lower coercive force (therefore, planar anisotropy), the natural magnetic resonance is observed as shown in Fig. 2(b), Fig. 2(c) and Fig. 2(d). The resonance frequency is 12.3 GHz, 1.5 GHz and 2.6 GHz for the specimen of $x=0.8, 1.2, 1.3$, respectively. Planar anisotropy makes it possible for the magnetization to

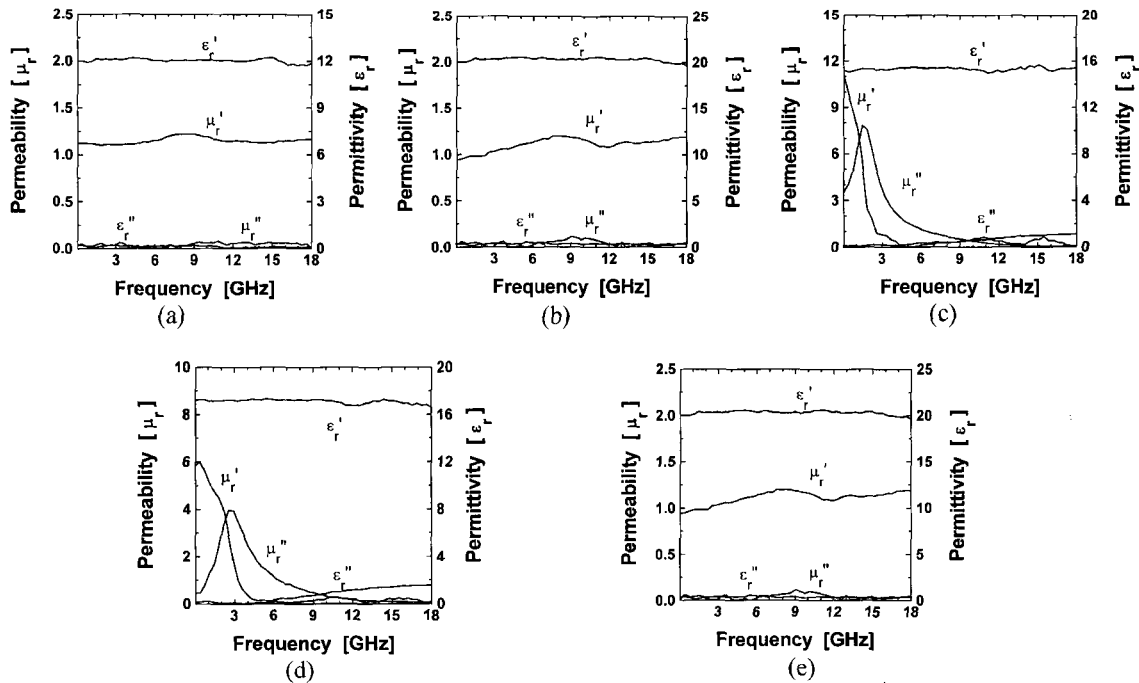


Fig. 2. Frequency dispersion of complex permeability and permittivity of $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$ specimens; (a) $x=0.0$, (b) $x=0.8$, (c) $x=1.2$, (d) $x=1.3$, (e) $x=2.0$.

rotate on the basal plane, which produces a magnetic resonance at a specified frequency of the applied alternating magnetic field. For the specimen of highly substituted with Ti and Co ($x=2.0$), no magnetic resonance is observed, (shown in Fig. 2(e)), which is considered to be due to low magnetization.

As shown in the results, it is evident that the magnetic resonance is closely related with coercive force and saturated magnetization. According to the magnetic resonance theory [3], the natural resonance frequency (f_r) is proportional to the anisotropy field (H_A), as given by

$$f_r = \frac{g}{2\pi} H_A \quad (1)$$

where g is gyromagnetic constant. Since the coercive force is, in general, proportional to the anisotropy field, the natural resonance frequency is increased with the coercive force. For the specimen of $x=1.2$, H_c is 185 Oe and f_r is 1.5 GHz. On the while, for $x=0.8$, H_c is 407 Oe and f_r is 12.3 GHz. It is evident that the more the coercive force, the higher the resonance frequency.

The real part of permittivity (ϵ_r') of the $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$ specimens is 12-20 and has a tendency to increase with the substitution of Ti and Co as shown in Fig. 2. The dielectric loss (ϵ_r'') is negligibly small for all the specimens.

3.3. Microwave Absorbing Properties

The attenuation of microwave inside a lossy material is expressed as the attenuation in decibels per length. If the field strength falls from V_0 to V_d over a length d of the sample, the absorption loss is defined as

$$20 \log \frac{V_0}{V_d} = 8.686 \alpha d \quad [\text{dB}] \quad (2)$$

where α is attenuation factor, which is given as for a magnetically lossy material [8]

$$\alpha = \frac{38.6}{\lambda_0} [\epsilon_r'^2 (\mu_r''^2 + \mu_r'^2) - \epsilon_r' \mu_r']^{1/2} \quad (3)$$

Here, λ_0 is the wavelength in free space.

Fig. 3 shows the absorption loss of $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$ specimens predicted by using Equations (2) and (3). For the specimen of $x=0.8$, the absorption loss is very high (about 60 dB/cm) in the frequency range above 12 GHz. For the specimen of $x=1.3$, the absorption loss is 30 dB/cm in the frequency range of 4~7 GHz. As seen in Equation (3), the attenuation constant is proportional to the magnetic loss (μ_r''), the absorption loss becomes large above the resonance frequency. The predicted microwave attenuation is quite well consistent with the result of permeability spectrum shown in Fig. 2.

As compared with the spinel ferrites (Ni-Zn or Mn-Zn ferrites), which is mainly used as the noise suppressor in the RF frequency (below 1 GHz), the M-type barium ferrites substituted Ti and Co can be used as the high-fre-

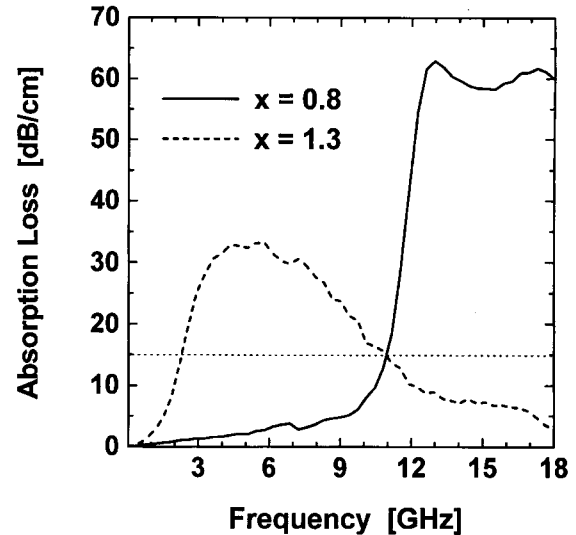


Fig. 3. Microwave absorption loss of $\text{BaFe}_{12-2x}\text{Ti}_x\text{Co}_x\text{O}_{19}$ specimens.

quency absorber above 1 GHz and the operating frequency can be controlled by the substitution ratio of Ti and Co.

4. Conclusion

This study has demonstrated that the Ti and Co substituted M-hexaferrites can be used as a microwave absorber used at high frequencies above 1 GHz, and the operating frequency can be controlled by the substitution ratio of Ti and Co for Fe^{+3} ions. For the substitution ratio $x=0.8$, the very high absorption loss (60 dB/cm in 12~18 GHz) was predicted from the measured complex permeability and dielectric constant.

It was also demonstrated that the frequency dispersion of complex permeability has a strong relationship with the basic magnetic properties (especially, coercive force). The natural resonance frequency increases with coercive force which, in turn, controlled by the substitution ratio of Ti and Co. The magnetic resonance observed in the Ti-Co substituted M-hexaferrite is attributed to the magnetization rotation from the planar magnetic anisotropy.

Acknowledgement

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References

- [1] K. Ishino and Y. Narumiya, "Development of Magnetic Ferrites: Control and Application of Losses", *Ceram. Bull.*, **66**, 1469 (1987).
- [2] J. Woody, "The Use of Ferrites as Suppressors of EMI", *IEEE Spectrum*, **1**, 21 (1990).
- [3] S. Chikazumi, *Physics of Ferromagnetism*, 2nd Ed., Clarendon Press, Oxford (1997).

- [4] J. Smith and H. P. J. Wijn, Ferrites, Phillips Technical Library, Eindhoven (1960).
- [5] H. Kojima, Ferromagnetic Materials (E. P. Wohlfarth Ed.), Vol 3, North-Holland, Amsterdam (1982).
- [6] Z. Simsa, S. Lego, R. Gerber, E. Pollert, "Cation distribution in Ti-Co-substituted barium hexaferrites: a consistent model", J. Mag. & Mag. Mater., **140-144**, 2103 (1995).
- [7] H. Vincent, E. Brando, and B. Sugg, "Cationic Distribution in Relation to the Magnetic Properties of New M-Hexaferrites with Planar Magnetic Anisotropy $BaFe_{12-2x}Ir_xMe_xO_{19}$ (Me=Co, Zn, $x=0.85$ and $x=0.50$)", J. Solid State Chem., **120**, 17 (1995).
- [8] A. Von Hippel, Dielectrics and Waves, John Wiley and Sons, New York (1959).