

## Magnetic Properties and Reflection Loss Characteristic of Mn-Ti Substituted Barium-Strontium Hexaferrite

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**Keywords:** Microwave absorber properties, magnetic properties, reflection loss, Mechanical alloying, Barium hexaferrite.

**Abstract.** The magnetic and microwave absorbing properties of the Mn-Ti substituted barium strontium hexaferrite,  $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{12-x}\text{Mn}_{x/2}\text{Ti}_{x/2}\text{O}_{19}$  ( $x$ : 0; 0.2 and 0.4) prepared by a mechanical alloying technique were investigated. Material phases in synthesized samples were identified by XRD from which results confirmed that the synthesized material was a single phase material. The hysteresis loop for the samples as analyzed by a permagraph showing those of typical of hexaferrite permanent magnet but with both remanence and coercivity decreased remarkably with the increasing fraction of Mn-Ti. The reflection coefficients  $S_{11}$ ,  $S_{21}$  of the ferrite samples were measured in the X-band frequency range (9-15 GHz) and their absorbing properties were calculated according to transmission line theory. It was observed that the absorption was taking place in the whole frequency range between 9 and 15 GHz. In addition, resonance frequencies have shifted due to the Mn-Ti substitution in barium-strontium hexaferrite. It is concluded that the Mn-Ti substituted barium-strontium hexaferrites would be a good potential candidate for electromagnetic absorbers and for other practical applications at high frequencies.

### Introduction

Technological advances information and communications equipment based on electronic and electricity is growing rapidly nowadays. It follows that electromagnetic waves which used in atmosphere will be in various frequencies covering from low to ultra-high frequency. Thus, the used of electromagnetic wave (EMW) in current advancement would bring serious concerns for all human activities. Electromagnetic interference (EMI) is one of problem encountered in the electronic based equipment system like electronic control which is being applied in hospital, military installation etc. The need for EMW absorbers with wider absorbing band-widths are ever increasing [1, 2]. The materials should have a capability to absorb and dissipate the EMW energy into heat though magnetic losses and dielectric losses once the EMW enter absorber. Compared with the ferrites with spinel and garnets structure, the ferrites with hexagonal structure is anisotropic and has a larger intrinsic magnetocrystalline anisotropy field. Because of their in-plane anisotropy, the natural resonance occurs in the GHz range that has a resonance frequency of 47.6 GHz and a big opportunity as a wave absorbent material on the microwave range [3]. So as a kind of microwave absorber, hexagonal ferrite is more adapted to the application in high frequency band. It is widely applied to the electromagnetic wave shielding and stealth technology. The hexagonal ferrites are special kinds of absorbing material due to their dielectric and magnetic losses in microwave frequency band [4]. The magnetic loss of these materials results from their ferimagnetisms, the resonance absorption of moving magnetic domains and spin relaxation in the high frequency. There are two important conditions required to be satisfied by lossy materials. The first is matched characteristics impedance, in which the intrinsic impedance of free space. This entails making the dielectric constant and magnetic permeability of the material equal each other. Second, the incident electromagnetic wave must enter and attenuated rapidly through the material layer, thus reducing the emerging wave to an acceptably low magnitude [5]. In this study, the magnetic properties and

reflection loss (*RL*) characteristics are investigated in  $\text{Mn}^{2+}$ - $\text{Ti}^{4+}$  substituted barium strontium hexaferrite in 9-15 GHz. Substitution of non-magnetic ions such as  $\text{Mn}^{2+}$ - $\text{Ti}^{4+}$  to  $\text{Fe}^{3+}$  ions are expected to change the value of a static magnetic like, coercivity, saturation magnetization which have consequences to shifting the resonant frequency of barium hexaferrite ferrite materials.

### Materials and Method

Material  $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{12-x}(\text{Mn}_{x/2}\text{Ti}_{x/2})\text{O}_{19}$  (BaSrHFO) where the substitution  $x$  was varied from 0 to 0.4 in steps of 0.2 is made from a mixture of powder  $\text{BaCO}_3$ ,  $\text{SrCO}_3$ ,  $\text{MnCO}_3$ ,  $\text{TiO}_2$ ,  $\text{Fe}_2\text{O}_3$  which has reached 99.99% purity. To get barium strontium hexaferrite, the powder of oxide must get through stoichiometric calculated and in wet milling use of acetone for 20 hours. Then it was calcinated for 4 hours at a temperature of  $1050^\circ\text{C}$  to the process of carbonization. After that, it was suppressed in the form of pellets with a diameter of 2.5 mm with pressure 70 kN. During the calcination, the evolution of gases takes place and ferrite is partially formed. This is an important homogenization operation that produces ferrites with single phase. It is heated again for 5 hours at a temperature of  $1100^\circ\text{C}$  to form crystals barium hexaferrite phase with reasonably high densities and uniform crystal structure, chemical composition and particle size. The phase analysis committed uses X-ray diffraction (XRD) Phillip PW 7310 to the source of radiation  $\text{Co-K}\alpha$  at room temperature. The XRD patterns for samples were recorded from  $20^\circ$  to  $100^\circ$  with a step scanning  $0.02^\circ/\text{min}$ . Observation of magnetic and absorption of electromagnetic waves is conducted by the use of permagraph magnetphysik-Steingroever Gmb and vector network analyzer (VNA) Advantest at the range of 9 to 15 GHz.

### Results and Discussion

Sample of BaSrHFO with doped of Mn-Ti at  $x = 0$ ,  $x = 0.2$ ,  $x = 0.4$  forms a single phase of barium hexaferrite ferrite pattern with rietveld analysis using software GSAS EXPGUI. All identified peaks correspond to the phases of barium hexaferrite ferrite based on data base of COD (Crystallography Open Database) with the entry data number 96-100-8322 which has hexagonal crystal structure with space group  $\text{Pb}63/\text{mmc}$  (194) and lattice constants  $a = 5.9290\text{\AA}$ ,  $c = 23.4130\text{\AA}$ . Figure 1 shows diffraction XRD patterns for barium strontium hexaferrite undoped and doped with  $x = 0.2$  and  $x = 0.4$ .

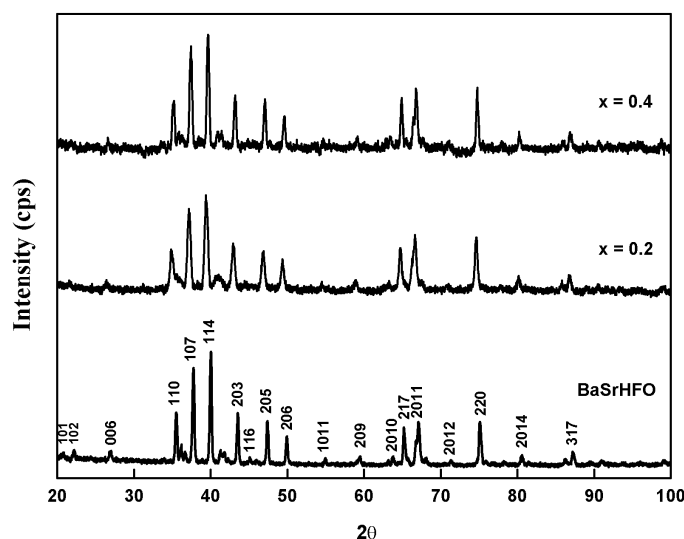


Figure 1. XRD patterns for  $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{12-x}(\text{Mn}_{x/2}\text{Ti}_{x/2})\text{O}_{19}$

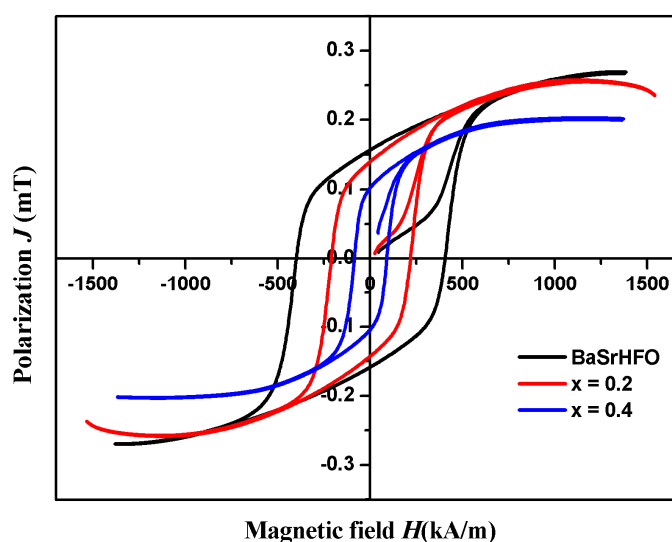
The results of diffraction are refined by Rietveld analysis. In refinement analysis, the refined were background parameters, lattice parameters, isotropic thermal parameters, scale factor, profile half-width parameters ( $u$ ,  $v$ ,  $w$ ), occupancy, and atomic functional positions. The initial Rietveld refinement was done by zero point shift, unit cell, and background parameters. The background was modeled using linear interpolation function and peak shapes were described by pseudo-Voigt.

Diffraction patterns of all samples identify the magnetic phase of barium hexaferrite although there is a difference in the intensity of the diffraction peaks. The substitution of ions  $Mn^{2+}$  and  $Ti^{4+}$  causes the changes in lattice parameter. After refinement the parameters  $R_p$  (profile fitting R-value),  $R_{wp}$  (weighted profile R-value) and  $\chi^2$  (goodness of fit quality factor) for all samples are presented in table I. From the refinement data, that derived samples are of better quality and refinements of samples are effective. The values of  $\chi^2$  show the best criteria it is between 1-1.3. With doped values, the value of “a” almost remains the same but the value of “c” is various due to difference ionic radii  $Mn^{2+}$ ,  $Ti^{4+}$ .

**Table 1.** Refined structural parameters of all prepared samples

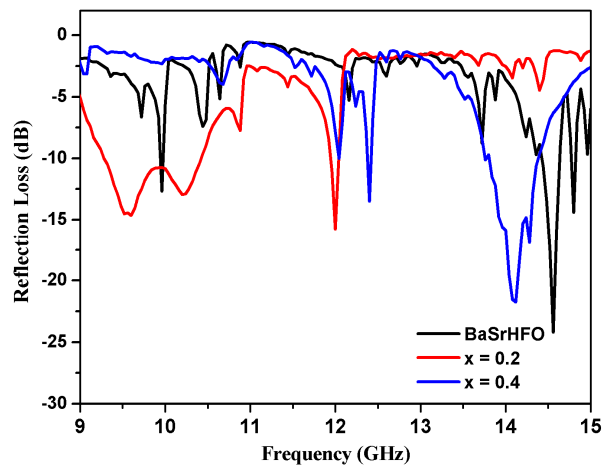
Sample	$R_p$	$R_{wp}$	$\chi^2$	a (Å)	c (Å)
Undoped (BaSrHFO)	0.0422	0.0536	1.153	5.8841	231447
Doped $x = 0.2$	0.0386	0.0483	1.148	5.8872	23.1295
Doped $x = 0.4$	0.0368	0.0467	1.172	5.8973	23.1702

Figure 2 is the hysteresis curve material barium strontium hexaferrite with undoped and doped Mn-Ti. On undoped barium strontium hexaferrite ferrite the value is higher than the doped. Magnetic polarization ( $J$ ) of 0.27 mT with coercivity field ( $H$ ) 398.7 kA/m, decreases because of the substitution  $x$ . At  $x = 0.2$  obtained magnetic polarization value of 0.25 mT and coercivity field is 209.9 kA/m while for  $x = 0.4$  obtained magnetic polarization value 0.2 mT and coercivity field worth 84.77 kA/m. Changes in the value of magnetic polarization and coercivity field due to the paramagnetic ion  $Mn^{2+}$  and diamagnetic ion  $Ti^{4+}$  which replace most of the position occupied by  $Fe^{3+}$  ions. Hexagonal structure, consist of 64 ions per unit cell. It is divided into 11 different symmetry which consists of 24  $Fe^{3+}$  ions which is distributed in five different crystallographic sites, such as, tetrahedral (4f1), octahedral (12k, 2a, 4f2) and hexahedral (2b) site of oxygen lattice. In the magnetically ordered state of  $BaFe_{12}O_{19}$ , the 12k, 2a, and 2b sites there are spins parallel to the crystallographic  $c$  axis, whereas those of 4f1 and 4f2 points are anti-parallel, constituting a net magnetic moment of 40  $\mu_B$ .  $Ti^{4+}$  ions are expected to occupy the electronegative on the side of the octahedral 4f2 while  $Mn^{2+}$  ions occupy tetrahedral side on the side of the 4f1. The result interpreted to be due to the reduction in crystal anisotropy field by changing of easy axis of magnetization from  $c$ -axis to basal plane [6, 7].



**Figure 2.** Hysteresis curve polarization of  $Ba_{0.5}Sr_{0.5}Fe_{12-x}(Mn_{x/2}Ti_{x/2})O_{19}$ , with BaSrHFO undoped and doped Mn-Ti with  $x = 0.2$ ,  $x = 0.4$

Figure 3 shows the effective value of electromagnetic wave absorbing materials as a function of frequency. At BaSrHFO material without doped seen some absorption peaks in the range of 9.5 to 11 GHz and 13.5 to 15 GHz with an effective depth of absorption at 14.6 GHz frequency with a value of -24.8 dB absorption effectiveness. On the substitution  $x = 0.2$ , it is obtained some extreme peaks occur in the frequency range of 12 to 12.5 GHz and 14 GHz in the range up to 14.5 GHz with a maximum reflection loss occurred at 14.1 GHz with a value of -22.5 dB. While at  $x = 0.4$ , there are two extreme absorption peaks in the frequency range of 9 to 11 GHz and at 12.1 GHz with a maximum reflection loss -15.2 dB.



**Figure 3.** Variation of reflection loss vs. Frequency spectra of barium strontium hexaferrite undoped and doped with variation  $x = 0.2, 0.4$

The factors resulting in the multippeak phenomenon during the ferromagnetic resonance include the motion of domain walls, the polycrystalline effect and the spin wave instability [8]. Magnetic crystalline field of barium hexaferrite is as large which corresponds to a natural resonance frequency due to its large crystalline magnetic anisotropy. The substitution of magnetic and non-magnetic such as  $\text{Mn}^{2+}$  and  $\text{Ti}^{4+}$  ions for  $\text{Fe}^{3+}$  into ferrites changes the direction of the magneto-crystalline anisotropy of the material from the  $c$ -axis towards the  $a$ - $b$  basal plane. The cause of this anisotropy is the effective coupling of the spins of the magnetic ions and the crystalline electric fields that act upon the ions via spin-orbit coupling. Such electric fields depend on the magnitude and symmetry of the positions of the neighboring ions [9]. So with such  $\text{Mn}^{2+}$ - $\text{Ti}^{4+}$  substitutions, the resonance frequency can be shifted to lower frequencies.

## Conclusion

This study has demonstrated that Mn-Ti substituted barium strontium hexaferrite as a microwave absorber. The Static of magnetic properties of barium strontium hex ferrite material changes after the substitution of  $\text{Mn}^{2+}$ - $\text{Ti}^{4+}$ . The minimum value of coercivity and magnetic saturation occurs in the substitution  $x = 0.4$  and a value of saturation magnetization and coercivity of each field is 0.2 mT and 84.77 kA/m. While the absorption of electromagnetic waves occur in all samples with different absorption peaks which indicates the material barium strontium hexaferrite with variations substitution  $x = 0.2$  and  $x = 0.4$ . All are the wave absorbent materials from this research, in the microwave frequency range.

## Acknowledgment

This research was funded by Hibah DIKTI for the fellowship granted to first author of this study through research grant number 2026/H2.R12.3/PPM.00 Penelitian/2011 and research competitive of decentralization with grant number 014/K3.KU/2012. Thanks to Mr. Asep Yudi research staff of LIPI Bandung for help in the use of VNA from which the authors are very grateful.

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10.4028/www.scientific.net/AMR.896

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10.4028/www.scientific.net/AMR.896.418