Synthesis and Characterization of Barium-Hexaferrite-Based Nanocomposite on X-Band Microwave

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Abstract. In this research, synthesis and characterization of barium-hexaferrite-based nanocomposites were conducted with silicone rubber as a microwave absorber in x-band frequency. Barium-hexaferrite nanoparticle powder is prepared with high energy milling for 75 h. Nanocomposites were prepared by weight percentage of barium-hexaferrite to silicon rubber with a variation of 20% to 50%. X-ray diffraction (XRD) is used for structural analysis and phase in still-powder conditions. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) are used for surface morphology analysis. The vibration of sample magnetometer (VSM) and vector network analyzer are respectively used for static magnetic analysis and microwave absorption properties. The characterization results show that the sample has a single phase with a hexagonal crystal structure. The surface morphology is almost homogeneous with an almost spherical grain shape and a nanoparticle size of 20 nm to 30 nm. Magnetic saturation (Ms) reached 52.0 emu·g⁻¹, magnetic remanent (Mr) 33.0 emu·g⁻¹ and coercivity (Hc) 0.32 T. Ability to absorb microwaves expressed by reflection loss (RL) reached the maximum for the sample with 20% composition, i.e. about −15 dB at a frequency of 10.8 GHz. While for the composition of 30% and 50% the ability to absorb microwaves in a row is −7 dB (10.8 GHz) and −5.2 dB (10.6 GHz).

Keywords. Barium-hexaferrite, microwave absorber, nanocomposite.

1. Introduction

Technological developments, especially in microwave absorption technology, attracted the attention of many researchers. The type of material that can be used as a microwave absorber has a magnetic, electrical and dielectric loss. Among the magnetic materials that can be used as a microwave absorber is barium hexaferrite which has a hexagonal structure, high magnetic saturation and chemical stability. To further enhance its ability to absorb microwaves, it is first processed through doping procedures in the position of Ba, either in the Fe position or both at once [1–7]. The work already carried out refers to variations in magnetic permeability, dielectric permittivity at GHz, and absorption properties with composition and sample-making procedures but has not much been studied about the composite of the compound.
Polymers have several advantages, including ease of processing and formation, thermomechanical stability, and low cost. Epoxy resin is commonly used because it is extensively used for the production of high-tech composite in the fields of automotive, aerospace and electronics industries, mostly due to their high stiffness [8, 9]. The barium ferrite composite as the content with the epoxy resin will have dielectric permittivity, magnetization and magnetic saturation increased with the increased barium ferrite content [10]. Particles of nanosize in carbon-epoxy resin composites will produce better microwave absorption [11]. Conductivity, saturation magnetization and coercivity decrease with the incorporation of MnO₂ contents [12]. We hope that the thinner thickness of silicon rubber and barium-strontium hexaferrite can improve the ability to absorb microwaves.

In this paper, the nanocomposite is formed from silicone rubber and barium-strontium hexaferrite nanoparticles with a content variation of 20 % to 50 % by weight. Nanocomposite printed sheets with a thickness of 1 mm. Analysis of structures and phases in the form of powder used X-ray diffraction (XRD) and for surface morphology analysis used scanning electron microscopy (SEM) and transmission electron microscopy TEM, while the analysis of the ability to absorb microwaves used vector network analyzer (VNA).

2. Materials and methods
The Ba₀.₆Sr₀.₄Fe₁₀MnTiO₁₉ sample with certain composition was prepared using a conventional milling technique through stoichiometric quantities of analytical-grade BaCO₃, SrCO₃, Fe₂O₃, MnCO₃, and TiO₂ precursors with high purity were mixed and milled using high-energy milling (HEM) type Spex 8000 for up to 5 h. The samples were characterized by powder X-ray diffraction (XRD) using Pan Analytical diffractometer with CuKα radiation (λ = 1.5406 Å). The powders were then compacted into pellets and sintered in the electric chamber furnace Thermolyne at 1000°C for 5 h to obtain crystalline materials. The Ba₀.₆Sr₀.₄Fe₁₀MnTiO₁₉ sample was re-milled for 75 h to obtain nanoparticles.

The absorber nanocomposite was made from silicon rubber and Ba₀.₆Sr₀.₄Fe₁₀MnTiO₁₉ nanoparticle. The matrix of nanocomposite absorber was methyl vinyl silicone rubber and toluene as vulcanized assistants. The silicon rubber and Ba₀.₆Sr₀.₄Fe₁₀MnTiO₁₉ nanoparticle were mixed and guaranty a better dispersion. The diameter of nanocomposite absorber was about 15 cm and the thickness was about 2 mm. The various volume fractions of the Ba₀.₆Sr₀.₄Fe₁₀MnTiO₁₉ nanoparticle were added into the silicone rubber with the weight ratio was 20 wt%, 30 wt% and 50 wt% then were mixed in each composite. Nanostructural of the samples were observed using the scanning electron microscope (SEM) and transmission electron microscope (TEM). The magnetic properties were evaluated using an Oxford Instrument Vibrating Sample Magnetometer (VSM). Meanwhile the reflection loss of microwave was measured using the vector network analyzer (VNA) with a frequency range of 8 GHz to 12 GHz.

3. Results and discussion
Figure 1a shows the refinement pattern of X-ray diffraction sample Ba₀.₆Sr₀.₄Fe₁₀MnTiO₁₉ with milling time of 75 h. The result of this refinement produces excellent quality fittings with a very small R factor as well. The R factor is a very small criterion of fit and χ² (chi-squared) (1.036). The complete refinement results can be seen in Table 1.

| Lattice parameters, crystal structure, R-factor and χ² |
|-------------------------------------------------------|
| Phase BaFe₁₂O₁₉                                      |
| Space group: P 63/m m c (194), crystal structure: Hexagonal |
| Lattice parameters: a = b = 5.8984(3) Å and c = 23.171(1) Å, α = β = 90° and γ = 120°, V = 698.1(1) Å³ and ρ = 5.614 g.cm⁻³ |
| R-factor   Rp = 3.10                                       |
| χ² = 1.036                                          |
Figure 1a. Refinement of x-ray diffraction pattern of Ba$_{0.6}$Sr$_{0.4}$Fe$_{10}$MnTiO$_{19}$

Figure 1b. Cell unit of Barium Hexaferrite [13]

Figure 1b shows crystal structure of M-type barium hexaferrite BaFe$_{12}$O$_{19}$ [13]. This hexagonal structure is constructed into 4 sub-unit blocks called the sub-units S and S* blocks with the chemical formula (Fe$_{6}$O$_{8}$)$_{2}^{2+}$ and sub-unit blocks R and R* with the chemical formula (Ba$_{2+}$Fe$_{6}^{3+}$O$_{11}$)$_{2}^{-}$. The (*) indicates that the sub-unit relationship is rotated 180° around the hexagonal axis. The sub-unit blocks S and S* are spinels with 2 layers O binding to 6 Fe$^{3+}$ ions (4 Fe$^{3+}$ ions form the octahedral and 2 Fe$^{3+}$ ions form the tetrahedral). While the sub-unit blocks R and R* consisting of 3 layers of O, 1 layer O bind to the Ba$^{2+}$ ions and the other 2 layers O bond with 6 Fe$^{3+}$ ions to form octahedral. The colors of spheres; red, pink, gray, blue and yellow represent iron atoms in 2a,4f$_{1}$,12k,4f$_{2}$, and 2b sites respectively. Small green spheres represent oxygen atoms, while larger cyan spheres represent barium atoms [13].

The single phase Ba$_{0.6}$Sr$_{0.4}$Fe$_{10}$MnTiO$_{19}$ has a homogeneous microstructure of surface morphological observations as shown in Figure 2. Surface morphological observation results show that the sample has a shape and a particle size distribution of about 20 nm to 30 nm. This result is very much in accordance with the characterization results using TEM which can be seen in Figure 3. S.S.S.Afghahi et al obtained an average particle size of about 250 nm to 300 nm [14].

Figure 2. Surface morphology of Ba$_{0.6}$Sr$_{0.4}$Fe$_{10}$MnTiO$_{19}$ using SEM

Figure 3. Characterization of sample particles Ba$_{0.6}$Sr$_{0.4}$Fe$_{10}$MnTiO$_{19}$ using TEM.
The result of characterization of magnetic properties of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Fe}_{10}\text{MnTiO}_{19}$ particle can be seen in Figure 4. The loop of the hysteresis curve consists of the intrinsic saturation of $\text{Ms}$, remanence $\text{Mr}$, and coercivity $\text{Hc}$. This saturation is an area where the material is unable to absorb strong magnetic fields such as magnetization strengthening and does not produce significant changes in magnetic flux density. Remnant $\text{Mr}$ for the samples shows that the magnetization left in the media after the outside magnetic field has been removed and the coercivity indicating the coercive power of the material is the same as the demagnetizing force required to reduce residual induction to zero in the magnetic field after the magnetization is saturated. The complete result of magnetization can be seen in Table 2.

![Hysteresis curve of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Fe}_{10}\text{MnTiO}_{19}$ using VSM](image)

**Figure 4.** Hysteresis curve of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Fe}_{10}\text{MnTiO}_{19}$ using VSM

| t (h) | Coercivity, $\text{Hc}$ (T) | Saturation, $\text{Ms}$ (emu$\cdot$g$^{-1}$) | Remanence, $\text{Mr}$ (emu$\cdot$g$^{-1}$) |
|-------|-----------------|-----------------|-----------------|
| 75    | 0.32            | 52.0            | 33.0            |

**Table 2.** Summary of magnetic properties data $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Fe}_{10}\text{MnTiO}_{19}$

Ability to absorb microwaves is done using VNA. For composites with variations of the filler composition $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Fe}_{10}\text{MnTiO}_{19}$ 20 % to 50 % can be seen in Figure 5. It appears that the 20 % composition actually shows as the best absorbent compared to the larger composition. The maximum value of Reflection Loss (RL) is about $-15$ dB at a frequency of 10.8 GHz. S.S.S.Afghahi et al found that the maximum value of reflection loss was about $-2.4$ dB [14].

![Reflection loss curve (RL) $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Fe}_{10}\text{MnTiO}_{19}$ with filler variation between 20 % to 50 %](image)

**Figure 5.** Reflection loss curve (RL) $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{Fe}_{10}\text{MnTiO}_{19}$ with filler variation between 20 % to 50 %.
4. Conclusions
Nanocomposite has been successfully manufactured with barium-strontium hexaferrite nanoparticles as fill and silicon rubber with a content variation of 20 wt% to 50 wt%. The characterization of the ability to absorb microwaves using a vector network analyzer (VNA) found that the higher the content, the weaker the ability to absorb microwaves. The 20 % composition has a maximum reflection loss value (RL) of about –15 dB at a frequency of 10.8 GHz with a bandwidth of 1.0 GHz.

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