Effects of the geometry factor on the reflection loss characteristics of the modified lanthanum manganite

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Abstract. The synthesis and characterization of modified lanthanum manganite materials by using mechanical milling technique have been performed. This magnetic material is prepared by oxides, namely lanthanum oxide, barium carbonate, iron oxide, titanium dioxide, and manganese carbonate. The mixture was milled for 10h, compacted at 5000 psi into pellets with three kinds of different thickness (d = 1.5, 2.0 and 4.0 mm, respectively) and then sintered at temperature of 1000 °C for 10h. The refinement results of x-ray diffraction pattern showed that the sample is single phase. The sample had composition in accordance to stochiometry composition. The geometry factor consists of the particle morphology observed using scanning electron microscope and thickness measured millimeter device. The microstructure analyses shows that the particle shapes was aggregates with the particle sizes distributed homogeneously on the surface of the sample. The results of the microwave absorption indicated that there were three of absorption peak frequency at 9.9 GHz, 12.0 GHz, and 14.1 GHz. The microwave absorption of the sample increases with increasing thickness of absorption area. We concluded that the increasing thickness of absorption region resulted new resonance frequencies, and the new resonance frequencies are joined to each other to form a wider resonance frequency and it is known as a broadband absorber. The first section in your paper.

1. Introduction

The demand for the reduction of electromagnetic wave interference is a topic of interest at the moment. Electromagnetic wave interference is a emission of high frequency noise that poses a problem for electronic devices which are working at high frequency due to its ability to reduce the performance of the electronic equipments. Therefore, an absorber material is needed which is capable to absorb electromagnetic waves in such manner so that the electromagnetic wave interference could be eliminated. Recently some modified materials such as hexaferrite-based, perovskite-based, and carbon-based have been found to exhibit the microwave absorption characteristics [1-5]. Development of magnetic material as absorber materials is required by a structural-engineering technique in which it is expected that the end product should be a superior material suitable for microwave absorber applications [6-9].
There are at least two factors that affect the high performance of absorbing material, namely intrinsic and extrinsic factors. The prerequisite of intrinsic factor that is required of an electromagnetic wave absorber material is that a material must have both the permeability and the permittivity, meanwhile the extrinsic factor is the geometry factor such as particle shape and thickness of absorption area [10-12] have reported that the authors have succeeded in the modification of a manganite-based material in which they performed a material modification of Fe substituted lanthanum manganite system in order to increase the material’s permeability. The nano-crystalline powders, is a type of an excellent microwave absorbing material, which could be synthesized by the sol–gel method. The authors determined the relationship between reflection loss and frequency for their samples with three different thicknesses \((d = 1.80, 2.00\) and \(2.21\) mm, respectively). The absorption peaks increase with the thickness [12] reported that they have carried out a convenient graphical method for the determination of the optimal thickness for single-layer absorbers under normal wave incidence. Matching condition demands coincidence of thicknesses of equal amplitudes and phases for reflection and transmission coefficients of the wave reflected from the first-boundary air-absorber and the wave transmitted and reflected from second-boundary absorber-metal, respectively. Reflection loss maps are constructed as functions of layer thickness and one of the material parameters \(tg\varepsilon\) or \(tg\mu\) for constant ratio \(\varepsilon/\mu\) and another parameter \(tg\mu\) or \(tg\varepsilon\) ((\(\varepsilon\) and \(\mu\) are the real parts of complex relative permittivity and permeability and \(tg\varepsilon\) and \(tg\mu\) are the dielectric and magnetic loss tangents of the material) [13].

In this work, the authors performed a further study on this particular topic based in part on the results of previous studies. More specifically, in this work synthesis of modified lanthanum manganite nanoparticles is carried out, and in addition, the effect of absorption thickness on the absorption properties of the material has also been investigated. The aim of this study is to synthesize nanoparticles of modified lanthanum manganite and to understand the changes in the extrinsic factors of microwave absorption performance of this material caused by the particle shape and thickness of absorption area.

2. Materials and Methods
Modified lanthanum manganite referred is a perovskite-based magnetic material and its molecular formula is \(\text{La}_{0.3}\text{Ba}_{0.2}\text{Fe}_{0.3}\text{Mn}_{0.35}\text{Ti}_{0.35}\text{O}_{3}\). The material is synthesized by a solid-state reaction method through mechanical milling technique using a high-energy milling (HEM) of the type Spex 8000. The synthesis process of the material was carried out according to research result of Azwar et al [14]. However, the sample was re-milled for 30 hours to obtain nanocrystallites powders. The aim of the 30 hours re-milling was to obtain a more perfect quality improvement over the previous research results. The re-milled material was compacted at 5000 psi into pellets with three different thickness \((d = 1.5, 2.0\) and \(4.0\) mm, respectively).

The characterization of the product materials consist of phase analysis using Miniflex Rigaku diffractometer (XRD) equipped with a tube of cooper anode (CuK\(\alpha\)). An additional Rietveld analysis was performed applying the GSAS program [15]. Elemental composition of the material was measured by using energy dispersive spectroscopy (EDS) method, meanwhile the particle morphology of the sample is observed by using the scanning electron microscope (SEM). The microwaves absorption characteristic investigation was carried out by using vector network analyzer (VNA) in order to obtain the effects of reflection and transmission of electromagnetic waves from signal source with a certain frequency. The reflection loss (RL) property of the samples was measured by using a Vector Network Analyzer ADVANTEST type R3770 in the frequency range of 300 kHz - 20 GHz (Figure 1(a)). For the analysis of microwave reflection and transmission, testing was conducted in the frequency range of 5 GHz to 18 GHz with a prototype form of bulk samples with a diameter of 1.5 mm.
The electromagnetic wave propagation scheme in adapter of vector network analyzer (VNA) is shown in Figure 1(b). Wave coming from the Port 1 and the reflected wave (S11) are also received by the Port 1 and wave transmission (S21) was received by the Port 2. Polarization of electromagnetic waves in the waveguide is very useful for analyzing the electrical properties of the material thickness d. To obtain accurate data on the measurement of reflection and transmission coefficients, the measurement needs to be performed using TRL calibration (reflection transmission line).

3. Results and Discussion

Figure 2(a) shows the refinement results, which indicate that a very good quality of fitting with a small $R_{wp}$ factor of 7.63 and a goodness of fit value $\chi^2$ (chi-squared) of 1.302 have been achieved; and which also are in good agreement with the established rietveld refinement standards [15]. The refinement results of x-ray diffraction pattern show that the sample is a single phase of lanthanum manganite. The structure parameter of the sample compared with previous research [14]

| Sample | Previous research [11] |
|--------|------------------------|
| Monoclinic, Space group : $I1\bar{2}/a\bar{1}$, $a = 5.5386(9)$ Å, $b = 5.577(1)$ Å, $c = 7.8465(1)$ Å, $\alpha = \gamma = 90^\circ$, $\beta = 89.80(2)^\circ$ $V = 242.88(7)$ Å$^3$, and $\rho = 6.407$ gr.cm$^{-3}$ Single phase | Monoclinic, Space group : $I1\bar{2}/a\bar{1}$, $a = 5.5182(8)$ Å, $b = 5.5442(8)$ Å and $c = 7.822(1)$ Å $\alpha = \gamma = 90^\circ$ and $\beta = 89.63(1)^\circ$ $V = 239.68(3)$ Å$^3$, $\rho = 6.559$ gr.cm$^{-3}$ Single phase |
In Table 1 the lattice parameter values, the volume unit cell, and atomic density values (which are approximately similar to the results of a previous work) are tabulated so that required support data to exact elemental content in the sample. The elemental content of the $\text{La}_{0.8}\text{Ba}_{0.2}\text{Fe}_{0.3}\text{Mn}_{0.35}\text{Ti}_{0.35}\text{O}_3$ is shown in Figure 2(b) and Table 2.

**Table 2.** The results of element analysis by using energy dispersive spectroscopy

| No. | Unsure | Content (wt.%$\pm$0.14) | Content (wt.%$\pm$0.14) [14] |
|-----|--------|--------------------------|-----------------------------|
| 1.  | Lanthanum (La) | 48.64 ± 0.49 | 16.64 |
| 2.  | Barium (Ba) | 12.44 ± 0.40 | 4.30 |
| 3.  | Titanium (Ti) | 4.10 ± 0.19 | 4.07 |
| 4.  | Manganese (Mn) | 6.27 ± 0.29 | 5.42 |
| 5.  | Iron (Fe) | 7.17 ± 0.29 | 6.10 |
| 6.  | Oxygen (O) | 21.38 ± 0.14 | 63.47 |

EDS spectra shows that the elements composition consist of lanthanum (La), barium (Ba), titanium (Ti), manganese (Mn), iron (Fe) and oxygen (O). This means that the sample had composition in accordance with the stoichiometry composition. The Williamson-Hall method was used to calculate the mean crystallite size by X-ray broadening peaks analysis of the sample [14].

![Williamson hall curve](image1)

**Figure 3.** The crystallite size and particle size of the samples

Figure 3(a) shows a curve of $B_{1}$ Cos $\theta$ vs. Sin $\theta$ plot for the sample according to the results based on the Williamson-Hall calculation method. The linear regression graph from the curve can be used to determine the mean crystallite size of the sample. The mean crystallite size of the sample is around 41 nm. Figure 3(b) is the particle size of the sample observed by using transmission electron microscope (TEM). TEM image shows that the sample has particles with a varying diameter of around 100 – 200 nm and a spherical shape. Thus, the analysis result of X-ray diffraction profile shows that the synthesis of modified lanthanum manganite have been successfully accomplished with a single phase and the mean crystallite size of 41 nm. This result signifies that the method is reproducible and has a similarity with the previous research result [14].

For further analysis, the authors measured the performance of the sample’s absorbing properties with three kinds of different thickness ($d$ = 1.5, 2.0 and 4.0 mm, respectively). The measurement result of the total absorption of the sample in the frequency range 9-15 GHz is shown in Figure 4. The sample has a relatively significant absorption rate, which occurs in a relatively wide range. There is an indication that an absorption of microwaves in the frequency range of 9 to 15 GHz has been found.
Microwave absorption in the samples increases with increasing thickness of the absorption field. For the thickness of the sample $d = 1.5$ mm it seems that there is an absorption of microwaves in the frequency range between 9-15 GHz with three peaks of frequency absorption at 9.9 GHz (peak – 1), 12.0 GHz (peak – 2), and 14.1 GHz (peak – 3), with reflection loss of $\sim 9 \text{ dB}$, $\sim 8 \text{ dB}$, and $\sim 23.5 \text{ dB}$, respectively. For the thickness of $d = 2.0$ mm it seems that there are four absorption peaks, namely at the frequencies of 10.1 GHz (peak – 1), 11.5 GHz (peak – 2a), 13.0 GHz (peak – 2b), and 14.2 GHz (peak – 3) with reflection loss of $\sim 13 \text{ dB}$, $\sim 10 \text{ dB}$, $\sim 7 \text{ dB}$ and $\sim 23.0 \text{ dB}$, respectively. Then when the thickness of $d$ is equal to $4.0$ mm, it seems that there are four absorption peaks, appearing at the frequencies of 9.8 GHz (peak – 1), 11.3 GHz (peak – 2a), 13.1 GHz (peak – 2b), and 14.3 GHz (peak – 3) with reflection loss of $\sim 7 \text{ dB}$, $\sim 13 \text{ dB}$, $\sim 20 \text{ dB}$ and $\sim 22.5 \text{ dB}$, respectively.

The three sharp peaks (peak – 1, peak – 2 and peak – 3) are known as the resonance absorber when the thickness $d$ is equal to $1.5$ mm. Resonance absorber is the absorbing characteristics of microwaves which occur at certain frequencies. It is suspected that these peaks are part of the microwaves absorption characteristics in this material. Then the thickness increased up to $d = 2.0$ mm, absorbing energy (peak – 1) also increases which is characterized by the reflection loss (RL) value increase from $\sim 9 \text{ dB}$ up to $\sim 13 \text{ dB}$ (the absorption percentage increases from 70 % to 80 %). And then the absorbing energy (peak – 2) also increases from $\sim 9 \text{ dB}$ up to $\sim 10 \text{ dB}$ but the resonance frequency shifts to the left because it displays a new peak of resonance frequency at 13 GHz (peak – 2b) with RL of $\sim 9 \text{ dB}$. Absorbing energy (peak – 3) appears to be relatively fixed, but it is slightly shifted to the right due to the presence of the resonance frequency on the peak – 2b increases. Then, when the thickness of the samples increased to 4.0 mm, the absorbing energy (peak – 1) decreases but this peak is broad. It shows that this peak occurs at the resonance frequency more evenly and it is known as a broadband absorber. Absorbing energy (peak – 2a) increases from $\sim 10 \text{ dB}$ up to $\sim 13 \text{ dB}$ (the absorption percentage up to 80 %). The absorbing energy (peak – 2b) increases significantly from $\sim 7 \text{ dB}$ up to $\sim 20 \text{ dB}$ (the absorption percentage up to 90 %). And the absorption energy (peak – 3) appears relatively fixed and is shifted slightly to the right due to the presence of the resonance frequency on the peak – 2b increase. Increasing reflection loss allegedly came from the influence of the geometry (shape of the particles) in the material as shown in Figure 4 (b). The microstructure analysis shows that the particles’ shape was spherical aggregates with the particle sizes distributed homogeneously on the surface of the sample. If there is a microwave radiation incident upon the material, the microwave beam will be scattered in all directions so that the beam is not reflected back, as though it was being absorbed due to the scattering process. This scattering factor increases with increasing sample thickness (geometrical factors). In the end, the geometry factor (extrinsic) can increase the absorption of microwave in addition to the intrinsic factors such as the materials permeability and permittivity.
4. Conclusion
Sample of modified lanthanum manganite have been prepared by mechanical milling technique to study the effect of the geometrical factor on the reflection loss characteristic. The sample is single phase with a monoclinic structure, having a crystalline phase of I12/a1, the particle size of 100-200 nm, and the particles have a spherical shape. The microstructure analyses showed that the particles morphology consists of aggregates structures with the particle sizes distributed homogeneously on the surface of the sample. The microwave absorption of the sample increases with increasing thickness of absorption area. We concluded that the increasing thickness of absorption region have resulted in the appearance of new resonance frequencies, and the new resonance frequencies are joined to each other to form a wider resonance frequency which is known as a broadband absorber.

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