Structural, Magnetic and Electrical Properties of La$_{1-x}$Ba$_x$MnO$_3$ (x = 0 – 0.7) for Absorber Electromagnetic Wave

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ABSTRACT

The synthesis and characterization of La$_{1-x}$Ba$_x$MnO$_3$ magnetic materials by mechanical alloying method have been performed. This magnetic material is prepared by oxides, namely La$_2$O$_3$, BaCO$_3$ and MnCO$_3$. The mixture was milled for 10 h then sintered at temperature of 1000 °C for 10 h. The refinement results of x-ray diffraction pattern showed that the sample La$_{1-x}$Ba$_x$MnO$_3$ (x = 0.2) is the best composition with monoclinic structure, space group of $I 2/a$ 1, the lattice parameters of $a = 5.5182(8)$ Å, $b = 5.5442(8)$ Å and $c = 7.822(1)$ Å, $\alpha = y = 90^\circ$ and $\beta = 89.63(1)^\circ$, $V = 239.68(3)$ Å$^3$ and $\rho = 6.559$ gr.cm$^3$. The microstructure analyses showed that the particle shapes was aggregate with the particle sizes around 500 nm and distributed homogeneously on the sample surface. The elementary analysis results indicated that the sample has a composition according to expected. The hysteresis curve showed that the presence of barium caused the change magnetic ordering of the sample from antiferromagnetic to ferromagnetic. The electric and magnetic properties of this material is affected by the very complex interaction between charge, spin-ordering and orbital ordering of the electron spin. The results of the electromagnetic wave absorption curve analysis by using a vector network analyzer indicated that that there has been absorption of EM waves in the frequency range between 9-15 GHz with two peak frequency at 11.1 and 14.2 GHz with absorption of ~ 4.8 and 6.8 dB, respectively. The EM wave absorption is calculated reaches 55% with a thickness of 1.5 mm. We concluded that the material lanthanum manganite perovskite can be used as preliminary study development new materials for electromagnetic wave absorber.

KEYWORDS: perovskite, lanthanum manganite, substitution, absorber, electromagnetic wave, structural, magnetic, and electric.

1. INTRODUCTION

Magnetic material utilization as an electromagnetic wave absorber materials become very popular today especially for electronic applications due to the presence of bias field is caused by electromagnetic wave interference that able to reduce the performance of the electronic equipment. And to eliminate the bias field is required a materials that able to resonate at a particular frequency so that it can absorb the electromagnetic radiation is undesirable. Necessary prerequisite as an electromagnetic wave absorber material is the material had high permeability and permittivity. At first the magnetic materials of ferrite-based that are excellent in engineered-structures to be developed into the absorber material of electromagnetic wave due to they has a high permeability. The last decade also has be discovered an electromagnetic wave absorber materials that developed by a materials of ABO$_3$ composition based-manganite due to they had a high permittivity. However this material is originally applied to the magnetic data storage (magnetic storage devices) or as cathodes in solid oxide of fuel cells. One of ABO$_3$ composition based-manganite is lanthanum manganite (LaMnO$_3$).

The lanthanum manganite is the parent of one of the family of magnetic materials which the last few decades attracted much attention due to they show the electric and magnetic properties are varied depend on the composition. The previous study of the modifying lanthanum manganite has carried out by some researcher with various dopant of a particular elements in lanthanum ions. Pissas et al. has studied the crystal and magnetic structure of the La$_{1-x}$Ca$_x$MnO$_3$ compounds. Ulyanov et al. has studied the importance of metamagnetism for the giant magnetocaloric effect in lanthanum manganites and the key role of oxygen deficiency in L$_{1-x}$Ca$_{0.33}$Sr$_x$MnO$_{3.6}$ manganites. Alifanti et al. has also studied the effect of lanthanum substitution by cerium on the catalytic activity of La$_{1-x}$Ce$_x$MnO$_3$ catalysts and its relation to their physico-chemical characteristics. And Li et al. has studied the microwave-absorbing properties of La$_{1-x}$Sr$_x$MnO$_3$ powders.

Magnetic properties of this system are affected by a change valence on the Mn atom. These changes are referred to as charge-ordered, which may affect the magnetic structure of Mn atoms in lanthanum manganite system. A very strong correlation between structure, charge carriers, magnetic properties, and the phenomenon of absorber electromagnetic wave become this material very interesting to be understood. This study will be carried out synthesis of lanthanum barium manganite material. So the discussion in this study focused on the synthesis and characterization of structural and magnetic properties of materials lanthanum manganite.

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perovskite system as preliminary study of new materials development for electromagnetic wave absorber. Thus the aim of this study was to synthesize and characterize magnetic materials lanthanum manganite systems and understand the characteristics of the crystal structure, magnetic, electrical and electromagnetic wave absorber properties of this material.

2. MATERIALS AND METHODS

The synthesis of La$_{1-x}$Ba$_x$MnO$_3$ ($x = 0 – 0.7$) materials are carried out by using the solid reaction method. The samples are prepared by oxide materials, namely La$_2$O$_3$, BaCO$_3$, and MnCO$_3$ from Merck product with high purity. The mix is milled by using a high-energy milling (HEM) type Spex 8000 for 10 hours at room temperature. The finally mix powder is compacted at 5000 psi into pellets and sintered in the electric chamber furnace THERMOLYNE at 1000°C in the air at atmosphere pressure for 10 hour.

The elemental composition analysis and surface morphology are measured by using an energy dispersive spectroscopy and scanning electron microscope of JEOL instrument, respectively. The phase qualitative and quantitative of analysis is carried out by using the x-ray diffraction, type of PW1710 Philips with cooper anode tube. The range of diffraction angles is from 20° to 80° and step size of 0.02°. The data is analyzed by GSAS program (Rietveld code). The pseudo-Voigt function was used in the refinement of diffraction line profiles [9]. The magnetic properties are measured by VSM Merk Oxford VSM1.2H Instrument. The electric properties are measured by LCR of Hioke instrument. And the reflection and transmission of microwave were carried out using the vector network analyzer (VNA) of ADVANTEST R3770 types with frequency range of 300 kHz - 20 GHz.

3. RESULTS AND DISCUSSION

The identification results of x-ray diffraction pattern showed that the samples La$_{1-x}$Ba$_x$MnO$_3$ ($x = 0 – 0.7$) has been well established as shown in Figure 1.

![Figure 1](image1.png)

Figure 1. The phase identification of the samples La$_{1-x}$Ba$_x$MnO$_3$ ($x = 0 – 0.7$).

Figure 1 showed that the samples have been peaks formed are believed to consist of single phases for $x \leq 0.2$. However, the increasing of barium content furthermore ($x \geq 0.3$) was found foreign peaks. This means that the samples contained multi-phase And base on the XRD profile result showed that the maximum doping concentration of substitution barium into lanthanum is suspected of $x = 0.2$.

![Figure 2](image2.png)

Figure 2. The refinement result of XRD pattern on the La$_{1-x}$Ba$_x$MnO$_3$ ($x = 0.2$).
The refinement results of x-ray diffraction pattern shows that the sample La$_{1-x}$Ba$_x$MnO$_3$ (x = 0.2) is single phase with monoclinic structure, space group of I 2 / a 1, the lattice parameters of $a = 5.5182(8)$ 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$^3$ and $\rho = 6.559$ gr.cm$^{-3}$.

Morphological observations on the sample surface by scanning electron microscope showed that the sample La$_{1-x}$Ba$_x$MnO$_3$ (x = 0.2) has been well established as shown in Figure 3.

![Figure 3. Surface morphology photo of the sample La$_{1-x}$Ba$_x$MnO$_3$ (x = 0.2)](image)

The microstructure analyses showed that the particle shapes was aggregate with the particle sizes around 500 nm and distributed homogeneously on the sample surface.

Figure 4 is showed the results of the elementary analysis by using energy dispersive spectroscopy on the sample La$_{1-x}$Ba$_x$MnO$_3$ (x = 0.2).

![Figure 4. The elementary analysis of the sample La$_{1-x}$Ba$_x$MnO$_3$ (x = 0.2)](image)

The energy dispersive spectrum as in Figure 4 shows that the dominant elements are lanthanum (La), Barium (Ba), Manganese (Mn) and oxygen (O) at 4.648 keV (L$_\alpha$), 4.464 keV (K$_\alpha$), 5.894 keV (K$_\alpha$) and 0.525 keV (K$_\alpha$), respectively. The detail content of the sample are showed in Table 1.

<table>
<thead>
<tr>
<th>No.</th>
<th>Unsure</th>
<th>Content (wt.%)</th>
</tr>
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<tbody>
<tr>
<td>1.</td>
<td>Lanthanum (La)</td>
<td>48.62 ± 0.45</td>
</tr>
<tr>
<td>2.</td>
<td>Barium (Ba)</td>
<td>8.47 ± 0.39</td>
</tr>
<tr>
<td>3.</td>
<td>Manganese (Mn)</td>
<td>22.19 ± 0.30</td>
</tr>
<tr>
<td>4.</td>
<td>Oxygen (O)</td>
<td>20.72 ± 0.10</td>
</tr>
</tbody>
</table>

Table 1. The results of element analysis by using energy dispersive spectroscopy

Figure 5 is showed hysteresis curve of samples La$_{1-x}$Ba$_x$MnO$_3$ (x = 0 – 0.7). The structure LaMnO$_3$ (x = 0) has arranged antiferomagnetic. It is appears a linierity magnetization as magnetic field function. And the presence of barium caused the change magnetic ordering of the sample from antiferomagnetic to ferromagnetic.
This condition causes an imbalance of valence on Mn ions, as consequences will be the present the mix-valence on Mn ions. The fundamental characteristic of the mix-valence was a close linkage between electric and magnetic transfer. A key feature is the simultaneous transition of magnetic properties from antiferromagnetic to ferromagnetic. Basic theory that can explain this phenomenon was found a theory of double exchange by Zener [10]. The magnetic interaction occurred in Mn ions is very complex, namely between Mn$^{3+}$ and Mn$^{4+}$ ions by stoichiometry of La$_{1-x}$Ba$_x$[Mn$^{3+}$(1-x)Mn$^{4+}$]O$_3$. The presence of mix-valence on manganese ions can cause two kinds of magnetic interactions, namely interaction of superexchange and double exchange.

Figure 6 was shown the electrical properties of the samples La$_{1-x}$Ba$_x$MnO$_3$.

It is appears that the increasing of barium content caused the characteristic of inductance and capacitance increase up to x = 0.2 and then decrease after x > 0.2. This means that the permeability and permittivity characteristic also increase up to x = 0.2 and then decrease after x > 0.2. The electric and magnetic properties of the perovskite material is affected by the very complex interaction between charge, spin-ordering and orbital ordering of the electron spin. The presence of spin ordering in this materials will affected the electric and magnetic properties overall.

The reflection loss total of the sample is measured in the frequency range 9-15 GHz as shown in Figure 7.
Figure 7 indicated that there has been absorption of electromagnetic waves in the frequency range between 9-15 GHz with two peak frequency at 11.1 and 14.2 GHz with absorption of ~ 4.8 and 6.8 dB, respectively. The electromagnetic wave absorption were calculated reaches 55 % with a thickness of 1.5 mm. It can be concluded that the material lanthanum manganite perovskite system can be used as preliminary study development new materials for electromagnetic wave absorber.

4. CONCLUSIONS

The structural, magnetic and electrical properties of samples La$_{1-x}$Ba$_x$MnO$_3$ ($x = 0 - 0.7$) have been carried out. The refinement results of x-ray diffraction pattern showed that the sample La$_{1-x}$Ba$_x$MnO$_3$ ($x = 0.2$) is the best of composition with monoclinic structure, space group of I 1 2 / a 1, the lattice parameters of $a = 5.5182(8)$ Å, $b = 5.5442(8)$ Å and $c = 7.822(1)$ Å, $\alpha = \gamma = 90^\circ$ and $\beta = 89.63(1)^\circ$, $V = 239.68(3) \, \text{Å}^3$ and $\rho = 6.559 \, \text{g/cm}^3$. The microstructure analyses showed that the microstructures were aggregate with the particle sizes around 500 nm and distributed homogeneously on the sample surface. The microstructure analyses results showed that the sample contained lanthanum (La), Barium (Ba), Manganese (Mn) and oxygen (O) with mass fraction are 48.62, 8.47, 22.19, and 20.72 wt%, respectively. The hysteresis curve showed that the presence of barium caused the change magnetic ordering of the sample from antiferomagnetic to ferromagnetic. The electric properties showed that the sample had relatively high permeability and permittivity. The results of the electromagnetic wave absorption curve analysis by using a vector network analyzer indicated that there has been absorption of EM waves in the frequency range between 9-15 GHz with two peak frequency at 11.1 and 14.2 GHz with absorption of ~ 4.8 and 6.8 dB, respectively. The electromagnetic wave absorption is calculated reaches 55 % with a thickness of 1.5 mm. This study has successfully carried out the synthesis of La$_{1-x}$Ba$_x$MnO$_3$, with the best composition can be used as preliminary study development new materials for electromagnetic wave absorber.

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