EFFECT OF Mn-Ti IONS DOPING AND SINTERING TEMPERATURE ON PROPERTIES OF BARIUM HEXAFERRITE

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Diterima: 18 November 2013 Diperbaiki: 1 April 2014 Disetujui: 19 Mei 2014

ABSTRACT

EFFECT OF Mn-Ti IONS DOPING AND SINTERING TEMPERATURE ON PROPERTIES OF BARIUM HEXAFERRITE. Mn-Ti doped (0, 0.1, 0.4, 0.5, and 0.6 mole%) barium hexaferrite powders have been prepared from BaCO₃, Fe₂O₃, TiO₂ and MnO powder by mechanical alloying technique for 20 hours. The mixture powder were grinded and then dried at 100 °C for 24 hours, followed by calcination at 1,000 °C for 2 hours. The calcined powder was then crushed into 400 mesh (38 μm) in particle size. X-Ray Diffraction analysis was performed to determine the phase formed. The powder was mixed with 3 wt% Celina WE-518 polymer, and compressed with applied force of 5 tons. The sintering process was done at temperatures of 1100 °C and 1150 °C for 2 hours. The microstructure of sintered samples was observed by Scanning Electron Microscope - Energy Dispersive X-Ray Spectroscopy (SEM-EDS). The magnetic properties and Reflection Loss (RL) was measured by permeograph Magnetic-Physikal and Vector Network Analyzer (VNA), respectively. The results show that the remainance (B) of samples are likely to decrease with increase in % mol of Mn-Ti and the optimum coercivity (HcR) 4.42 kOe was achieved at 0.5 mole% Mn-Ti. The maximum reflection loss of 25.6 dB was obtained at 0.4 mole% Mn-Ti with sintering temperature of 1100 °C for 2 hours. Accordingly, it can be potentially used for microwave absorption application.

Keywords: Barium hexaferrite, Ion Mn-Ti, Reflection loss, Flux density, B-H curve

ABSTRAK

PENGARUH DOPING ION Mn-Ti DAN SUHU SINTERING PADA SIFAT BARIUM HEKSAFERIT. Telah dilakukan preparasi serbuk barium heksaferit dari bahan baku BaCO₃, Fe₂O₃, TiO₂ dan MnO, dengan doping Mn-Ti (0 %mol, 0.1 %mol, 0.4 %mol, 0.5 %mol dan 0.6 %mol) melalui teknik metaliurgi serbuk selama 20 jam. Campuran bulat digiling dan kemudian dikeringkan pada suhu 100 °C selama 24 jam, kemudian dioksilasi pada suhu 1100 °C yang ditanah selama 2 jam. Serbuk yang telah dikalsinasi digiling hingga kelos ayakan 400 mesh (38 μm). Analisis X-Ray Diffractionometer (XRD) dilakukan untuk menentukan fase yang terbentuk. Selanjutnya serbuk diompor dengan polimer Celina WE-518 sebanyak 3 % berat dan dikompaksi dengan gaya tekan sebesar 5 ton. Proses sintering dilakukan pada suhu 1100 °C dan 1150 °C, masing-masing ditahan selama 2 jam. Analisis mikrostruktur dari sampel tersebut diamati dengan menggunakan Scanning Electron Microscope - Energy Dispersive X-Ray Spectroscopy (SEM-EDS). Sifat magnitik dan Reflection Loss (RL) diskusi dengan permeograph Magnetic-Physikal dan Vector Network Analyzer (VNA). Dari hasil pengamatan menunjukkan bahwa sifat remanensi (B) cenderung turun dengan meningkatnya % mol Mn-Ti dan nilai limit koersitisitas (HcR) tetap pada 0.5 % mol Mn-Ti sebesar 4.42 kOe. Nilai reflection loss optimum adalah pada 0.4 % mol Mn-Ti, suhu sinter 1100 °C selama 2 jam yaitu sebesar -25.6 dB, sehingga potensial digunakan sebagai bahan penyerap gelombang mikro.

Kata kunci: Barium heksaferit, Ion Mn-Ti, Reflection loss, Kerapatan flaks, Kurva B-H
INTRODUCTION

Generally, the magnetic materials can be divided into soft and hard magnets. The hard magnetic materials called as permanent magnet, can keep its magnetization effect after magnetized. On the contrary, the magnetic effect of soft magnetic materials can only be maintained when the external magnetic field was applied [1-2]. Barium Hexaferrite (BaFe$_{12}$O$_{19}$) is a ceramic magnetic material which is popular in industries to be applied as a permanent magnet owing to their high magnetic properties, high corrosion resistant, and economically cheap. Moreover, this material has a high Curie temperature among the rare-earth based permanent magnets such as NdFeB [4-5] and also has good electrical properties, such as high permeability and resistivity [6]. The typical magnetic properties of barium hexaferrite, such as remanence (B), coercivity (Hc) and energy product (BH$_{max}$), Curie temperature (Tc), and resistivity (ρ) are 3.2 kOe, 3 kOe, and 2.5 MGOe, 450 °C, and ~10$^4$ ohm, respectively. The coercivity of hard magnetic materials usually have a value > 10 kA/m, while the soft magnetic materials are < 10 kA/m [3,7-8].

Amiri et al. analyzed and characterized the electromagnetic properties [2] of NiZn-ferrite (Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$) in frequencies of 12 GHz (X-band) as the radar absorption materials. It was found that the particle size and magnetic properties of the prepared ferrite sample showed strong dependence on the annealing temperature [9]. The effect of particle size and concentration of Cu Co$_{3-y}$Fe$_y$ (x = 0.1) hexaferrite composites on microwave-absorbing properties was also studied [10, 11]. The results showed that the particle size and chemical composition affect the susceptibility and permeability of composites. The microwave absorbing material with 10 GHz (8GHz) bandwidth for attenuation of 5 dB (10dB) can be achieved. Powder compaction has been done using a magnetic orientation press to get the anisotropic ferrite. The doping agents that were usually used for substituting the Fe in barium hexaferrite are Al, Co, Ni [12-13]. In addition, thick barium M-type hexaferrite (BaFe$_{12}$O$_{19}$) or BaM films deposited on alumina substrates are potentially used for self-biased planar microwave devices [14].

This paper discussed the effect of Mn-Ti ions doping and sintering temperature on the properties of BaFe$_{12}$O$_{19}$ materials to be applied as radar absorption material (RAM). The variations of Mn-Ti ions concentration are 0.1, 0.4, 0.5 and 0.6 % mol. The hysteresis curve, microstructure analysis and reflection loss (RL) characteristic of barium hexaferrite was carried out by using permagraph, SEM, XRD and vector network analyzer (VNA), respectively.

EXPERIMENTAL METHOD

BaCO$_3$, Fe$_2$O$_3$, and Mn-Ti ions from TiO$_2$ and MnO powder, were mixed by using ballmill with deionized water medium for 20 hours. The mixed powder was dried at the temperature of 100 °C for 24 hours, and then calcined at 1000 °C and hold for 2 hours. The XRD analysis was performed to identify the phases formed after calcinations. The calcined powder was then crushed (passed the 400 mesh/38 μm sieve), then mixed with 3 %wt celulosa WE-518 binder and compressed with applied force of 5 tons. The sintering process was carried out at temperatures of 1100 °C and 1150 °C, separately with 2 hours in holding time. The phase composition of calcined samples was analyzed by XRD. The microstructure of sintered samples was observed by SEM-EDS. The samples were then magnetized using impulse magnetizer K-series. The magnetic properties and microwave absorbing properties reflection loss (RL) were measured by permagraph Magnet-Physik and Vector Network Analysis (VNA). The flow chart of sample preparation and characterization is shown in Figure 1.

RESULT AND DISCUSSION

The X-Ray diffraction patterns of barium hexaferrite (BaFe$_{12-x}$Mn$_x$Ti$_x$O$_{19}$) material with x = 0, 0.1, 0.4, 0.5, and 0.6 mol% of Mn-Ti is shown in Figure 2. It can be determined that the calcined powders at 1000 °C with 2 hours in holding time were composed of a single phase hexagonal barium ferrite. This means that almost all of the added Mn-Ti ions were mixed in the hexagonal system of barium ferrite crystals.

The surface morphology and EDS analysis of BaFe$_{12}$O$_{19}$ sintered at 1100 °C for 2 hours is shown in Figure 3a and 3b, respectively. The SEM image shows that the particle distribution of barium hexaferrite is relatively homogeneous with a particle size < 3μm. Energy Dispersive X-Ray Spectrometry (EDS) analysis of BaFe$_{12}$O$_{19}$ was also performed to identify the composition of Ba, Fe, and O elements. The amount of each elements
Figure 2. XRD patterns of BaFe$_{12.2}$Mn$_x$Ti$_{10}$O$_{19}$ ($x = 0, 0.1, 0.4, 0.5$ and $0.6$ mole%) Mn-Ti) after calcination at 1000 °C for 2 hours

Table 1. Magnetic properties of barium hexaferrite with Mn-Ti addition

<table>
<thead>
<tr>
<th>Mn-Ti (mole%)</th>
<th>Sintered Temp. (°C)</th>
<th>Br (kOe)</th>
<th>Hc (kOe)</th>
<th>BH$_{max}$ (MGOe)</th>
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<tr>
<td>0</td>
<td>1150</td>
<td>2.13</td>
<td>2.786</td>
<td>0.64</td>
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<tr>
<td>0.1</td>
<td>1150</td>
<td>1.85</td>
<td>2.627</td>
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<tr>
<td>0.4</td>
<td>1100</td>
<td>1.76</td>
<td>2.845</td>
<td>0.70</td>
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<tr>
<td>0.5</td>
<td>1100</td>
<td>1.71</td>
<td>2.729</td>
<td>0.95</td>
</tr>
<tr>
<td>0.6</td>
<td>1100</td>
<td>1.48</td>
<td>2.536</td>
<td>0.96</td>
</tr>
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B-H curve of BaFe$_{12.2}$Mn$_x$Ti$_{10}$O$_{19}$ magnets is shown in Figure 4. The value of remanence (B$_r$), intrinsic coercivity (H$_c$), and energy product (BH$_{max}$) of barium hexaferrite with $0, 0.1, 0.4, 0.5$ and $0.6$ mole% of Mn-Ti ions addition is presented in Table 1.

According to Table 1, as the Mn-Ti ions increases, the $B_r$ were slightly decreased and the BH$_{max}$ were increased. The intrinsic coercivity of samples were in the range of 2.64 to 4.42 kOe. The typical barium hexaferrite usually has a value of $B_r=3.2$ kOe, $H_c=3$ kOe and BH$_{max}=2.5$ MGOe [2]. The value was different with the present study. This could be related to the raw material purity, particle size and fabrication process.

The reflection loss (RL) of magnetic material needs to be known for applications of Radio Detection and Ranging (RADAR). The RL show the capability of material to absorb the microwave which can be measured using Vector Network Analyzer (VNA). The measurement results of reflection loss (RL) of BaFe$_{12.2}$Mn$_x$Ti$_{10}$O$_{19}$ magnetic material at frequency of 4-10GHz are shown in Figure 5.

The VNA measurement results show that the substitution effect of Mn-Ti ions on the barium hexaferrite magnetic materials were significant. The 0.1 mole% of Mn-Ti ions doping sintered at 1150 °C for 2 hours give the maximum RL of -22.2 dB in the frequency of 9.13 Hz. For $x = 0.4$ mole% and sintering temperature of 1100 °C, RL is -25.6 dB in the frequency of 7.90 GHz. Referring to these RL values and the requirement for the absorbing material as RL < -20 dB [15], it can be seen

Figure 3. (a) SEM image and (b) SEM-EDS analysis of barium hexaferrite

Figure 4. Magnetic hysteresis curves of BaFe$_{12.2}$Mn$_x$Ti$_{10}$O$_{19}$ magnets after sintering (according to Table 1)

in barium hexaferrite are Ba = 3.07, Fe = 39.52 and O = 57.41 atom%, respectively.

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the potential used of these Mn-Ti doped barium hexaferrite as microwave absorbing materials.

CONCLUSIONS

Barium hexaferrite with Mn-Ti doping has been successfully developed for microwave absorption material applications by using mechanical alloying technique from raw materials BaCO₃, Fe₂O₃, TiO₂, and MnO. The best reflection loss (RL < -25.6 dB in the frequency of 7.90 GHz) is obtained by the addition of 0.4 mole% Mn-Ti and sintered at 1100 °C for 2 hour.

REFERENCES

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