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

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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 $BaCO_3$, Fe_2O_3, TiO_2 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 calcined 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% Celuna 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 permeagraph Magnet-Physik and Vector Network Analyzer (VNA), respectively. The results show that the remanance (B_r) of samples are likely to decrease with increase in % mol of Mn-Ti and the optimum coercivity (HCJ) 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 $BaCO_3$, Fe_2O_3, TiO_2dan MnO, dengan *doping* Mn-Ti (0 %mol, 0,1 %mol, 0,4 %mol, 0,5 %mol dan 0,6 %mol) melalui teknik metalurgi serbuk selama 20 jam. Campuran bubuk digiling dan kemudian dikeringkan pada suhu 100 °C selama 24 jam, kemudian dikalsinasi pada suhu 1000 °C yang ditahan selama 2 jam. Serbuk yang telah dikalsinasi digiling hingga lolos ayakan 400 *mesh* (38 πm). Analisis *X-Ray Diffractometer* (*XRD*) dilakukan untuk menentukan fasa yang terbentuk. Selanjutnya serbuk dicampur dengan polimer Celuna 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 magnetik dan *Reflection Loss* (*RL*) diukur dengan *permeagraph* (Magnet-Physik) dan *Vector Network Analyzer* (*VNA*). Dari hasil pengamatan menunjukkan bahwa nilai remanansi (B_r) cenderung turun dengan meningkatnya % mol Mn-Ti dan nilai intrinsik koersifitas (HCJ) tertinggi 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 fluks, 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_r), coercivity (H_c) and energy product (BH_{max}), Curie temperature (T_c), and resistivity (ρ) are 3.2 kG, 3 kOe, and 2.5 MGOe, 450 °C, and ~10^4 ohm, respectively. The coersivity 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 of NiZn-ferrit ($Ni_{0.7}Zn_{0.3}Fe_2O_4$) in frequencies of 8-12 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 $Cu_xCo_{2-x}Y$ (x = 0,1) hexaferrite composites on microwave-absorbing properties was also studied [10, 11].The results showed that the particle size and chemical composition affected the susceptibility and permeability of composites. The microwave absorbing material with 10 GHz (8GHz) band width 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_2O_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 celuna 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.

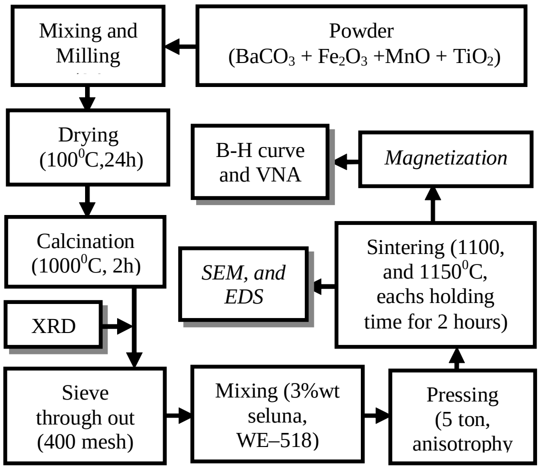

Figure 1. Flow chart of $BaFe_{12-x}(MnTi)_xO_{19}$ sampel preparation and characterization

RESULT AND DISCUSSION

The X-Ray diffraction patterns of barium hexaferrite ($BaFe_{12-2x}Mn_xTi_xO_{19}$) material with x = 0, 0.1, 0.4, 0.5 and 0.6 mole% 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 1,100 °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

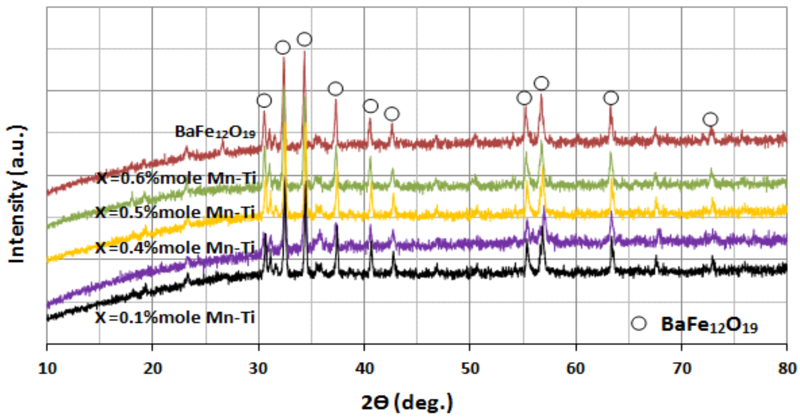

Figure 2. XRD patterns of $BaFe_{12-2x}Mn_xTi_xO_{19}$ (x = 0, 0.1, 0.4, 0.5 and 0.6 mole% Mn-Ti) after calcination at 1000 °C for 2 hours

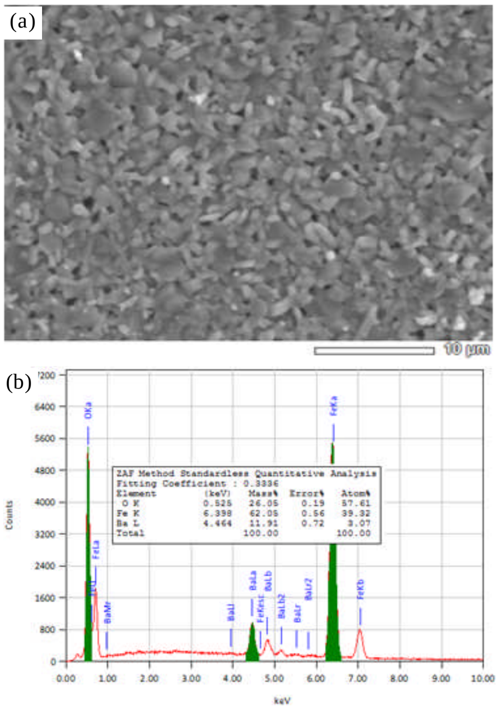

Figure 3. (a). SEI image and (b). SEM-EDS analysis of barium hexaferrite

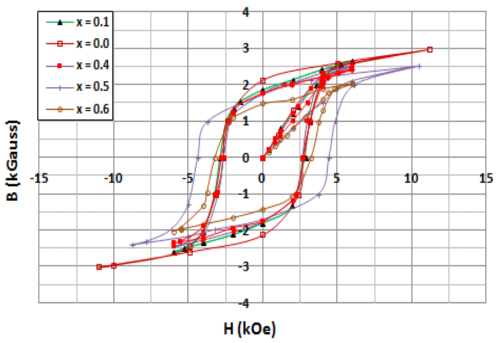

Figure 4. Magnetic hysteresis curves of $BaFe_{12-2x}Mn_xTi_xO_{19}$ magnets after sintering (according to Table 1)

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

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

Mn-Ti (%mol)	Sintered Temp. (°C)	Br (kG)	Hcj (kOe)	BHmax (MGOe)
0	1150	2.13	2.786	0.64
0.1	1150	1.85	2.627	0.65
0.4	1100	1.76	2.845	0.70
0.5	1100	1.71	2.729	0.95
0.6	1100	1.48	2.536	0.96

B-H curve of $BaFe_{(12-2x)}Mn_xTi_xO_{19}$ magnets is shown in Figure 4. The value of remanence (B_r), intrinsic coercivity (H_{CJ}), 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 kG, H_{cj} = 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 $BaFe_{12-2x}Mn_xTi_xO_{19}$ 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

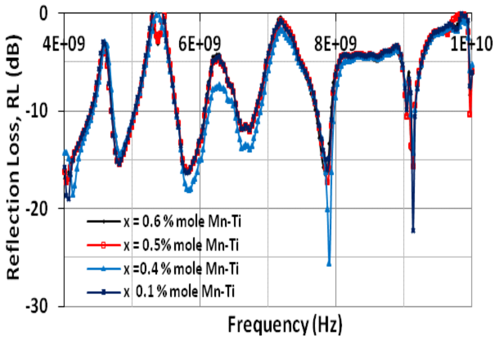

Figure 5. The reflection loss (RL) of $BaFe_{12-2x}$ $MnxTixO_{19}$ at 4-10 GHz

the potential used of these Mu-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 $BaCO_3$, Fe_2O_3, TiO_2 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.

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