International Journal of Applied Engineering Research ISSN 0973-4562 Volume 12, Number 17 (2017) pp. 6586-6590

Microstructure and Magnetic Properties of $BaFe_{12-2x}Mg_xAl_xO_{19}$ for Microwave Absorbing Materials

Martha Rianna[1], Timbangen Sembiring[1], Candra Kurniawan [2,3], Eko Arief Setiadi[2], Silviana Simbolon[2,3], Masno Ginting[2], Perdamean Sebayang[1,2,3]

[1]Physics Department Faculties of Mathematics and Natural Science, Universitas Sumatera Utara, Medan, North Sumatera-20155, Indonesia.

[2]Research Center for Physics, Indonesian Institute of Sciences (LIPI), South Tangerang, Banten, 15314, Indonesia.

[3]Department of Mechanical Engineering, Universitas Pamulang, South Tangerang, Banten- 15417, Indonesia.

Orcid ID: 0000-0002-8860-8365

Abstract

The preparation of $BaFe_{12-2x}Mg_xAl_xO_{19}$ (x = 0, 0.5, 1, and 2) of magnetic materials for microwave absorber has been done using the mechanical alloying method. $BaFe_{12-2x}Mg_xAl_xO_{19}$ material was synthesized using $BaFe_{12}O_{19}$, MgO and Al_2O_3 powders. Mixing and milling processes was held using wet High Energy Milling (HEM). Sintering process was done at 1200ºC for 2 hours. Samples were characterized using XRD, SEM-EDS, VSM and VNA. The effect of ion Mg and Al addition was not change the M-hexagonal structure of barium hexaferrite. The Mg and Al ions have been substituted Fe ion in barium hexaferrite system which is indicated by change of lattice parameters. Besides that, the substitution decreases saturation (M_s), remanence (M_r) and energy product (BH_{max}) value, but increases coercivity (H_c). Optimum condition was obtained on $BaFe_{11}Mg_{0.5}Al_{0.5}O_{19}$ with M_s = 129.0 emu/g, M_r = 69.0 emu/g, H_c = 3.90 kOe, and BH_{max} = 3.40 MGOe. Moreover, the value of the maximum reflection loss is -36.4 dB at 10.6 GHz.

INTRODUCTION

Barrium hexaferrite ($BaFe_{12}O_{19}$) have attracted more attention due to low cost, good chemical stability, high magnetic saturation, electrical resistively, and Curie temperature [21]. Barrium hexaferrite is applicable for microwave absorber in the gigahertz (GHz) range because it has frequency resonance quite wide (S-band, X-band and K-band radar) [1–4]. The magnetic properties of $BaFe_{12}O_{19}$ are affected by the substitution of ion Fe and Ba. At microwave technology, the resonance frequency of absorber ferromagnetic material can be controlled with certain level of ion substitution [5,6], such as with tetravalence, divalence of non and magnetic cation, ie: Co, Zn, Ti, Mo, Al, Ni [6,7].

Abhishek Kumar et al. had substituted Fe with Mg at BaFe12O19 using sol-gel method with molecule formula: $BaFe_{12-x}Mg_xO_{19}$ (x = 0, 0.5 and 1% mol). Reflection loss around -24.86 dB is obtained at frequency of 9.04 GHz with the concentration of x = 1% mol. This result is suitable with its application as absorption material for Radio Detection and Ranging (RADAR) [8]. While Sun Chang et al. experimented by using high energy ball milling (HEM) method and was succeeded in synthesizing $BaFe_{12}O_{19}$ by doping with MgO. Reflection loss was obtained around -41 dB at frequency of 4.27 GHz [9].

In this study, $BaFe_{12}O_{19}$-MgO-Al_2O_3 powder was mixed with mechanical alloying methods to produce a nanomagnetic $BaFe_{12-2x}Mg_xAl_xO_{19}$ (x = 0, 0.5, 1, and 2%). Additive material of MgO and Al_2O_3, were chosen because their stability of chemical behavior, abundant in nature and higher melting points, ie: 2852 and 2072˚C [10]. The purpose of using additive MgO-Al_2O_3 in $BaFe_{12}O_{19}$ is to change its magnetic properties, so that it can be used as a Radar Absorbent Materials (RAM). For microwave absorption applications, nano are better than microns size, because smaller size can be increasing of interface polarization and multiple scattering, improving the microwave absorption properties [11].

EXPERIMENTAL

The materials used to synthesize $BaFe_{12-2x}Mg_xAl_xO_{19}$ by employing the mechanical alloying method included $BaFe_{12}O_{19}$, MgO powders (Sigma-Aldrich, Germany) and Al_2O_3 (Sigma-Aldrich, Germany). The additive concentration was variated with x values about 0, 0.5, 1, and 2 % mol. Mixing and milling process were done by using wet High Energy Milling (HEM) for 3 hours in toluene medium. Ratio of milling ball and powder was 10:1. The powder was dried in oven at 100ºC for 24 hours and then shaped into pellet (diameter = 17.5 mm and thickness = 3 mm) using automatic axial hydraulic press at 1500 kgf/cm^2 pressure. For further crystal growth, the pellet was sintered at 1200ºC [12,13] for 2 hours, with heating/cooling speed of 10°C/minutes. Table 1 shows the composition of sample I, II, III, IV and V.

Table 1: Composition of $BaFe_{12-2x}Mg_xAl_xO_{19}$

Sample	Composition	Notes
I	$BaFe_{12}O_{19}$	Original
II	$BaFe_{12}O_{19}$	Sintered at 1200°C (2 hours)
III	$BaFe_{11}Mg_{0.5}Al_{0.5}O_{19}$	
IV	$BaFe_{10}MgAlO_{19}$	
V	$BaFe_8Mg_2Al_2O_{19}$	

Then samples were analyzed using X-Ray Diffraction (XRD), Scanning Electron Microscope (SEM), Energy Dispersive Spectroscopy (EDS), Vibrating Sample Magnetometer (VSM) and Vector Network Analyzer (VNA).

RESULTS AND DISCUSSION

XRD analysis of the powders and pellets of $BaFe_{12-2x}Mg_xAl_xO_{19}$ (x = 0, 0.5, 1, and 2) is depicted at Figure 1(a). It can be seen that all samples have single phase barium hexaferrite ($BaFe_{12}O_{19}$). From Figure 1(b), peak shift occurs at (114) plane to the smaller 2θ angle after sample was sintered. The differences of peak shift affect lattice parameter as shown in Table 2. On other hand, the sintering process causes the increasing of intensity, but decreases the FWHM value. It shows that sintering process really influences the grain growth of barium hexaferrite crystals [14–16], and increases the crystal diameter from 34.3 to 40 nm.

However, the lattice parameter values and crystal diameter decrease with increase additive concentration. The result indicated that Mg and Al ions substituted Fe ions. While, substitution of ions Mg and Al increase lattice strain of $BaFe_{12-2x}Mg_xAl_xO_{19}$, because of radius differences of ion Mg (0.72 Å) and Al (0.53 Å). The radius difference resulting lattice distortion at $BaFe_{12-2x}Mg_xAl_xO_{19}$. The additional of ion Mg and Al decreases the crystal density value, and crystal volume becomes smaller. The crystallite size remained the decreasing value of <5% Co doping to $BaFe_{12-x}O_{19}$ material [13]. While addition of Co increases the atomic diameter of $Ba_{1-x}Fe_{12}O_{19}$ [7].

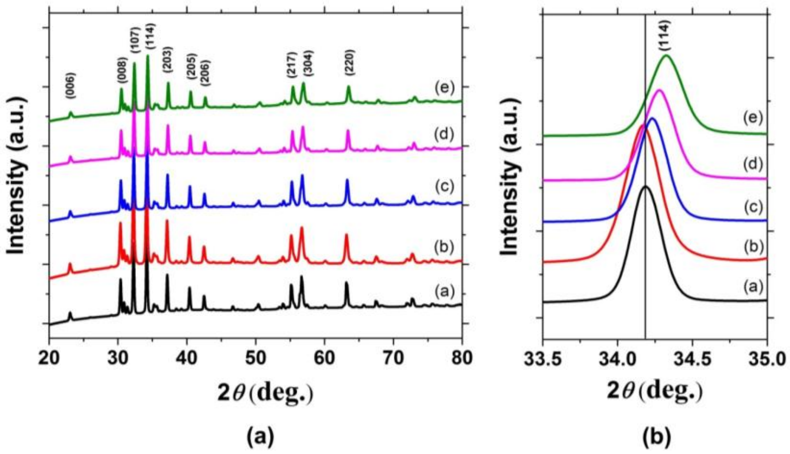
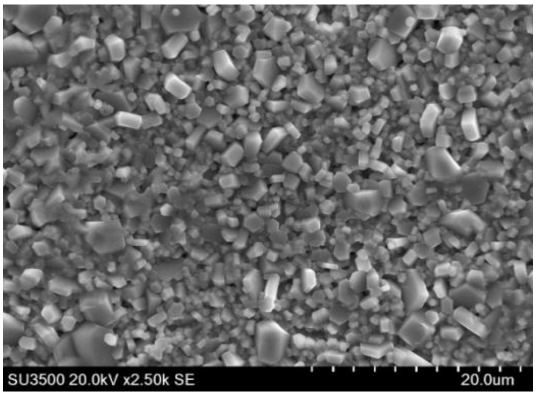


Figure 1: XRD Patterns of (a) $BaFe_{12}O_{19}$ powders (I and II) and pellet of sample $BaFe_{12-2x}Mg_xAl_xO_{19}$ (III, IV, and V), (b) Magnification of XRD pattern at (114) plane.

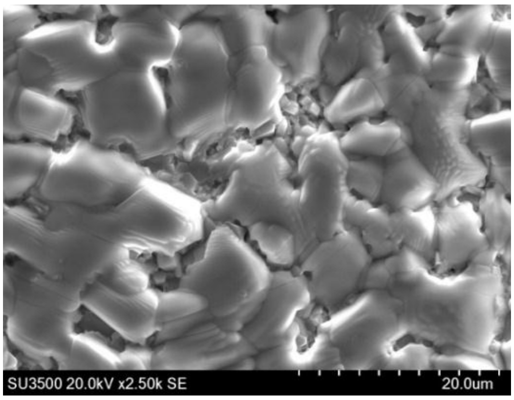
Table 2: Crystal parameters of $BaFe_{12-2x}Mg_xAl_xO_{19}$ at (114) plane for samples I, II, III, IV and V.

Sample	2θ (deg)	*FWHM* (deg)	*a* (Å)	*c* (Å)	*c/a*	*d*-spacing (Å)	*D* (nm)	Ξ (%)	ρ_{calc} (g/cm^3)
I	34.161	0.238	5.884	23.144	3.934	2.623	34.3	0.338	5.317
II	34.153	0.204	5.889	23.181	3.940	2.623	40.0	0.290	5.316
III	34.225	0.209	5.875	23.128	3.937	2.618	39.1	0.296	5.338
IV	34.262	0.241	5.865	23.094	3.938	2.615	33.9	0.341	5.363
V	34.277	0.268	5.859	23.064	3.937	2.614	30.4	0.379	5.387

SEM-EDS results for samples III and IV, it is showed in Figure 2. From Figure 2a shows that sample III has uniform particle size with average 2 µm. While from Figure 2b (sample IV) shows that the densification process occurs particle size around 4-6 µm. This is also due to the effect of the substitution of ions Mg and Al at $BaFe_{12-2x}Mg_xAl_xO_{19}$. From samples III and IV, substituted Mg and Al ions increase (shown at Table3) as proportional with additives concentrations.



(a)



(b)

Figure 2: SEM image of (a) $BaFe_{12}O_{19}$ (sample III) and (b) $BaFe_{11}Mg_{0.5}Al_{0.5}O_{19}$ (sample IV) after sintering process at 1200 °C for 2 hours.

Table 3: Other subtances found at $BaFe_{12x}Mg_xAl_xO_{19}$ (wt%)

Element	**Sample (wt%)**	
	C	**D**
O	32.59	32.60
Mg	0.01	0.10
Al	0.31	0.52
Fe	61.83	62.54
Ba	5.26	4.23

Hysteresis curve of $BaFe_{12-2x}Mg_xAl_xO_{19}$ for samples II, III, IV, and V is given in Figure 3. While magnetic behavior of $BaFe_{12-2x}Mg_xAl_xO_{19}$ is shown in Table 4.

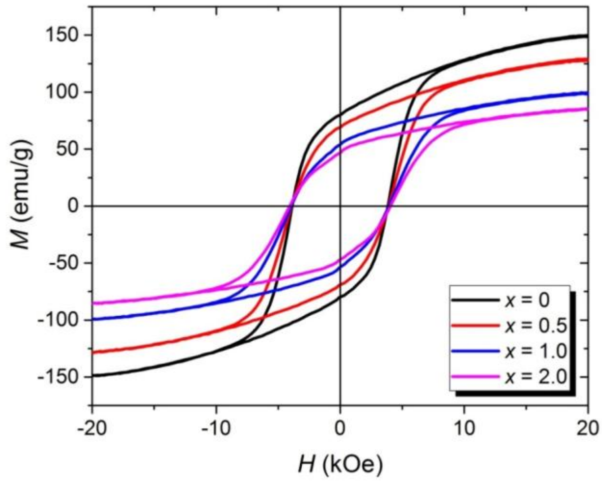


Figure 3: Hysteresis curve of $BaFe_{12-2x}Mg_xAl_xO_{19}$ for samples II, III, IV and V.

Table 4: Magnetic properties of $BaFe_{12-2x}Mg_xAl_xO_{19}$

Sample	Magnetic property			
	Ms (emu/g)	Mr (emu/g)	Hc (kOe)	BH_{max} (MGOe)
II	150.0	80.0	3.85	4.55
III	129.0	69.0	3.90	3.40
IV	99.8	53.9	3.94	2.10
V	85.5	46.7	4.06	1.66

From Figure 3, it shows that the addition of Mg and Al ions reduce magnetic behavior. The increasing of additives reduces value of Remanent (M_r), Saturation (M_s), [17], and Energy Product (BH_{max}), and increases value of Coercivity (H_c) [18]. This is due to Mg and Al which have paramagnetic behavior, while Fe is ferromagnetic. The presence of Mg and Al ions causes the changing of $BaFe_{12}O_{19}$ anisotropy constants [19], so that total of the magnetic flux decrease.

The microwave absorption results for samples II, III, IV and V is shown in Figure 4. From Figure 4, it can be seen that $BaFe_{12-2x}Mg_xAl_xO_{19}$ materials have microwave absorption for the range of 9-12 GHz. Optimum condition is obtained for sample III with optimal frequency at 10.6 GHz is -36.4 dB. Meanwhile, the other samples have no significant effect in absorbing the microwaves. For S_{11} plane, the absorption value at the position of optimum frequency of 10.6 GHz is -9.347 dB. While for S_{22} plane the absorption value at the position of optimum frequency of 10.1 GHz is -12.8 dB. As comparison, Qiu et al., the substituted Al and Cr into barium hexaferrite is obtained the absorption maximum of -34.76 dB [20]. While Shayan et al. using ion Al obtained the reflection loss of -43 dB at frequency of 15.7 GHz [21]. While, It was found that the sample of $BaMg_{0.9}Zn_{0.9}Zr_{1.8}Fe_{8.4}O_{19}$ with saturation magnetization and coercive force of 37.3 emu/g and 94 Oe respectively possessed the maximum reflection loss of -19.3 dB at 12.3 GHz [22].

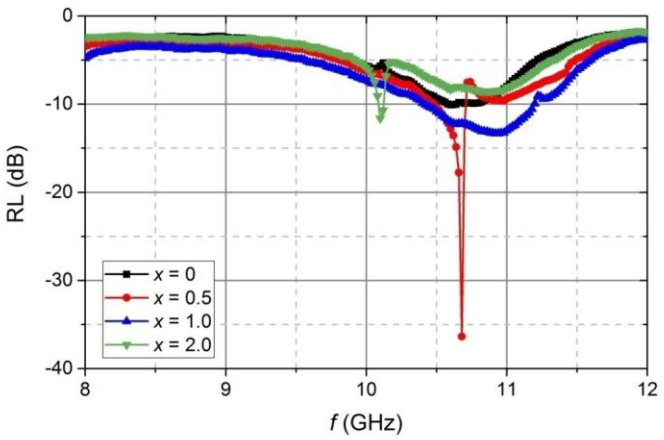


Figure 4: Reflection loss curve of $BaFe_{12-2x}Mg_xAl_xO_{19}$ for samples II, III, IV and V.

CONCLUSION

The substitution of Mg and Al ions into $BaFe_{12-2x}Mg_xAl_xO_{19}$ magnetic material was prepared by mechanical alloying method. It was followed by sintering at 1200ºC for 2 hours. The optimum condition is obtained 0.5% concentrations of Mg and Al ions (sample III) with single phase of barium hexaferrite. For this concentration, values of M_s = 129.0 emu/g, M_r = 69.0 emu/g, H_c = 3.90 kOe, and BH_{max} = 3.40 MGOe are obtained. Moreover, optimum value of the reflection loss is -36.4 dB for range of frequency about 9-12 GHz.

ACKNOWLEDGMENTS

This work is supported by Grand Master Program of Education Leading to Doctoral Degree for Excellent Graduates (Hibah PMDSU) 2015 from Directorate for Human Resource Qualifications Directorate General of Resource for Science, Technology & Higher Education Ministry of Research, Technology & Higher Education Indonesian (KEMENRISTEK-DIKTI RI), and the Research Center for Physics, Indonesian Instituted of Sciences (P2F-LIPI).

REFERENCES

[1] Choopani S, Keyhan N, Ghasemi A, Sharbati A and Alam R S 2009 Structural, magnetic and microwave absorption characteristics of $BaCo_xMn_xTi_{2x}Fe_{12-4x}O_{19}$ *Mater. Chem. Phys.* **113** 717–20 *doi:10.4028/www.scientific.net/SSP.241.93*

[2] Ghasemi A, Xiaoxi Liu and Morisako A 2009 Effect of Additional Elements on the Structural Properties, Magnetic Characteristics and Natural Resonance Frequency of Strontium Ferrite Nanoparticles/Polymer Composite *IEEE Trans. Magn.* **45** 4420–3 Ghasemi A, Xiaoxi Liu and Morisako A 2009 Effect of Additional Elements on the Structural Properties, Magnetic Characteristics and Natural Resonance Frequency of Strontium Ferrite Nanoparticles/Polymer Composite *IEEE Trans. Magn.* **45** No. 10 October 2009 4420–3

[3] Chen N, Yang K and Gu M 2010 Microwave absorption properties of La-substituted M-type strontium ferrites *J. Alloys Compd.* **490** 609–12

[4] Davoodi A and Hashemi B 2011 Magnetic properties of Sn–Mg substituted strontium hexaferrite nanoparticles synthesized via coprecipitation method *J. Alloys Compd.* **509** 5893–6 doi.org/10.1016/j.jallcom.2011.03.002

[5] Pereira F M M, Santos M R P, Sohn R S T M, Almeida J S, Medeiros A M L, Costa M M and Sombra A S B 2009 Magnetic and dielectric properties of the M-type barium strontium hexaferrite ($Ba_xSr_{1-x}Fe_{12}O_{19}$) in the RF and microwave (MW) frequency range *J. Mater. Sci. Mater. Electron.* **20** 408–17

[6] Chawla S K, Mudsainiyan R K, Meena S S and Yusuf S M 2014 Sol–gel synthesis, structural and magnetic properties of nanoscale M-type barium hexaferrites $BaCo_xZr_xFe_{(12-2x)}O_{19}$ *J. Magn. Magn. Mater.* **350** 23–9 doi: 10.1016/j.jmmm.2013.09.007

[7] Chavan V C, Shirsath S E, Mane M L, Kadam R H and More S S 2016 Transformation of hexagonal to mixed spinel crystal structure and magnetic properties of Co^{2+} substituted $BaFe_{12}O_{19}$ *J. Magn. Magn. Mater.* **398** 32–7

doi: 10.1016/j.jmmm.2015.09.002

[8] Kumar A, Agarwala V and Singh D 2012 Effect of Mg Substitution on Microwave Absorption of $BaFe_{12}O_{19}$ *Adv. Mater. Res.* **585** 62–6 doi: 10.4028/www.scientific.net/AMR.585.62

[9] Sun C, Sun K, Pang L and Liu J 2016 Preparation and Electromagnetic Properties of an Electromagnetic Wave Absorber *J. Magn.* **21** 61–4 http://dx.doi.org/10.4283/JMAG.2016.21.1.061

[10] Krämer S, Yang J, Levi C G and Johnson C A 2006 Thermochemical Interaction of Thermal Barrier

Coatings with Molten CaO-MgO-Al_2O_3-SiO_2 (CMAS) Deposits *J. Am. Ceram. Soc.* **89** 3167–75 doi**:** 10.1111/j.1551-2916.2006.01209.x

[11] Baniasadi A, Ghasemi A, Nemati A, Azami Ghadikolaei M and Paimozd E 2014 Effect of Ti–Zn substitution on structural, magnetic and microwave absorption characteristics of strontium hexaferrite *J. Alloys Compd.* **583** 325–8 https://doi.org/10.1016/j.jallcom.2013.08.188

[12] Castro W S, Corrêa R R, Paulim Filho P I, Rivas Mercury J M and Cabral A A 2015 Dielectric and magnetic characterization of barium hexaferrite ceramics *Ceram. Int.* **41** 241–6 https://doi.org/10.1016/j.ceramint.2014.08.064

[13] Shepherd P, Mallick K K and Green R J 2007 Dielectric properties of cobalt substituted M-type barium hexaferrite prepared by co-precipitation *J. Mater. Sci. Mater. Electron.* **18** 527–34 https://doi.org/10.1007/s10854-006-9059-6

[14] Naceur H, Megriche A and El Maaoui M 2014 Effect of sintering temperature on microstructure and electrical properties of $Sr_{1-x}(Na_{0.5}Bi_{0.5})_xBi_2Nb_2O_9$ solid solutions *J. Adv. Ceram.* **3** 17–30 https://doi.org/10.1007/s40145-014-0089-x

[15] Liu M, Shen X, Song F, Xiang J and Liu R 2011 Effect of heat treatment on particle growth and magnetic properties of electrospun $Sr_{0.8}La_{0.2}Zn_{0.2}Fe_{11.8}O_{19}$ nanofibers *J. Sol-Gel Sci. Technol.* **59** 553–60 https://doi.org/10.1007/s10971-011-2526-4

[16] Bezerra C W B, Zhang L, Liu H, Lee K, Marques A L B, Marques E P, Wang H and Zhang J 2007 A review of heat-treatment effects on activity and stability of PEM fuel cell catalysts for oxygen reduction reaction *J. Power Sources* **173** 891–908 https://doi.org/10.1016/j.jpowsour.2007.08.028

[17] Kim Y, Kim J and Kim S 2006 Magnetic and Microwave Absorbing Properties of Ti and Co Substituted M-hexaferrites in Ka-band Frequencies (26.5-40 GHz) (IEEE) pp 996–996 **doi:** 10.1109/INTMAG.2006.375462

[18] Yue Z, Zhong M, Ma H and Guo G 2008 Effect of Al-substitution on phase formation and magnetic properties of barium hexaferrite synthesized with sol-gel auto-combustion method *J. Shanghai Univ. Engl. Ed.* **12** 216–20

doi : https://doi.org/10.1007/s11741-008-0306-1

[19] Setiadi E A, Kurniawan C, Sebayang P and Ginting M 2017 Microstructures, physical and magnetic properties of $BaFe_{12}O_{19}$ permanent magnets with the addition of Al_2O_3-MnO *J. Phys. Conf. Ser.* **817** 012054 (http://iopscience.iop.org/1742-6596/817/1/012054)

[20] Qiu J, Wang Y and Gu M 2006 Effect of Cr substitution on microwave absorption of $BaFe_{12}O_{19}$ *Mater. Lett.* **60** 2728–32

[21] Shayan A, Abdellahi M, Shahmohammadian F, Jabbarzare S, Khandan A and Ghayour H 2017 Mechanochemically aided sintering process for the synthesis of barium ferrite: Effect of aluminum substitution on microstructure, magnetic properties and microwave absorption *J. Alloys Compd.* **708** 538–46 https://doi.org/10.1016/j.matlet.2006.01.079

[22] Afghahi S S S, Jafarian M and Atassi Y 2016 Microstructural and magnetic studies on $BaMg_xZn_xX_{2x}Fe_{12-4x}O_{19}$ (x=Zr,Ce,Sn) prepared via mechanical activation method to act as a microwave absorber in X-band *J. Magn. Magn. Mater.* **406** 184–91 https://doi.org/10.1016/j.jmmm.2016.01.020