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Synthesis and Characterization Materials M-Barium Hexaferrite Doping Ions Co-Mn Nano Particle

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Abstract. This research has been success in the synthesis of M-Barium hexaferrite (BaM) doping Co-Mn ions using coprecipitation method are expected to be applied as a base material in the coating RADAR. M-Barium hexaferrite (BaM) are $BaFe_{12-2x}Co_xMn_xO_{19}$ synthesized with various concentrations (x = 0.0, 0.1, 0.2, 0.3) and the calcinations temperature (T = 400°C, 600°C, 800°C).The materials characterization using a X-Ray Diffraction (XRD), Transmission Electron Microscope (TEM), Inductance Capacitance and resistance (LCR) meter, and Vibrating Sample Magnetometer (VSM) Instruments. The measurement results using XRD shows the material has a hexagonal crystalline structure. The measurement results using a TEM show a sample of nano crystal materials with grain diameters up to 40 nm and spacing of the crystal lattice. The measurement results using a LCR-meter shows the electric conductivity of 1.15 x 10^{-6} S/cm to BaM without doping, 3.75 x 10^{-6} S/cm to 0.1 doping concentration calcination temperature of 400 ºC, and 1,23 x 10^{-5} S/cm to 0.3 doping concentration calcination temperature of 800 ºC, thus including semiconductor materials. The magnetic properties of materials using a VSM test results show the value of coercivity of 0.1 T; remanence value of 0.06 emu/ g; and the saturation value of 0.42 emu/g. The results above show BaM Co-Mn metal doping potential as anti-radar material.

1. Introduction

The mechanism is not detecting an object by radar to be based on two aspects that is the places designed with the geometry of angular radar absorbing structure (RAS), so that the reflection of electromagnetic waves cannot be recaptured by the receiver, and the airframe is covered by absorbent material radar wave, radar absorbing materials (RAM) Barium in the form of M-Barium hexaferrite $BaFe_{12}O_{19}$ [1]. A material can absorb radar when it interacts with the electric field of the wave, so many developed materials that are magnetic. The materials that have the ability to absorb radar include M-barium hexaferrite $BaFe_{12}O_{19}$ [2], because of the nature of magnetocrystalline anisotropy is high, it is necessary to engineering material properties such as conductivity, coercivity, saturation magnetization, magnetization remanence, and the Curie temperature to fit the needs by giving Fe ion doping with other elements such as Co, Zn, Ni, Mn, Ti, and others up to a certain amount. Giving doping is not expected to cause changes in the structure of the base material $BaFe_{12}O_{19}$ considering the dimensions of the doped material and almost the same doping [3]. Research is growing rapidly with a variety of methods to obtain barium M-Barium hexaferrite (BaM) in nano size, because the particle size largely determines the magnetic characteristics of the BaM. The method used is the sol-gel, crystallization gas, ball milling, coprecipitation, aerosol, precipitation of hydrothermal and mechanical integration. The coprecipitation is one method of synthesis of inorganic compounds based on the deposition of more than one substance

together when passing through the point of saturation [4]. In this study synthesized material M-barium hexaferrite ($BaFe_{12}O_{19}$), Co-Mn ions doping using coprecipitation method with variation of dopant concentration and calcination temperature until $BaFe_{12-x}Co_xMn_xO_{19}$ generated. The characterization is performed to determine the structure, electrical properties and magnetic properties of materials $BaFe_{12-x}Co_xMn_xO_{19}$ formed.

2. Materials and Methods

$BaFe_{12-2x}Co_xMn_xO_{19}$ synthesized using coprecipitation method. The basic material used is a powdered barium carbonate ($BaCO_3$) with Mr = 197.34 g/mol, Iron (III) chloride hexahydrate ($FeCl_3.6H_2O$) with Mr =270.38 g/mol, Cobalt (II) chloride hexahydrate ($CoCl_2.6H_2O$) with Mr = 201.95 g/mol, and Manganese (II) chloride tetrahydrate ($MnCl_2.4H_2O$) with Mr = 197.96 g/mol is a product of Merck KGaA mixed with appropriate stoichiometric ratio. $BaFe_{12-2x}Co_xMn_xO_{19}$ synthesized with various concentrations (x = 0.0, 0.1, 0.2, 0.3) and the calcinations temperature (T = 400°C, 600°C, 800°C). Synthesis $BaFe_{12-2x}Co_xMn_xO_{19}$ with x = 0.3 is done in several stages, first dissolving 22.2856 g $FeCl_3.6H_2O$ in 1.552 HCl using a magnetic stirrer for 30 minutes without heating, dissolving 1.7732 g $BaCO_3$ added 0.6556 ml of HCl 37% with Mr = 12.063 using a hot plate and a magnetic stirrer at 70°C for 2 hours dissolve powder $CoCl_2.6H_2O$ 0.6414 and 0.5335 g $MnCl_2.4H2O$ in 0, 1619 ml H_2O. The second stage is the third mixed solution while stirring with a magnetic stirrer for 45 minutes. The third is dripped slowly 11.7815 ml NH_4OH with Mr = 35.06 last in the solution so that a precipitate is formed is then filtered and washed with distilled water until the normal pH (pH = 7).

The last stage in the sludge drying oven with a temperature of 80° C for 4 hours and then pulverized to form a fine powder. Samples were then characterized using XRD, TEM, LCR meters, and VSM instruments. The crystal structure of the products was characterized by X-ray Diffraction (XRD) brand Rigaku SmartLab operated at a voltage of 40 kV and current of 30 mA with a target Cu (λ 1.541862 Å) were recorded in the region of 2θ. Transmission Electron Microscope (TEM) brand Tecnai G2 SN-D6892 M operated at a voltage of 120 kV - 2000 kV was used for characterizing the size of nano particles. The electrical properties of the material was measured by using LCR-meter Yokogawa brand Hioki 3522-50. The magnetic properties of the materials measurement by vibrating sample magnetometers (VSM) OXFORD type VSM1.2H.

3. Results and Discussion

Synthesis of barium M-hexaferrite Co and Mn metal doping using dopant variations coprecipitation with x = 0.0 ; 0.1 ; 0.2 ; 0.3 and calcination temperature variations of 400, 600, and 800 ^oC. Samples with doping concentrations of 0.1 and 0.2 before calcination appear darker than the sample with a doping concentration of 0.3, this can happen because of possible formation of salt $MnCl_3$ (black) is not stable, reaction of $MnCl_2$ with hydrochloric acid (HCl) at low temperature, and will break down again at temperatures above 40 ^oC [5]**,** until after calcination 400 ^oC reddish brown color of the sample becomes brighter than the sample with a doping concentration of 0.3. Calcination gave very big influence on the synthesis process materials such as in the color of sample.

Figure 1 shows the diffraction pattern of the sample with the dopant concentration x = 3 and the calcinating temperature of 800 ^oC. Based on the results of XRD, the phase that is formed is a Manganese-Iron-Oxide hexagonal structure with identity $a = b = 5{,}04$ Å and $c = 13{,}76$ Å where $\alpha = \beta = 90^o$ and $\gamma = 120^o$; $V = 302{,}7$ A^3, in theory [6], barium M-hexaferrite own identity and where and refers to data crystallographic information file (CIF). These results indicate that the addition of the dopant concentration can regenerate identity and does not change the basic structure of barium M-hexaferrite, in accordance with the statement [7], that the addition of dopants with a small concentration not to alter the basic structure of BaM. Crystal size becomes finer with increasing calcination temperature. Components of the solid ingredients and spread evenly with statistical parameters: $\chi^2 = 1.0016$; S = 1.0008 ; $R_{wp} = 9.49$; and the position of the particle layer (d_{hkl}) constituent materials exhibit repetition with a regular pattern, indicating the material is formed consisting of layers of the crystal, where the size and the distance between the crystal layer is viewed using TEM test equipment.

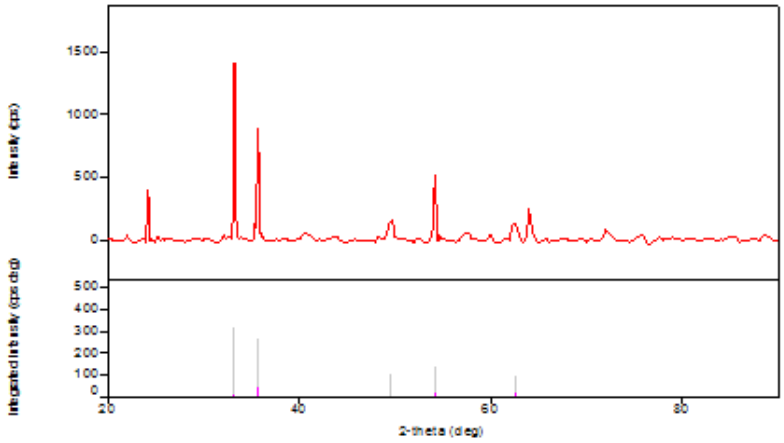

Figure 1. XRD diffraction patterns for material $BaFe_{11,4}Co_{0,3}Mn_{0,3}O_{19}$ and calcination temperature of 800 ^oC

Tests using a TEM are performed to determine the size and composition of the particles formed in the sample. The structure of the building blocks of the sample can be seen in more detail for TEM can measure up to on the order of nanoparticles. The observation using a TEM as shown in Figure 2(a,b).

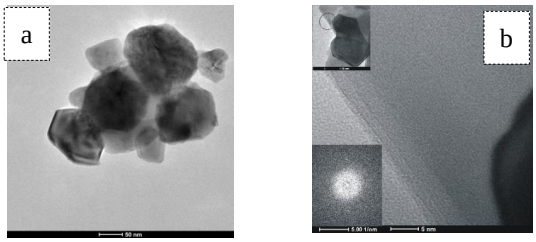

Figure 2(a,b). The observation using TEM , (a = picture crystal sizes on the order of 50 nm, b = image layer and the crystal lattice spacing on the order of 5 nm).

Figure 2a is a result of observation using a TEM with a magnification of up to 50 nm in order to determine the crystal grain size, by comparing measuring scale with the scale of the observable obtained crystal grain size vary, the diameter of the smallest and greatest diameter. In Figure 2b, the upper left corner is the result of observation of crystal layers on the order of 50 nm part circled and enlarged up to the order of 5 nm, revealing layers of crystal neat, ensuring constituent particles sampled BaM is the layers of the crystal, reinforcing the results of XRD before. Crystals formed pattern tends to elongate color to the gray- transparent, consistent with research conducted of the order of 10 nm observation [8]. The bottom left corner is the observation by the order of 5 nm to determine the distance between the crystal lattice, by comparing the measuring scale with the scale of the observable, the distance between the crystals obtained. Based on the above, the results and analysis of samples BaM in this study is a nanoparticle materials. Another study using sol-gel method to get the particle size of BaM by 38, 34, and 33 nm**,** 50-100 nm [9]**,** a method of self-propagating obtain a particle size of 40 nm [10]**.**

Table 1 shows the measurement results using a LCR meter to measure the electrical properties of materials.

Table 1. The results of testing the electrical properties of the metal doping samples BaM Co-Mn

Sample	Co-Mn 0,2 400 °C	Co-Mn 0,3 800°C	Nodoping
Diameter (cm)	1.17	1.17	1.17
Thick (cm)	0.3	0.304	0.394
$Z_{average}$ (Ω)	7.44E+04	2.30E+04	3.19E+05
A (cm^2)	1.075132	1.075132	1.075132
σ (S/cm)	3.75E-06	1.23E-05	1.15E-06

The average impedance of materials $3{,}19\ x\ 10^5\ \Omega$ (without doping); $7{,}44\ x\ 10^4\ \Omega$ ($x = 0{,}2;\ T = 400\ ^oC$), and $2{,}30\ x\ 10^4\ \Omega$ ($x = 0{,}3;\ T = 800\ ^oC$), so that the electrical conductivity of the material if it is calculated using the formula $\sigma = \frac{1}{R} = \frac{1}{Z}$, obtained as σ values in Table 1. of $1{,}15\ x\ 10^{-6}$ S/cm (without doping) ; $3{,}75\ x\ 10^{-6}$ S/cm ($x = 0{,}2;\ T = 400\ ^oC$); and $1{,}23\ x\ 10^{-5}$ S/cm ($x = 0{,}3; T = 800\ ^oC$). Based on these data, the dopant concentration and calcination temperature affect the conductivity properties of materials and samples that form a semiconductor material for conductivity value in the range of up to S/cm [11]**.** Another study by the sol-gel method to get conductivity value of 10.03 x 10^{-4} S/cm to BaM doping Mg [12], and without doping of 1 x 10^{-5} to 6 x 10^{-5} S/cm [13]**.** This research resulted in a better semiconductor material on doping concentration and calcination temperature of 800 °C ($BaFe_{11,4}Co_{0,3}Mn_{0,3}O_{19}$) with a conductivity of $1{,}23\ x\ 10^{-5}$ S/cm.

Figure 3 shows the results of measurement of magnetic properties of materials $BaFe_{11,4}Co_{0,3}Mn_{0,3}O_{19}$.

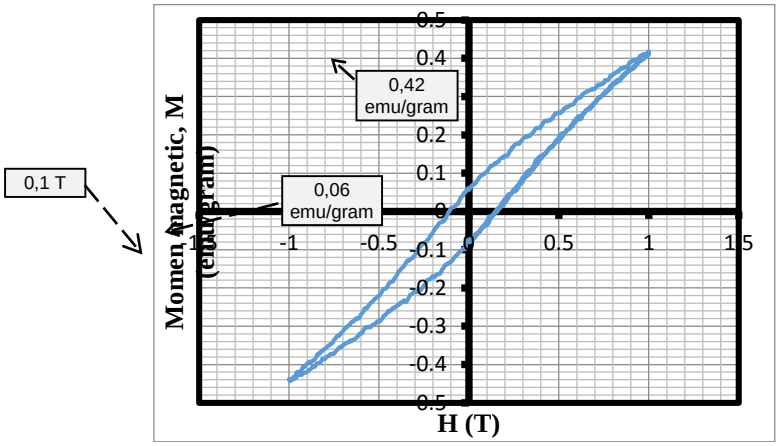

Figure 3. Hysteresis curve samples $BaFe_{11,4}Co_{0,3}Mn_{0,3}O_{19}$

The value of coercivity (Hc) of 0.1 T, saturation (Ms) of 0.42 emu/g and remanence (Mr) of 0.06 emu/g. In theory**,** Barium hexaferrite has a coercivity of 6700 Oe, Curie temperature of 450 °C, the saturation magnetization of 78 emu/g. Previous research, produce BaM coercivity value of 163 mT [14]**,** 0.32 T**,** with a solid reaction method acquire 98.22 kA/m to 27.4 Mn dopant to dopant Co [15]**,** BaM doping Mn-Ti obtain 557 Oe [16]. With the sol-gel method, getting 30.304 kA / m for doping Co-Ni and 42.472 kA/m for doping Co-Sr [20], getting 1181.07 Oe for doping La and Co, and get coercivity coefficient of 0.1104 for doping Co-Zn [17]**.** This research obtains remanence of 0.06 emu /g, the other study, by the reaction of a solid gain of 7.58 emu/g for doping Mn-Ti; through the sol-gel method acquire 29.22 and 26.12 emu/g for doping La and Co. Magnetic saturation in this study get 0.42 emu/g, more research with the sol-gel method to get Ms = 43.67 emu/g for doping Co-Zn**,** Ms = 32.07 emu/g for doping Mn-Cu-Sr, with a solid reaction method to obtain Ms = 0.35 and 0.38 Mn doping to doping Co [18]. Based on the resulting magnetic values, the BaM material doping metals Co and Mn for x = 0.3 and the calcination

temperature of 800 ºC with coprecipitation methods have successfully lowered the hard magnetic properties of BaM into a soft magnetic, so the potential to be applied as a base material absorbing microwaves.

4. Conclusion

Synthesis of BaM with Co and Mn metal doping with coprecipitation method has resulted $BaFe_{12-2x}Co_xMn_xO_{19}$ at the doping concentration x = 0.3 and the calcination temperature of 800 ºC to form $BaFe_{11,4}Co_{0,3}Mn_{0,3}O_{19}$. BaM produced is a hexagonal crystal structure of nanoparticles of materials with the identity of $a = b = 5{,}04$ Å; $c = 13{,}76$ Å and $\alpha = \beta = 90^0; \gamma = 120^0$. Electrical properties $BaFe_{11,4}Co_{0,3}Mn_{0,3}O_{19}$ in the range of a semiconductor with a conductivity value $1{,}23\ x\ 10^{-5}$ S/cm. The material of BaM to be soft magnetic field values coercivity (Hc) of 0.1 T; magnetic remanence (Mr) of 0.06 emu/g; and the value of magnetic saturation (Ms) of 0.42 emu/g. BaM produced can be applied as a base for anti- radar material.

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References

[1] Af'idah N, Indahnia E and Darminto 2011 Paper presented at the National Graduate Seminar XI – ITS Surabaya
[2] Feng Y B, Qiu T, Shen C Y 2007 *Journal of Magnetism and Magnetic Materials* ***318***
[3] Ahmeda Okashab, Kershi 2008 *Journal of Magnetism and Magnetic Materials* ***320***
[4] Susilawati, Doyan A, Taufik M, Wahyudi 2011 Research Report featured colleges, University of Mataram
[5] chem-is-try.org., (2008). (Online): http://www.chem-is-try.org/tabel periodik/kobal/, Accessed October 6, (2014)
[6] G B Teh, N Saravanan, D A Jefferson 2007 *Materials of Chemistry and Phy*sics **105** 253
[7] Kaur T and Srivastava A K 2013 *International Journal of Research in Mechanical Engineering & Technology* **3** 171-173
[8] Swamy P M P and Basavaraja S 2011 Barium ferrite nanoparticles prepared by self-propagating low-temperature combustion method and its characterization. *Indian Academy of Sciences* **34** 1319–1323.
[9] Said L M 2013 Personality Analysis of Electrical Conductivity in Some Kind of Material with Potential Methods Fall . *Journal Teknosains* **7** 66-77
[10] Chauhan C C, Jotania R B, Jotania K R 2012 *International Journal of Advanced Engineering Research and Studies.* I. 25-27
[11] Parween N 2012 Thesis. National Institute of Technology, Rourkela
[12] Hasanah E, Budi A S, Adi W A and Suguhartono I 2012 Paper presented at the National Seminar of Applied Physics III University of Airlangga, Surabaya , September 15.
[13] Sulistyo, Marhaendrajaya I and Priyono 2012 *Periodic Journal of Physics* **2** 63
[14] Priyono and Ahyani M 2010 MIPA UI .
[15] Rosler S, Wartewig P and Langbein H 2003 *Cryst. Res. Technol* **38** 927-934.
[16] Aylin G *et.al.* 2012 *Material and Techology* **46** 305
[17] Saidah I N and Zainuri M 2012 *Journal of Science and Arts of ITS* **1** 41
[18] Wisnu A A and Manaf A 2012 *Journal of Basic and Aplied Scientific Research* University of Indonesia. Jakarta.