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Electrical, Magnetic and Microwave Absorption Properties of M-type Barium Hexaferrites ($BaFe_{12-2x}Co_xNi_xO_{19}$)

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Abstract. M-type barium hexaferrites synthesis with Co-Ni doping ion ($BaFe_{12-2x}Co_xNi_xO_{19}$) based on natural iron sand of Loang Balok beach, Lombok, Indonesia, to be applied as a microwave absorbent material using co-precipitation method. The materials used in the synthesis process are magnetite minerals (Fe_2O_3 and Fe_3O_4), 12M HCl, NH_4OH 37%, $CoCl_2.6H_2O$ and $NiCl_2.6H_2O$. This research to investigate the effect of doping ion concentration variation (x = 0.0, 0.6 and 1.0) and calcination temperature (T = 80, 600, and 800°C) on electrical and magnetic properties and microwave absorption as well. The samples were characterized using Vibrating Sample Magnetometer (VSM) and Network Vector Analyzer (VNA). The result from VSM showed that the coercivity value decreased when doping ion concentration and calcination temperature increased (0.151 Tesla at 600°C for x = 0.0 and 0.044 Tesla at 800°C for x = 1.0. The value of magnetic saturation and the magnetic remanence increased with increasing ion concentration (Ms = 0.327 emu/g at x = 0.0 increased to 35.4 emu/g at x = 1.0) and Mr = 0.148 emu/g for x = 0.0 increased to 15.6 emu/g at x=1.0, this indicates that the sample has been soft magnetic. The result from VNA showed that the electrical conductivity values measured in the range 8.0-15.0 GHz indicate that the sample is a semiconductor (6.149 x 10^{-6} -5.975 x 10^{-4} S/cm). It also showed that the microwave absorption properties increased at higher concentration of doping ions and the calcination temperature would increase the value of Reflection Loss (RL). The maximum RL value of the sample is -14.47 dB at 12.38 GHz, and the absorption coefficient of 96.43%. These results indicate that the $BaFe_{12-2x}Co_xNi_xO_{19}$ sample can be applied as a microwave absorbent material on X-band to Ku-band frequency.

Keywords: Barium M-Hexaferrites, electrical and magnetic properties, natural iron sand, microwave absorber

1. Introduction

The rapid technological developments have an impact on increasing pollution of electromagnetic interferences (EMI) in the environment that can affect life, so research on microwave absorbent is an important topic. One of the materials that can be utilized as a microwave absorbent is Barium M-Hexaferrites (BaM). The ferrites (Fe) is basic material of synthesizing permanent magnet BaM, in which the iron element is obtained by separating the magnetite material from the natural iron sand. The utilization of natural iron sand is currently less optimal because it is only used as a mixture of cement. In fact, the natural iron sand contains iron oxide in the form of magnetite (Fe_3O_4), maghemit (Y-Fe_2O_3) and hematite (α- Fe_2O_3) which has the potential to be processed into various products with higher economic value. The natural iron sand used in this study came from the Loang Balok beach in Sekarbela Sub-district, Mataram City, Lombok, Indonesia. The BaM has a hexagonal molecular structure ($BaFe_{12}O_{19}$), high saturation magnetization (78 emu/g), large coercivity field (6700 Oe), high Curie temperature (450°C), chemical stability and good corrosion resistance, with magnetic and electric properties adjustable

according to the required application [1]. However, the high coercivity field causes weak absorption properties so it is less effective to use as microwaves absorbent material. To overcome this problem, we need substitution of a Fe^{3+} ion with other metal cations of similar sizes, such as: Al^{3+}, Ga^{3+}, and Cr^{3+} in order to decrease the magnetism of BaM from hard to soft magnetic [2]. A number of studies have been conducted to modify the magnetic properties of BaM such as the use of Co-Zn, Co- Mn, and Mn -Ni ions [3,4], Mn-Ti, Co-Ti, Ni-Ti, and Zn-Ti [5], As well as Co-Zn and Ni-Zn [6]. The substitution carried out in this study would use a combination of transition metal elements Co-Ni, because these two elements have radii and ionic configuration similar to Fe^{3+} (ionic radii Fe = 0.077 nm; Co = 0.072; and Ni = 0.069) was then analyzed for its effect on the electrical and magnetic properties of $BaFe_{12-2x}Co_xNi_xO_{19}$ based on natural iron sand produced to be applied as a microwave absorbent material.

2. Materials and Methods

The material used in sample synthesis was 368.937 grams of natural iron sand in magnetite compound (Fe_3O_4), $BaCO_3$ Merck KGaA 64271 Darmstadt. The solvent utilizes hydrochloric acid (HCl) 37% 12.063 M, NH_4OH solution 6.5 M and distilled water. The materials for doping Cobalt (II) Chloride Hexahydrate ($CoCl_2.6H_2O$) PA and Nickel (II) Chloride Hexahydrate ($NiCl_2.6H_2O$) PA. The synthesis process was carried out as follows. The separation of natural iron sand by using a permanent magnet (3.6 T) to separate the magnetite material from the sand. The Ferrite (Fe) content testing in a magnetic material with Atomic Absorption Microscopy (AAS) type Perkin Elmer Analyst 400 and X-Rays Fluorescent (XRF) Rigaku Supermini 200 type. The separation of Fe_3O_4 as the basic ingredient of synthesis. The sample synthesis using co-precipitation method [3]. The Vibrating Sample Magnetometer (VSM) Oxford type for its magnetic properties, the Network Vector Analyzer (VNA) Advantest R3770 in range frequency 300 kHz - 20 GHz at 220 volt and 1 ampere to measure the electrical properties and the microwave absorption.

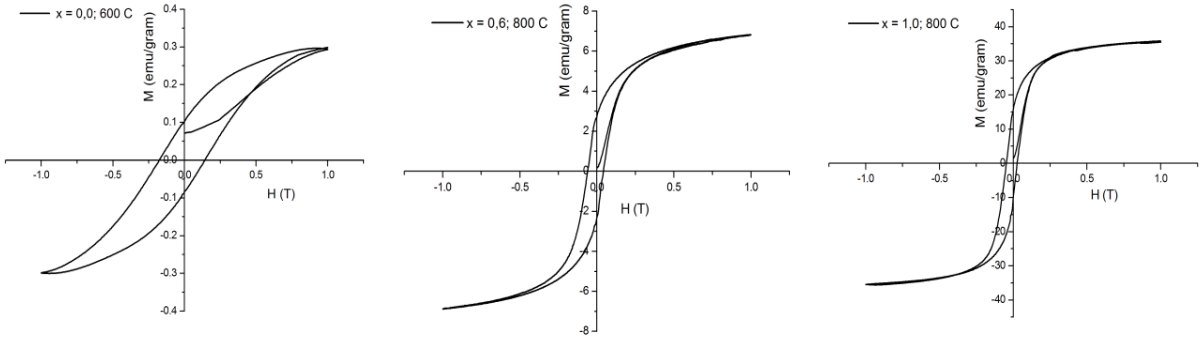
3. Results and Discussion

The magnetic material obtained from 50.033 g of sand of Loang Balok Beach after separation using permanent magnet is 46.775 g. The analyzed using AAS was obtained iron 12.816 mg/g, the result from XRF show that iron element is 62.1% weight in the Fe_2O_3 compound of 51.5% weight. These results were reinforced by XRD show that the dominant phase generated is hematite (Fe_2O_3) and magnetite (Fe_3O_4) phases corresponding to the average crystal size of 574.5967Å.

The magnetic properties of samples described in Table 1 and Figure 1. Theoretically BAM synthesized based $FeCl_3$ from the manufacturer has a value of Ms of 72 emu/ g and Hc value of 6.7 T. When compared with the results obtained have shown impaired Hc Which is very significant to 0.151 T for sample $BaFe_{12}O_{19}$ (x = 0,0); 0.060 T for samples $BaFe_{10,8}Co_{0,6}$-$Ni_{0,6}O_{19}$ (x = 0,6) and 0,044 T for sample $BaFe_{10}$ Co-NiO_{19} (x = 1,0). It can be concluded that the samples $BaFe_{12-x}Co_xNi_xO_{19}$ made from iron sand nature doped by metal Co-Ni could reduce the magnetic properties of samples from hard into a soft magnetic that can be applied as a mineral absorber of microwaves. The other studies have shown that mineral absorbing radar waves most
Preferably at low coercive value of 0.0506 Tesla at x = 0.3 [7].

Table 1. The Magnetic properties of BaM with different concentration of ion doping Co-Ni.

x	Sample	Hc (T)	Mr (emu/g)	Ms (emu/g)
0	$BaFe_{12}O_{19}$	0,151	0,148	0,327
0,6	$BaFe_{10,8}(Co\text{-}Ni)_{0,6}O_{19}$	0,060	2,84	6,79
1,0	$BaFe_{10}(Co\text{-}Ni)_{1,0}O_{19}$	0,044	15,6	35,4



(a) (b) (c)

Fig. 1. The Hysteresis loops of a). $BaFe_{12}O_{19}$ b). $BaFe_{10.8}Co\text{-}Ni_{0.6}O_{19}$ c). $BaFe_{10}Co\text{-}NiO_{19}$

The electrical properties of the sample were characterized by using VNA, the conductivity value was in the range of $2.892E^{-05}$ - 8.059 E^{-04} S/cm for sample $BaFe_{12}O_{19}$ (x = 0.0); $7.634E^{-05}$ - 8.078 E^{-04} S/cm for sample $BaFe_{10.8}Co_{0.6}\text{-}Ni_{0.6}O_{19}$ (x = 0.6); and $7.318E^{-05}$ - 6.086 E^{-04} S/cm for sample $BaFe_{10}Co\text{-}NiO_{19}$ (x = 1.0). These results indicate that the conductivity value is in the range of semiconductor properties since they are in the range of 10^{-7} - 10^{3} S/cm [8]. As a microwave absorbent, the mineral must be semiconductor because it can convert microwave into heat energy, form an electric field on an absorbent surface. The current will flow as the surface current, when the current flows into the absorber, the microwave energy will be converted in the form of heat energy [9].

Figure 2(a) showed that the permittivity (ε) and permeability (μ) values of the samples in complex numbers. The real value indicates the magnitude of the microwave energy stored in the material and the imaginary one shows the amount of microwave energy which is dissipated in the form of energy loss [10]. This combination of real and imaginary values are the determinant of the frequency of microwave absorbent material. The real permittivity value (ε') for all three samples tended to be constant at frequency (10.0-11.8) GHz and to be increased at frequency (12.0-15.0) GHz. The value of ε' increases with the increase of the doping mole fraction and the calcination temperature (from 17.8 at x = 0.0 to 20.09 at x = 1.0), but does not apply to doping x = 0.6 with the decreasing ε' value. The increase in the value of ε' is due to the transformation of Fe^{3+} ions into Fe^{2+} ions due to doping causing polarization on the sample surface [11]. A decrease in the value of ε' occurring in the sample by doping x = 0.6 because the on covering leads to the occurrence of lattice distortion thereby increasing electron scattering and resistivity [12].

Figure (2b) showed that a graph of the relationship between the imaginary permittivity (ε") of the sample at the measurement frequency range (10.0-15.0) GHz. Compared with the real permittivity value of the sample, the resulting imaginary permittivity value is lower. This means that the ability to form an electric field in the sample is higher when compared to the ability of samples in releasing electrical energy. The resulting permeability (μ) value (Figure 2b) decreases as the doping mole fraction and the calcination temperature increased. For the sample x = 0.6 it yields a real part of permeability (μ') lower than the sample x = 0.1 and results for imaginary part of permeability (μ") higher than the sample x = 0.1 compared to x = 0.6.

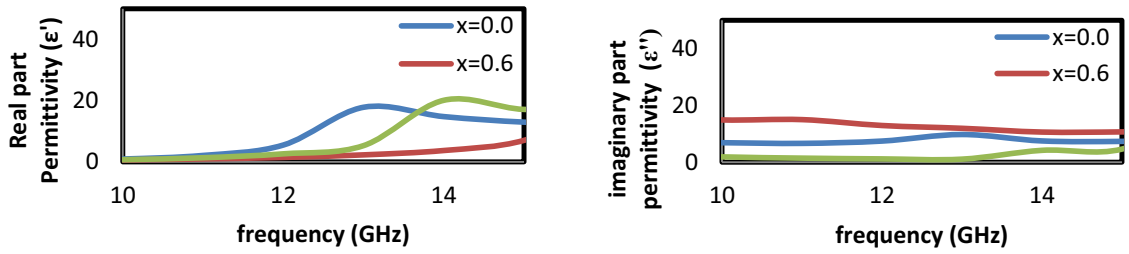


Fig.2(a). The Value of Sample Permittivity

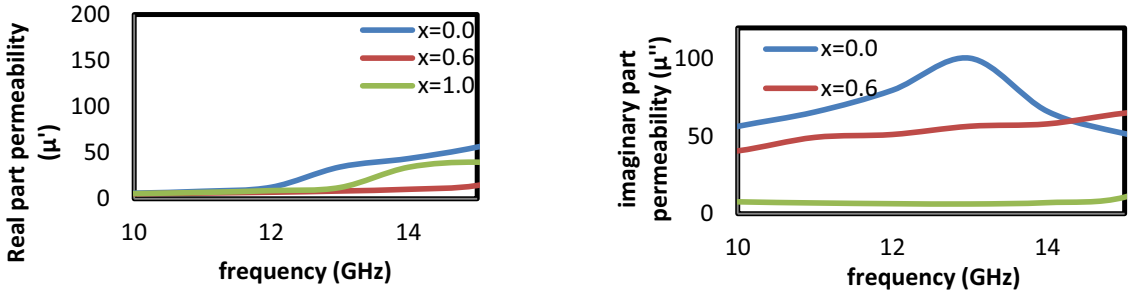


Fig. 2(b). The value of Permeability

The identification of microwave absorption properties was done through characterization using VNA resulting in data s parameters consisting of values S_{11} and S_{21} in the form of real and imaginary numbers used to determine the Reflection Loss (RL) value of the sample. The real and imaginary part of permittivity (ε) dan permeability (μ) was using equation (1) and (2)

$$\varepsilon = \frac{\lambda_o^2}{\mu}\left\{\frac{1}{\lambda_c^2} - \left[\frac{1}{2\pi L}\ln\left(\frac{1}{T}\right)\right]^2\right\} \quad (1)$$

$$\mu = \frac{1+\Gamma}{\Lambda(1-\Gamma)\sqrt{\frac{1}{\lambda_o^2}-\frac{1}{\lambda_c^2}}} \quad (2)$$

$$Z_{in} = Z_0\sqrt{\frac{\mu_r}{\varepsilon_r}}\tanh\left(i\left[\frac{2\pi L}{\lambda_0}\right]\sqrt{\mu_r\varepsilon_r}\right) \quad (3)$$

Where Z_{in} is impedance through to the sample and Z_0 is impedance free space ~ 377 Ω. The value of reflection loss (RL) was calculated using equation (4).

$$RL = 20\log\left|\frac{(Z_{in}-Z_0)}{(Z_{in}+Z_0)}\right| \quad (4)$$

Figure 3 shows the absorption intensity value (RL) in the frequency range (8.0-15.0 GHz) for each sample. The maximum RL value for the $BaFe_{10}Co\text{-}NiO_{19}$ sample at a frequency of 12.38 GHz was -14.47 dB with an absorbency coefficient of 96.43%. The RL value and absorption coefficient indicate the ability of microwave absorption. The greater the negative RL value caused the greater the absorption capacity of the material to absorbent microwave [13]. Based on the data it is found that the higher concentration of Co-Ni doping fraction and calcination temperature cause the higher RL value and the resulting absorption coefficient.

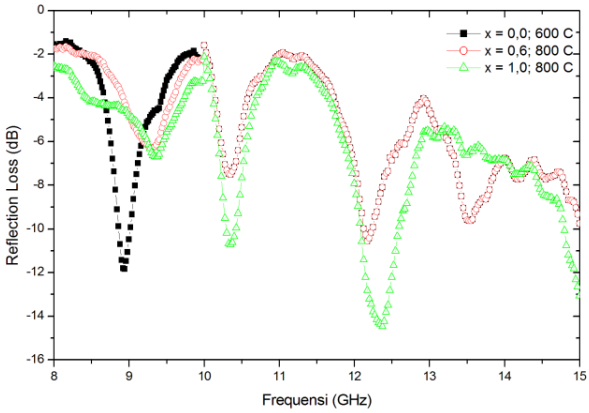


Fig. 3. The Value of Reflection Loss (RL) for Different Frequency and Concentration of Ion Doping

4. Conclusion

The $BaFe_{12-2x}Co_xNi_xO_{19}$ sample of the resulting natural iron sand has the dominant phase of Barium Iron Oxide and Hematite. The electrical properties of the sample show that the conductivity value in the range of 6.149 x 10^{-6} - 5.975 x 10^{-4} S/cm as a semiconductor. The magnetic properties of the samples indicate that soft magnetic, since the sample coercivity value has decreased significantly from 0.151 to 0.044 Tesla with an increase in the concentration of ion doping and temperature. The nature of microwave absorption yields a maximum RL value of -14.47 dB at 12.38 GHz frequency, and absorption coefficient of 96.43%. These results indicate that the $BaFe_{12-x}Co_xNi_xO_{19}$-based sample of natural iron sand can be used as a microwave absorbent material on x-band frequencies up to Ku-band.

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