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MAGNETIC PROPERTIES OF Cu^{2+} SUBSTITUTED $BaFe_{12-x}Cu_xO_{19}$ (x= 0.1,0.2,0.3,..., 4)

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ABSTRACT

Permanent magnet Barium M-Hexaferrite, $BaFe_{12-x}Cu_xO_{19}$ was synthesized with a wide range of x (x = 0.1, 0.2, …, 3 and 4mole% respectively). The precursor was made by mixing $BaCO_3$ powder and Fe_2O_3 powder, whereas CuO powder was used as additive. The powders were mixed together by wet milling in distilled water medium for 20 hours. The powders had been dried at a temperature of 100^0C for 24 h before calcined at a temperature of 1100^0C for 2 hours. The powders were then grained to prepare 400 mesh (38 µm) in diameter. The samples density was determined by using Archimedes method, magnetic properties by using permagraph, crystal structure by XRD-pattern and microwave absorbing properties using VNA. The hysteresis curve showed that the value of magnetic field ranging from 43.36 G to 100.42 G , remanence magnetic induction (Br) 20G-320G, coercivity (HcJ) 1.07 kOe to 9.22 kOe and energy production (BH) max from 0.05kGOe to 0.92 kGOe with a density from 3.82 g/cm^3 to 5.2 g/cm^3. The effect of Cu doping tent to increase the value of density with an optimum magnetic properties at x=0.3%mole.

Keywords: permanent magnet, $BaFe_{12-x}Cu_xO_{19}$, sintering, density, BH curve

ABSTRAK

Magnet permanen Barium M-Heksaferit, $BaFe_{12-x}Cu_xO_{19}$ disintesa dengan range x besar (x=0.1, 0.2, …, 3dan 4% mol). Bahan pelopor diuat dari pencampuran serbuk $BaCO_3$ dan Fe_2O_3, sementara serbuk CuO sebagai aditif. Bahan serbuk dicampur dengan miling basah dalam media air terdestilasi selama 20 jam. Serbuk dikeringkan pada 100ºC selama 24 jam sebelum dikalsinasi pada 1100ºC selama 2 jam. Serbuk kemudian diayak lolos diameter 400 mesh (38 µm). Densitas sampel ditentukan dengan metode Archimedes, sifat magnet dengan menggunakan permagraph, struktur kristal dengan pola-XRD dan sifat penyerap gelombang mikro dengan VNA. Kurva hysteresis menunjukkan nilai medan magnet dari 43,36G sampai 100,42G, Induksi magnetic remanensi (Br) dari 20G-320G, Koersivitas (HcJ) 1,07 kOe sampai 9,22 kOe dan energy produksi max (BH) dari 0,05kGOe sampai 0,92kGOe dengan nilai densitas dari 3,82 g/cm^3 sampai 5,2 g/cm^3. Penambahan Cu cenderung meningkatkan nilai densitas dengan sifat magnet optimum pada x=0,3mol persen.

Kata kunci: magnet permanen, $BaFe_{12-x}Cu_xO_{19}$, sintering, densitas, kurva BH

INTRODUCTION

Barium-Hexaferrite is a ceramic magnet and widely used in various applications, such as magnetic recording media. The raw materials of Barium Hexaferrite are relatively inexpensive and easy to manufacture as well [5]. The use in electromagnetic waves application has been extended in various fields such as telecommunications, military and civilian. The development of microwave absorbing material (microwave absorber) becomes very important nowadays [11].

Magnetic properties, especially the coercivity of permanent magnets depend on the grain size [4]. Magnetic materials with high coercivity, tend to have smaller crystallites of magnetic domains (about 1μm). For the processing of these materials, it has been developing several methods including crystallization of glass, mechano-chemical, powder metallurgy (mechanical alloying) and mechanical alloying [2]. In the mechanical alloying method, the materials are Fe_2O_3 and $BaCO_3$ (powders). This process consists of two stages of milling and annealing (ferritization). Milling process was done in the ball mill with water media in order to obtain a homogeny distribution, a more homogeneous particle size and to avoid agglomeration and adhesion [2]. Homogeneous distribution of the particles after milling is an important factor in affecting the magnetic properties after ferritization and magnetized.

The addition of metallic materials in the manufacture of a magnetic barium hexaferrite is aimed to change a soft magnetic to be a hard magnetic which has a good permeability, resistivity, and high conductivity and also low coercivity [1]. These properties are very important in terms of practical use of this magnetic as an absorber [8]. In this research we used Copper (Cu) in the form of copper oxide (CuO) as additive material, because Copper (Cu) has a high conductivity [3]. Our research was related to the manufacture of permanent magnets with barium hexaferrite by wet grinding (wet milling) with variation of Cu (in mole percent). The selection of this material because it had a good value of magnetic susceptibility and high permeability and also low coercivity, thus it can be used in absorption of microwave wave [3].

EXPERIMENT IN DETAILS

The samples were prepared by mixing raw materials: Hematit (Fe_2O_3) powder and Barium Carbonat ($BaCO_3$) with a stoichiometric reaction (1:6 mole ratio) and copper oxide powder (CuO) was used as a substitution agent. The powders were mixed by using a planetary ball mill for 24 hours. First stage was wet milling for 20 hours (ball mill) by adding 250 mL aquadest. The powders were then sieved to pass 400 mesh (38 μm) and Celuna WE - 518 at amount of 3% (by weight) was used as a polymer adhesive, the powders were then printed by pressing at 150 kgf/cm^2 (magnetic field press and held for 2 hours). We produce pellets with an outer diameter of 70 mm and 20 mm in inner diameter with 10 mm in thickness. The magnetic orientation using 1000 watts of power (AC). We prepared Barium Hexaferit ($BaFe_{12-x}Cu_xO_{19}$) with x = 0.1, 0.2, 0.3,..., 4%mol respectively. The chemical reaction can be written as follows:

$$BaCO_3 + 6Fe_2O_3 + xCuO \longrightarrow BaFe_{12-x}Cu_xO_{19} + CO_2 \quad (1)$$

The calcination process was carried at a temperature of 1100^{o}C with a rate of 3^{0}C per minute for 2 hours to produce Ferrite phase. At this phase we found the uniform grain size with a crystalline phase. The density of the samples was determined by using Archimedes' method (bulk density) and mathematically can be written as [7]:

$$\rho = \frac{M_k}{M_k - (M_b + \Delta M)} \tag{2}$$

with: ρ = bulk density (g/cm^3)
Mk = dried mass of the samples (g)
Mb = samples' mass after immersed for 24 h (g)
ΔM = Mass of the string (g)

The detail step works starting from the powder precursor and characterizations physical as well as magnetic properties is presented in below Figure 1.

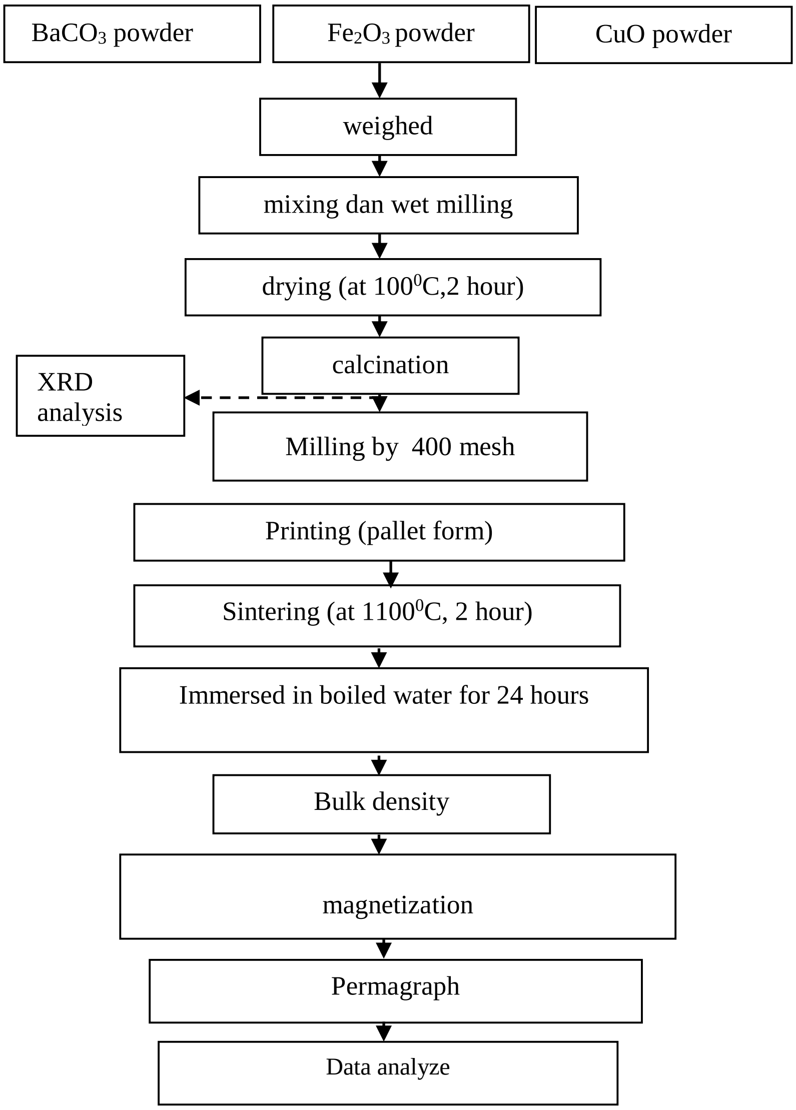

Figure 1. Flowchart of ($BaFe_{12-x}Cu_xO_{19}$) preparation and characterization

The hysteresis curves (BH curve) was obtained by using Magnet-Physics Dr. Steingroever GmbH permagraph C, whereas the flux density was determined by using a gauss meter. The reflection loss (RL) of the samples were measured in the range of 4 and 10 GHz (C-band) using Vector Network Analyzer (VNA).

RESULTS AND DISCUSSION

The permagraph result is presented in below Figure 2. From the figure, it is clear that our magnet were soft magnet. As such, our magnet are reasonable to be developed as

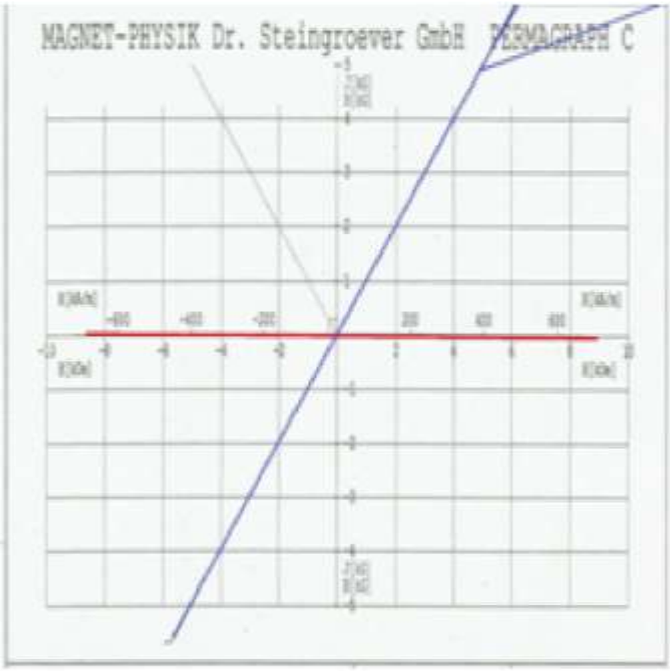

Figure 2. Permagraph result of $BaFe_{12-x}Cu_xO_{19}$ [13]

microwave absorber. To meet this point, we measured the Reflection Loss (RL) of the $BaFe_{12-x}Cu_xO_{19}$ with x=0.1 to x=0.3 as can be presented in below Figure 3. From the graph it can be seen that the optimum RL was achieved when x=0.3 with a reflection loss of -18 dB at a frequency of 5 GHz. In this experiment, we prepared a sheet of 3.57 mm in thickness, by reducing the sheet thickness we believed that an enhancement in reflection loss can be achieved [12].

Based on the measurement results of initial density (green density) in the ferrite-based magnetic (materials were not burned) ranging from 2.9 g/cm^3 to 3.9 g/cm^3 and after being burned (sintered density) is approximately 3.8 g/cm^3 to

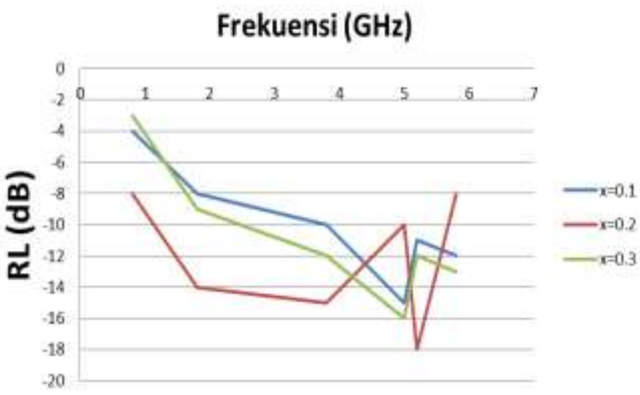

Figure 3. Reflection Loss of $BaFe_{12-x}Cu_xO_{19}$ [13]

5.2 g/cm^3. According to ChenYang Technologies [9] , the density of commercial magnet that made of Ferrite is between 2.6 g/cm^3 to 3.7 g/cm^3. Thus, our magnets are in the range of commercial magnets. The relationship between green density and sintered density is shown in Figure 3. From Figure 4, it can be seen that the value of the density tends to increase with the larger composition of CuO, it means the addition of CuO decreased the number of porous. The Cu atom filled the vacancy of the materials, hence the samples became dense. Gramatyk, P [4] reported that the density of the Soft magnetic $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ increased with the increasing of Fe content as an additive. Iqbal et al. [6] also reported that their magnet Zr-Cu with x=0-0,8 at a temperature of 780^{0}C, the density increased as x increased;

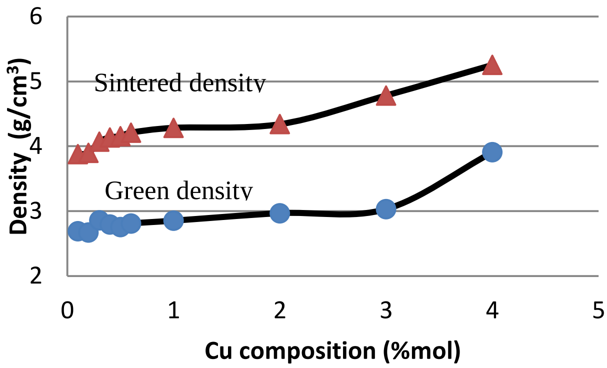

Figure 4. Relationship between density before and after sintering with Cu composition (%mol)[13].

The measurement of magnetic field strength (flux density measured) using gauss meter is shown in Figure 5. From the Figure 5, it is shown that the magnetic field strength values obtained is in the range of 43.36 gauss to 100.42 gauss, the highest value obtained when x = 0.3%mole. The value magnetic remanence, Br varies from 20kG-320 kG, with maximum remanence at x = 0.3%mole. The strongest magnetic field at x= 0.3% mole indicated that the maximum number of elementary magnetic of the materials has been achieved.

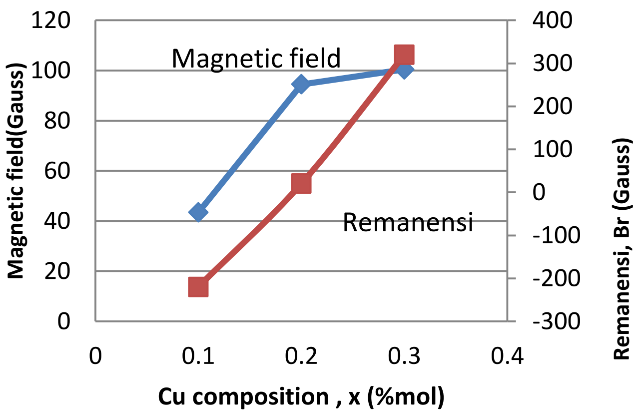

Figure 5. The relationship between magnetic field strength and remanence, Br[13].

The relationship between the maximum energy product (BHmax) and coercivity (HcJ) with addition of copper oxide (CuO) is presented in Figure 6. It can be seen that the values of BHmax obtained is in the range of 0.05kGOe to 0.9kGOe and the value of HcJ is in between

1.072 kOe and 9.22 kOe. These results also indicated that the optimum condition was achieved when x = 0.3% mole.

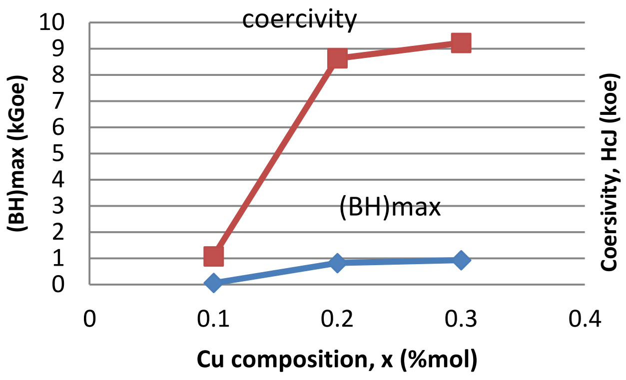

Figure 6. The relationship between (BHmax) and (HcJ) on Cu composition (%mol)[13].

The optimum magnetic values at x=0.3% mole implied that the stoichiometric reaction as well as phase stability have been occured at this composition. We could not observed the magnetic properties for x > 0.3% mole. The pattern result of X-ray diffraction (XRD) of the magnetic material hexaferrite ($BaFe_{12}O_{19}$) substituted by copper oxide (CuO) after sintering at 1100^{o}C is shown in below Figure 7.

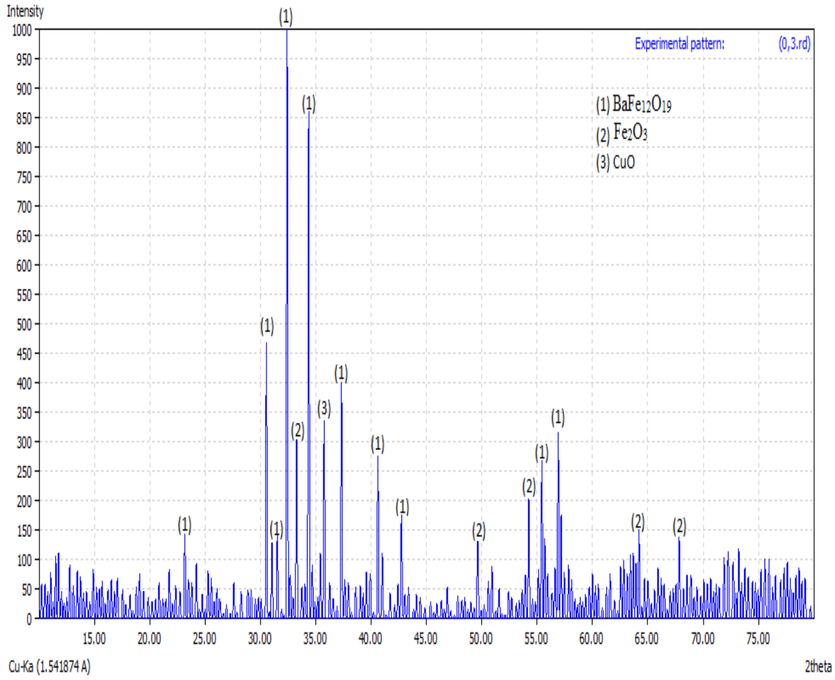

Figure 7. XRD patterns of $BaFe_{12}O_{19}$ [13].

From above XRD pattern, it can be seen that the occurence of the second phase (CuO) is around 6,25%, whereas the major phase is Ba-hexaferrite (94,75%), thus it can be noted that the sample sintered at a temperature of 1100^{o}C was formed $BaFe_{12}O_{19}$ phase as the dominant phase. It can be seen clearly that the first four maximum peaks occur at $30^{o} \leq 2\Theta \leq 40^{o}$. This implies that the characteristic peaks corresponding to the barium hexaferrite structure as a major phase. This confirms the completion of conversion the precursor powder material into $BaFe_{12}O_{19}$ [10].

CONCLUSION

We have synthesized $BaFe_{12-x}Cu_xO_{19}$ by wet milling process with an optimum condition at x = 0.3% and sintered at 1100^{0}C. The maximum flux density = 100.42 gauss, remanence = 320 gauss, coercivity, HcJ = 9.22 kOe, maximum energy product, BHmax = 0.92 kGOe. This composition can be considered as a starting composition to produce the microwave absorber material.

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