EFFECT OF FE^{3+}/BA^{2+} MOLE RATIO AND SINTERING TEMPERATURES ON THE MICROSTRUCTURE AND MAGNETIC PROPERTIES OF NANO PARTICLE BARIUM HEXAFERRITE (BAM) PRODUCED BY SOL GEL AUTO COMBUSTION

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ABSTRACT

Nanocrystalline of Barium Hexaferrite ($BaFe_{12}O_{19}$) powders have been synthesized by using the sol gel auto combustion method. The ferrite precursors were obtained from aqueous mixtures of Barium nitrate and Ferric nitrate by auto combustion reaction from gel point. These precursors were sintered at different temperatures ranging from 700 to 1000° C for constant calcinations time 2,5 h in a static air atmosphere. Effects of Fe^{3+}/Ba^{2+} mol ratios and sintering temperatures on the microstructure and magnetic properties were systematically studied. The powders formed were investigated using X-ray diffraction (XRD), scanning electron microscope (SEM) and VSM. The results obtained that the phase $BaFe_{12}O_{19}$ powders were achieved by the Fe^{3+}/Ba^{2+} mole ratio from the stoichiometric value 11, 11.5 and 12 at temperature 950° C. With increasing of temperature sintering, coercivity and magnetization value tends to rising. The maximum saturation magnetization (66.16 emu/g) was achieved at the Fe^{3+}/Ba^{2+} mole ratio to 11.5 and the sintering temperature 950° C. The maximum coercivity value 3542 Oe achieved at mole ratio sample 12 with sintering temperature 950° C. Maximum saturation 6616 emu/g achieved at mole ratio sample 115 with the same temperature.

Keywords: *barium hexaferrite, sol gel auto combustion, nanocrystalline*

INTRODUCTION

Barium Hexaferrite with hexagonal structure ($BaFe_{12}O_{19}$) has been recognized as a permanent magnetic material that has high - performance, theoretically it have large the crystalline magnetic anisotropy, high coercivity (6700 Oe), high Curie temperature (450° C), a relatively large magnetization (78 emu/g), cheese chemical stability, and corrosion resistant. In the military field Hexaferrite Barium is used as a radar absorbing material (RAM)(Hessein et al, 2007).

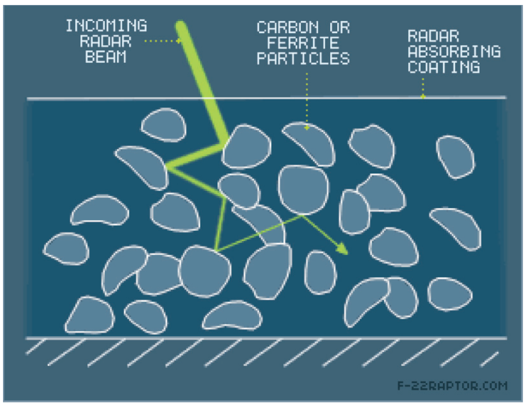

Figure 1. Energy Dissipation In Layers RAM

Topal (2007) states that the increase from 3 to 10 ratio influential to significant increase $BaFe_{12}O_{19}$ levels. In the ratio 3, phase barium monoferrite $BaFe_2O_4$ become dominant phase with less formed barium hexaferrite. When the ratio increased to 12, barium hexaferrite becomes the dominant phase. The presence of barium monoferrite no longer be found but there are impurity phases such as α-Fe_2O_3 in a number of quite a lot. Barium hexaferrite powders obtained all of the sample size was smaller than 1 μm. Mali (2006) state the influence of sintering temperature on the microstructure of barium Hexaferrite on the molar ratio of 8 with the sol gel method. This is showed that the morphology of the samples is affected by temperature. At temperatures of 900° C showed that morphology are like the burning powder. Many pores are visible at this temperature for samples with gas liberation process during sintering. Samples at temperatures of 1000° C showed spherical morphology experiencing agglomeration. On further increase in temperature of sample morphology looks like a elongated hexagonal plates with random orientation direction. Average particle size of Barium Hexaferrite is 0.4–2.5 μm.

From several studies that have been done by various methods, have been known that to obtain hexaferrite barium particles with good magnetic characterization, it must have a mole ratio of Fe and Ba are suitable for single phase. If the amount of barium that is used less, then the according to stoichiometry would produce Fe_2O_3 impurity phase due to the excessive amount of Fe, and if an excess amount of barium used, it will produce an intermediate phase $BaFe_2O_4$ (Mendoza, 2000). The more impurity phases and intermediates on $BaFe_{12}O_{19}$ powder, it becomes less good magnetic properties. Sintering temperature is more influential on the formation of barium hexaferrite and appropriate reactivity of particle size of barium hexaferrite. Sintering temperature also affect the value of coercivity (Nowosielski, 2007). In this study, using a variation of temperature 750 to 950° C to investigate the influence temperatures to single phase of Barium Hexaferrite.

METHODOLOGY

The method used in this study is the sol gel auto combustion. This method has several advantages, such as the experiments have been performed by Wang, et al. (2007) that used readily available reagents, using a mixing solution to obtain the expected chemical composition, dopants can easily be added to the end product, the shorter the time required, no special equipment needed. By using the sol gel method, we also can get the size of nano-sized powder particles are homogeneous.

Barium Nitrate and Ferrite Nitrate powder dissolved in distilled water with a ratio of mole ratio of 11, 11.5 and 12. Barium Ferrite solution and mixed and stirrer. Citric acid solution were added with the ratio of mole ratio 2:1. Solvent re-stirrer for 15 minutes. Ammonia is added by way of dripped through the pipette until the pH of the solution into 3. Thermal dehydration then performed until the solution reaches the gel point. Temperature increased until the gel expands due to auto combustion reaction. These reactions result in auto combustion powder. These Powder disinterring to temperature variations 750, 850 and 950oC. After sintering was examined using XRD, SEM/EDX, SEM (JEOL type JSM-6510LA) and VSM (Oxford VSM 1.2H).

RESULTS AND DISCUSSION

Effect of Mole Ratio on Microstructure and Magnetic Properties of Barium Hexaferrite

Figure 2. Test results show the XRD curve of sample mole ratio of 11, 11.5 and 12 with a temperature of 750° C. It is clearly seen that increasing the mole ratio of 11 to 11.5 is not resulted new phase change quite a lot. But with the addition of up to 12 mole ratio of reducing the amount of Fe_2O_3 phase and an increase in phase $BaFe_{12}O_{19}$.

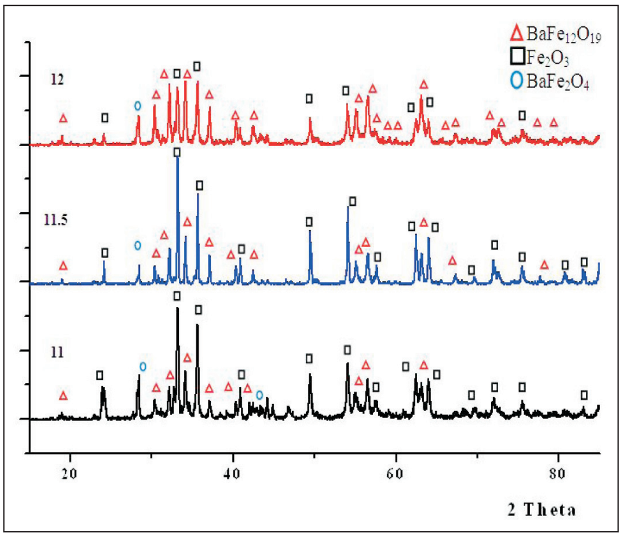

Figure 2. XRD BaM was influenced by mole ratio of Fe/Ba sintering temperature of 750° C

In contrast to previous results, from Figure 3 shows that the barium hexaferrite is the dominant phase of all the ratios at sintering temperature 950° C. Intermediate phase is still visible at 2θ of 28.46 $BaFe_2O_4$ (PDF # 46-0113) on at all mole ratios.

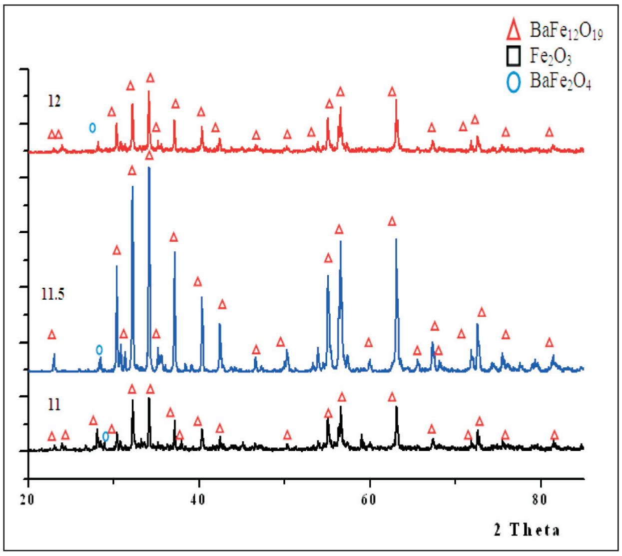

Figure 3. XRD BaM was influenced by mole ratio of Fe/Ba with sintering temperature of 950° C

Fe_2O_3 phase ending up at this temperature causes $BaFe_2O_4$ intermediate phase cannot react with Fe_2O_3 phase $BaFe_{12}O_{19}$ thus forming an intermediate phase $BaFe_2O_4$ still exist and are identified on the XRD curves.

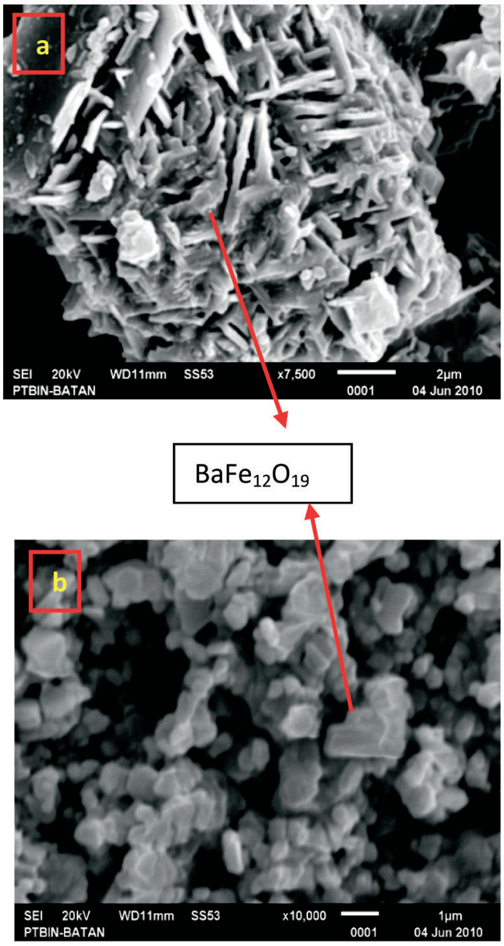

Figure 4. The morphology of the Barium Hexaferrite with a mole ratio of (a) 11.5 and (b) 12 at sintering 950° C (mag 10.000 x)

SEM examination showed that the value of different mole ratio is less affected by particle morphology. This is evidenced by the sample morphology with a mole ratio of Fe^{3+}/Ba^{2+} 11.5 and 12 have similar shapes. Form of particles with a mole ratio of 11.5 resembles the shape of elongated hexagonal plates. Whereas the mole ratio of 12 hexagonal shape resembling morphology but not elongated. Such a forms also have been reported by (Liu Junliang, 2009) and (Salemizah, 2009). After knowing Barium Hexaferrite morphology of continued testing to find out the magnetic properties of each sample. Powder with a lot of barium hexaferrite phase, homogeneous particle size and the equitable distribution would have a better magnetic properties.

Although the mole ratio of 11.5 and 12 still have an intermediate phase which has properties anti ferromagnetic $BaFe_2O_4$, but due to the existence of these phases is a minority phase, so it does not affect the shape of the hysteresis curve. The shape of the hysteresis curve width at this mole ratio. The value of magnetic saturation of the sample with a mole ratio

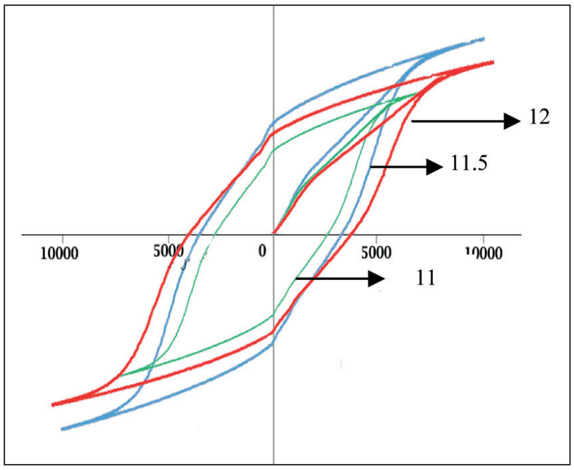

Figure 5. The hysteresis curve of the Barium Hexaferrite with mole ratio (a) 11 (b) 11.5 and (c) 12 sintering temperature of 950° C

greater than 11.5 ratio of the sample with 12 mol. But it's coersivities lower than the sample with a mole ratio of 12. The value of the magnetization hysteresis curve above show in Table 2.

Table 2. Magnetic properties of the BaM which mole ratio of 11, 11.5 and 12 (sintering temperature of 950° C).

Ratio	Sintering Temperature (° C)	Magnetic Properties	
		Hc (Oe)	Ms (emu/g)
11	950	2653	52.59
11.5	950	3234	66.16
12	950	3542	63.51

Effect of Sintering Temperature on Microstructure and Magnetic Properties of Barium Hexaferrite

Increased temperature also influential the reactivity of formation $BaFe_{12}O_{19}$ because at the time of heat treatment (sintering) took place, the metal oxide of barium and iron began to react. Increasing the sintering temperature also affects the morphology, magnetic properties of each sample. The following are the results of phase identification from XRD curves for several samples with different temperatures.

From the identification phase Figure 6 shows that BaM with a mole ratio 12 and sintering temperature 750, 850 and 950° C the dominant phase is α-Fe_2O_3 at sintering temperature 750 and 850° C. At sintering temperature 950° C dominant phase is $BaFe_{12}O_{19}$. These results indicate the formation of Barium Hexaferrite reactivity correlated with temperature in accordance with the reaction rate theory in which a process of chemical reaction rate will be increased

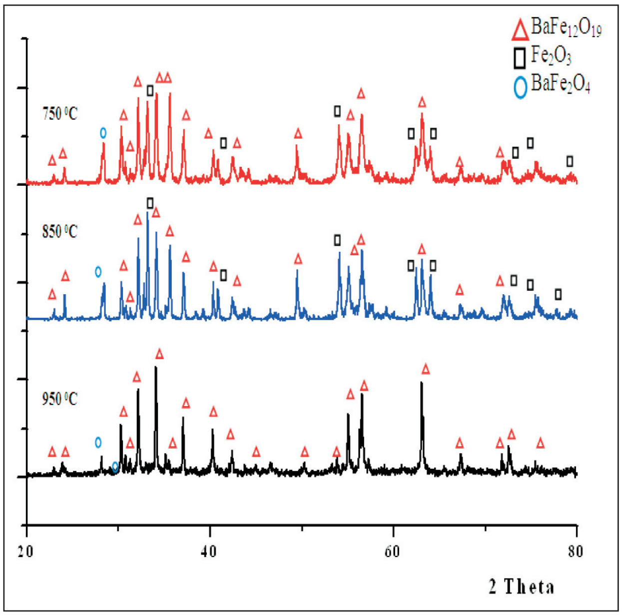

Figure 6. Effect of sintering temperature on BaM with the mole ratio of 12

along with increasing the temperature provided that the amount products increases.

SEM Tests conducted on BaM with mole ratio of 12 with sintering temperature 750, 850 and 950° C. Thus can be known whether there is a difference of particle size and morphology of the BaM as Figure 7.

At the sintering temperature 750° C, Barium Hexaferrite with mole ratio of 12 appears fine, porous or hollow with average particle size of 0.2 μm. Pores were triggered by the release of gases during the process of sintering. At temperature 850° C agglomeration spherical particles began to appear clearly. Still there are pores between particles. Cavities or pores may inhibit the diffusion rate so that the particle growth becomes obstructed. Average particle size increased up to 0.7 μm. In the increase temperature (950° C), particle shape resembles a hexagonal plate but does not extend. The average of particle size of BaM with sintering temperature 950° C is 0.5 to 1 μm. This due to the occurrence of a homogeneous nucleation in a uniform temperature with a short sintering time (Junliang, 2009). Magnetic propeties of Barium hexaferrite with mole ration 12 and variation sintering temperature show in Figure 8 and Table 3.

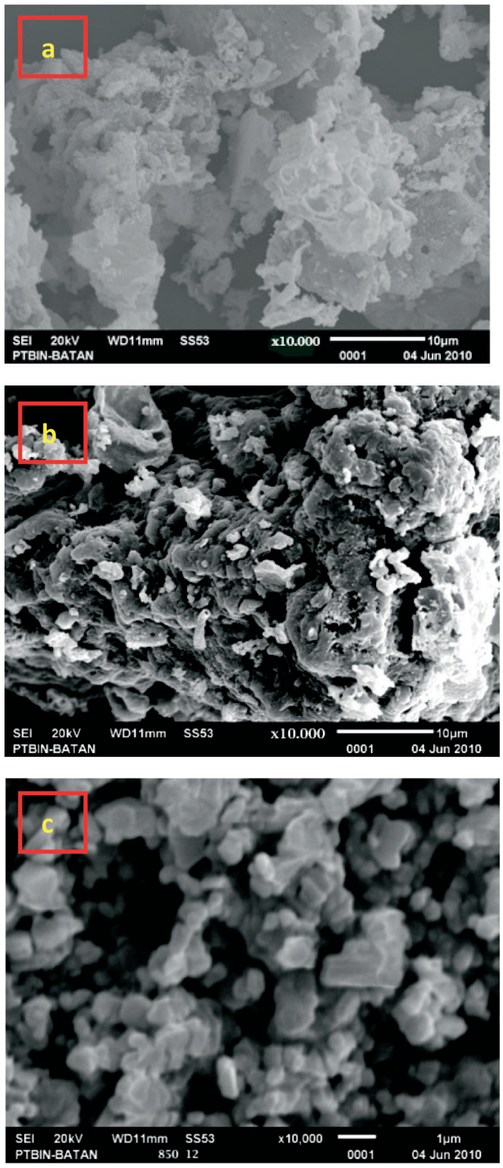

Figure 7. Effect of sintering temperature on microstructure of barium hexaferrite (mole ratio 12) with temperature sintering (a) 750 (b) 850 and (c) 950° C

Table 3. Effect of sintering temperature on magnetic properties of the Barium Hexa ferrite Mole ratio 12

Sintering Temperatur (° C)	Magnetic Properties	
	Hc (Oe)	Ms (emu/g)
750	2563	28.96
850	2612	56.76
950	3542	63.51

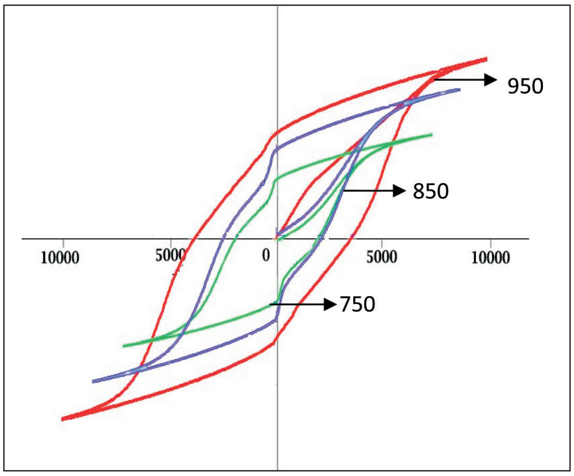

Figure 8. Effect of sintering temperature on hysteresis curve BaM with mole ratio 12

At the sintering temperature of 750 and 850° C, there are many phases hematite and monoferrite. Both these phases are antiferomagnetic. So that it can interfere with the response of magnetization when an external magnetic field imposed. Anti-parallel domains tend to be difficult to demagnetization accordance with the direction of external magnetic field. other causes of differences in crystal structure (crystalline anisotropy). Hexagonal structure of barium hexaferrite easier demagnetization at c axis direction [0001] than the other orthorhombic ($BaFe_2O_4$), demagnetization easier the other direction.

The low value of magnetization at a temperature of 750° C caused of least phase barium hexaferrite formed. Magnetic saturation significant increase from 28.96 emu/g after sintering 750° C and then slowly rises to 63.51 emu/g after sintering 950° C

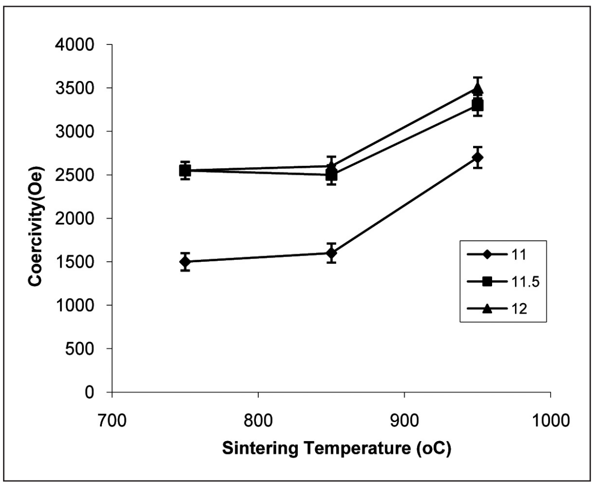

Figure 9. Effect of sintering temperature on magnetic properties of (coercivity)

For all samples, coercivity and magnetization tends to increase with increasing temperature increased. Figure 9 and 10 shows the effect of sintering temperature on magnetic properties

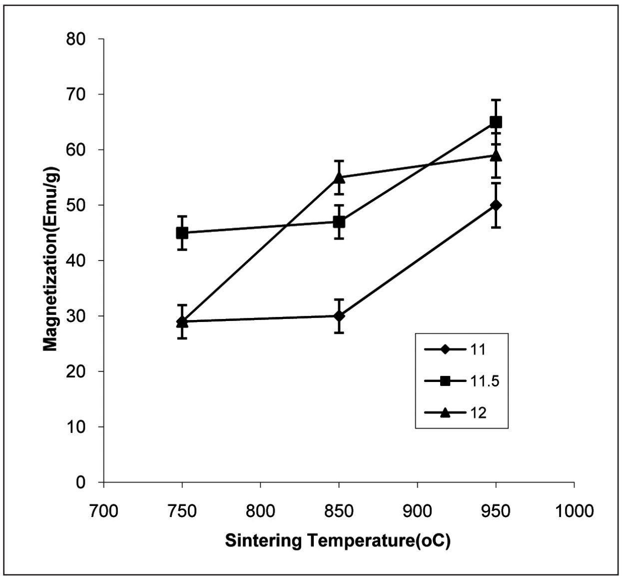

Figure 10. Effect of sintering temperature on magnetic properties (saturation)

Figure 9 and 10 shows that sintering temperature affects the value of coercivity and magnetization Barium Hexaferrite. With increasing sintering temperature up to 950° C, coercivity and magnetization Barium Hexaferrite tends to rise. These results indicate that the formation of Barium Hexaferrite phase continues to occur from 750° C temperature until it reaches the optimum point of 950° C.

CONCLUSION

Nano particles of Barium Hexaferrite successfully synthesized using the sol gel auto combustion method with a temperature of 950° C. Crystal and particle of Barium Hexaferrite increase with inreasing sintering temperature. Magnetic properties of Barium Hexaferrite increase. with increasing sintering temperature up to 950° C. At sintering temperature 950° C, the largest saturation magnetic properties (66.51emu/g) achieved at the BaM occured at mole ratio 11.5 and the largest coersivity (3542 Oe) reached at BaM mole ratio 12 with sintering temperature.

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