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SYNTHESIS AND CHARACTERIZATION OF BARIUM HEXAFERRITES DERIVED FROM STEEL WASTE BY AMMONIUM NITRATE SALT MELT SYNTHESIS

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ABSTRACT

In this paper, a series of barium hexaferrite $BaFe_{12}O_{19}$ with different powder to salt ratio (1:3, 1:4, 1:5) were prepared using ammonium salt melt technique. Iron oxide were process from steel waste product were mixed with barium carbonate, used as starting materials to produce barium hexaferrites. The ammonium nitrates in this experiment act as oxidizing agent in this synthesis. The $BaFe_{12}O_{19}$ powders were sintered at 1300°C for six hours and characterized using X-ray diffraction (XRD), Fourier transform Infrared (FTIR), Vibrating Sample Magnetometer (VSM) and Field emission Scanning Microscope (FeSEM) to investigate its crystallography, magnetic properties and morphology. The maximum coercivity and saturation magnetization obtained for sample ratio 1:3 of 1017 G and 90.9 emu/g, respectively. Increase the salt ratio decrease the coercivity and saturation magnetization values.

Steel waste or well known as mill scale is the iron oxide form on the surface of the steel during the steel making process. Steel mill factories produce an amuse amount of steel waste product every year [1-2]. Steel waste is a composed of metallic iron with different types of grease and oil contents as well as iron oxides [1, 3]**.** Mill scale are interesting industrial waste due to its high iron content about 72% [1, 4] low impurities and stable chemical composition [5]**.** It is also contains three types of iron oxides which are wuestite (FeO), hematite (α-Fe_2O_3) and magnetite (Fe_3O_4) [1, 6-7]. Barium hexaferrites $BaFe_{12}O_{19}$ with magnetoplumbite structure is one of interesting materials due to its high coercivity, high magnetic anisotropy, high saturation magnetization, excellent chemical and resistance properties [7-9]. These materials have been extensively used in various applications as magnetic recording media, magnetic tapes, permanent magnet, microwave devices and motor [7, 8, 10]. To synthesis $BaFe_{12}O_{19}$, several method such as high energy ball milling [11], sol gel [12-13], hydrothermal technique [14] and co-precipitation [15] have been developed. In this study we present ammonium nitrate salt melt technique [16]. The method is based on melting of ammonium nitrates by dissolving metal oxides to form metal nitrates. The heating of the mixture of ammonium nitrate solution and iron oxide at 260 °C is required to obtain an oxygen rich precursor which is suitable for the formation of complex oxides. Thus, this paper report the synthesis of $BaFe_{12}O_{19}$ powder using ammonium nitrate salt melt technique using different ratio of powder to salt. The effect on powder to salt ratio on magnetic properties of $BaFe_{12}O_{19}$ has been investigated. The sample has been characterized using X-ray diffraction (XRD), Field emission Scanning Microscope (FESEM) and Vibrating sample magnetometer (VSM).

The mill scales was milled for a several hours to obtain fine powder. The impurities separation and magnetic separation was conducted as follows work by R.S.Azis [1]. The magnetic and non-magnetic materials were separated using Curie separation technique. The magnetic powder was then oxidized at 500°C to obtain α-Fe_2O_3. Barium carbonate ($BaCO_3$), iron (III) oxide α-Fe_2O_3 obtained from mill and ammonium nitrate (NH_4NO_3) will be used as starting materials to synthesis pure $BaFe_{12}O_{19}$. The ammonium nitrate solution was pour in a transparent beaker and melted at 170°C. The mixture was then add the $BaCO_3$ and α-Fe_2O_3 and stirred until a thick solution was formed. The thick formed solution then heated at 260°C for one day until it produces ash like reddish precursor. The ash precursor was then grind and crush using a mortar and the powders are calcined at 1300°C in air using a high temperature box furnace. Structural and composition were investigated by using X-ray diffraction (XRD). X-ray powder diffraction data were collected in a Philips Expert PW3040 diffractometer operating at 40 kV/30 mA using Cu Kα (0.154 nm) range from 10° to 80°. Infrared spectra (280–4000 cm^{-1}) is recorded using an FTIR spectrometer (Perkin Elmer model 1650). The XRD and FTIR results were used to establish the structural

study of the prepared $BaFe_{12}O_{19}$ powder at different salt ratio. Magnetic characterization of the $BaFe_{12}O_{19}$ was performed by using a vibrating sample magnetometer (VSM) (Lake Shore 4700) at room temperature with a maximum magnetic field of 15 kOe. The particle morphology was examined by a field emission electron microscope (FESEM).

Figure 1 shows XRD spectra of the $BaFe_{12}O_{19}$ powder at different varied salts ratio 1:3, 1:4 and 1:5. Crystallinity and phase composition of the $BaFe_{12}O_{19}$ samples was investigated by XRD technique. All XRD peaks were fitted with powder-X software and indexed by JCPDS file 98-001-9939. XRD result confirms the formation of mono phase for ratio 1:3 and 1:4. For powder ratio of 1:5, shows the presence of intermediate Fe_2O_3 phase at 2θ (33.2117°). With increase salt ratio, the intensity counts of all peaks increased. The FWHM of the spectra are decreased. The possible explanation for this kind of structural modification induced by modified salt ratio can be related with change in grain size. The observed enhancement can be explained due to the formation of stable ferrite structure and effect of salt ratio induced texturing of the samples. Decreased FWHM value for increased salts ratio sample confirms the enlargement in grain size of Ba-M ferrite.

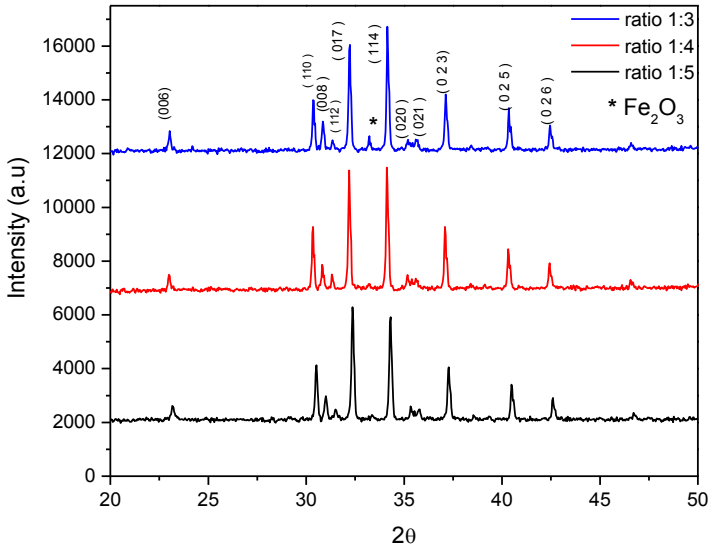

Figure 1: X-ray diffraction spectra of $BaFe_{12}O_{19}$ at different salt ratios.

Table 1: The XRD and magnetic parameters of $BaFe_{12}O_{19}$ powder.

Ratio	Position (°2θ)	FWHM (°2θ)	Intensity (counts)	*d* spacing (Å)	Crystallite size (μm)	M_r (emu/g)	M_s (emu/g)	H_c (Oe)
1:3	34.29	0.10	3199.28	2.63	2.37	43.83	90.87	1017.00
1:4	34.12	0.08	4747.01	2.62	3.40	32.57	56.18	752.02
1:5	33.21	0.09	4063.25	2.62	4.94	23.42	61.06	665.67

FTIR analysis of the precursor (Figure 2(a)) shows the absorption peak of 1413 cm^{-1} is attribute to C-O stretching, CH_2 stretching and C=O. The absorption band at 815.25 cm^{-1} [17] and 728.12 cm^{-1} are assigned to N-O bending vibration of NO_3^- and C-N stretch respectively from ammonium nitrate used in the reaction. The characteristic of absorption band of $BaFe_{12}O_{19}$ appears at 542.71 cm^{-1} and 432.48 cm^{-1} corresponding to metal-oxygen bending and vibration of octahedral sites of $BaFe_{12}O_{19}$ [18]. FTIR spectra of barium ferrite powder heated at 1300°C (Figure 2(b)) show the characteristic of absorption band of $BaFe_{12}O_{19}$ appear at 543-549 cm^{-1} which attributed to the metal oxygen stretching of the samples [17].

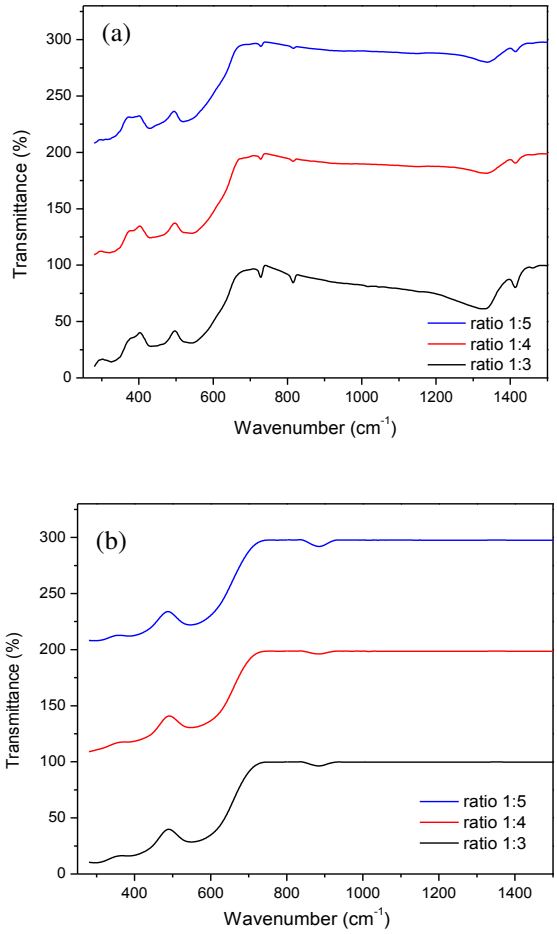

Figure 2: FTIR spectra of (a) precursor and barium ferrite (b) heated barium ferrite powder, in the range of 250–1400 cm^{-1}.

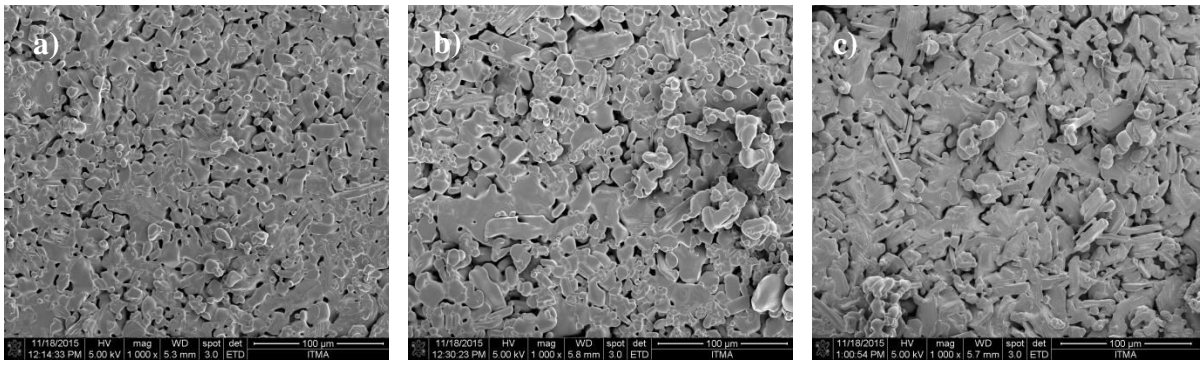

Figure 3: FESEM micrograph of samples a) ratio 1:3 b) ratio 1:4 c) ratio 1:5

Figure 3 shows FESEM micrograph of the samples heat at 1300°C at different ratio of powder to salts ratios. The average grain size of each samples are 2.5 µm, 3.4 µm and 4.6 µm for ratio of 1:3, 1:4, 1:5 respectively. The hysteresis loop of barium ferrite powder are shown in Figure 4. The M_s and H_c value show a decreased with increase the salts ratio. The maximum coercivity H_c value of 1017 Oe and saturation magnetization M_s of 90.87

emu/g for ratio 1:3 powder. This value is compared to Haneda [19] and Chin [20] that reported the coercivity H_c value of 6000 Oe and 1825 Oe. The reducing of coercivity value is due to impurities of Cr and Mn contain in the steel waste product. The coercivity H_c value decreases for salt ratio 1:4 and 1:4 to 752.02 Oe to 665.67 Oe.

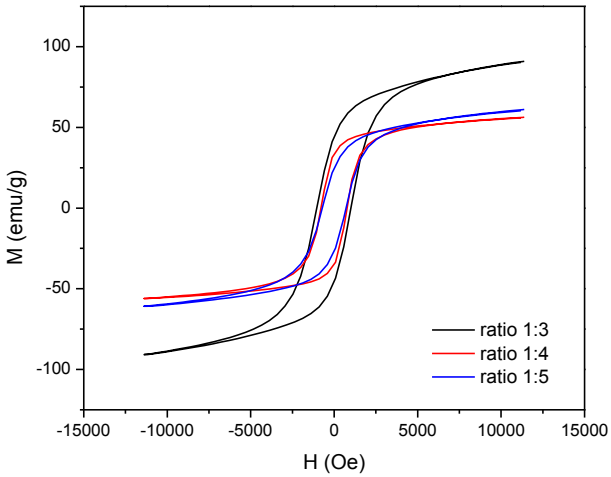

Figure 5: Hysterisis loop of samples at ratio 1:3, 1:4 and 1:5.

$BaFe_{12}O_{19}$ have been successfully prepared by ammonium nitrate salt melt technique. The study shows that as the ratio of powder to salt increases, the coercivity decrease. Thus, 1:3 ratio of powder to salt gives the maximum coercivity of 1017 Oe and saturation magnetization 90.87 emu/g. The recycled steel waste product is an alternative to produce low cost high performance ferrites, permanent magnet materials to be commercial.

REFERENCES

[1] R. S. Azis, M. Hashim, N. M. Saiden, N. Daud, and N. M. M. Shahrani, (2015). "Study the Iron Environments of the Steel Waste Product and its Possible Potential Applications in Ferrites," *Adv. Mater. Res.*, vol. 1109, pp. 295–299.

[2] M. Z. Ruhiyuddin, K. R. Ahmad, and S. S. Rizam, (2007). "Effect of Milling Periods on the Iron Mill Scale Particle Size and Properties," *Powder Metall.*, vol. 1, no. May, pp. 235–238.

[3] M. a. Legodi and D. de Waal, (2006). "The preparation of magnetite, goethite, hematite and maghemite of pigment quality from mill scale iron waste," *Dye. Pigment.*, vol. 74, no. 1, pp. 161–168.

[4] O. Benchiheub, S. Mechachti, S. Serrai, and M. G. Khalifa, (2010). "Elaboration of iron powder from mill scale," *J. Mater. Environ. Sci.*, vol. 1, no. 4, pp. 267–276.

[5] N. a. El-Hussiny, F. M. Mohamed, and M. E. H. Shalabi, (2011). "Recycling of mill scale in sintering process," *Sci. Sinter.*, vol. 43, pp. 21–31.

[6] M. I. Martín, F. a López, and J. M. Torralba, (2012). "Production of sponge iron powder by reduction of rolling mill scale," *Ironmak. Steelmak.*, vol. 39, no. 3, pp. 155–162.

[7] R. S. Azis, M. Hashim, N. Yahya, N. M. Saiden, and Z. Hari, (2002). "Effect of Milling Time on the Particles Size and Microstructure of Millscale Derived," in *Solid State Ionics*, World Scientific, pp. 575–582.

[8] P. Campbell, (1996). *Permanent magnet materials and their application*. Cambridge University Press.

[9] K. S. Martirosyan, E. Galstyan, S. M. Hossain, Y.-J. Wang, and D. Litvinov, (2011). "Barium hexaferrite nanoparticles: Synthesis and magnetic properties," *Mater. Sci. Eng. B*, vol. 176, no. 1, pp. 8–13.

[10] H.-S. Cho and S.-S. Kim, (1999). "M-hexaferrites with planar magnetic anisotropy and their application to high-frequency microwave absorbers," *Magn. IEEE Trans.*, vol. 35, no. 5, pp. 3151 – 3153.

[11] G. Sadullahoğlu, B. Ertuğ, H. Gökçe, B. Altuncevahir, M. Öztürk, R. Topkaya, N. Akdoğan, M. L. Öveçoğlu, and O. Addemir, (2015). "The Effect of Milling Time and Sintering Temperature on

Crystallization of $BaFe_{12}O_{19}$ Phase and Magnetic Properties of Ba-Hexaferrite Magnet," *Acta Phys. Pol. A*, vol. 128, no. 3, pp. 377–383.

[12] A. Mali and A. Ataie, (2004). "Influence of the metal nitrates to citric acid molar ratio on the combustion process and phase constitution of barium hexaferrite particles prepared by sol–gel combustion method," *Ceram. Int.*, vol. 30, no. 7, pp. 1979–1983.

[13] S. Kanagesan, M. Hashim, T. Kalaivani, I. Ismail, N. Rodziah, I. R. Ibrahim, and N. A. Rahman, (2015). "Sintering temperature dependence of optimized microstructure formation of BaFe12O19 using sol–gel method," *J. Mater. Sci. Mater. Electron.*, vol. 26, no. 3, pp. 1363–1367.

[14] Y. Liu, Y. Li, Y. Liu, H. S. Yin, L. L. Wang, K. Sun, and Y. Gao, (2011). "Structure Information of Barium Hexaferrite and Strategies for its Syntheses," *Appl. Mech. Mater.*, vol. 69, pp. 6–11.

[15] M. M. Rashad, M. Radwan, and M. M. Hessien, (2008). "Effect of Fe/Ba mole ratios and surface-active agents on the formation and magnetic properties of co-precipitated barium hexaferrite," *J. Alloys Compd.*, vol. 453, no. 1–2, pp. 304–308.

[16] U. Topal, H. Ozkan, and K. G. Topal, (2006). "Improved properties of BaFe12O19 prepared by ammonium nitrate melt technique and washed in HCl," *J. Alloys Compd.*, vol. 422, no. 1–2, pp. 276–278.

[17] A. Mali and A. Ataie, (2005). "Structural characterization of nano-crystalline BaFe 12 O 19 powders synthesized by sol–gel combustion route," *Scr. Mater.*, vol. 53, no. 9, pp. 1065–1070.

[18] S. Mandizadeh, F. Soofivand, M. Salavati-Niasari, and S. Bagheri, (2015). "Auto-combustion preparation and characterization of BaFe12O19 nanostructures by using maleic acid as fuel," *J. Ind. Eng. Chem.*, vol. 26, pp. 167–172.

[19] K. Haneda, (1973). "Magnetization reversal process in chemically precipitated and ordinary prepared $BaFe_{12}O_{19}$," *J. Appl. Phys.*, vol. 44, no. 8, p. 3760.

[20] T. Chin, S. L. Hsu, and M. C. Deng, (2000). "Barium ferrite particulates prepared by a salt-melt method," *J. Magn. Magn. Mater.*, vol. 120, no. 1993, pp. 64–68.