



Trade Science Inc.

# Materials Science

An Indian Journal

Full Paper

MSAIJ, 6(4), 2010 [199-207]

## Effect of Ba/Fe and annealing temperature on the synthesis and characterization of barium ferrite synthesized via oxalate precursor route

Q. Mohsen

Department of Chemistry, Materials Science Unit, Faculty of Science, Taif University, Taif, (SAUDIARABIA)

E-mail : mohsenqahtany@yahoo.com

Received: 29<sup>th</sup> April, 2010 ; Accepted: 9<sup>th</sup> May, 2010

### ABSTRACT

Barium hexaferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) is of great importance as permanent magnets, particularly for magnetic recording as well as in microwave devices. The aim of this study was to synthesize stoichiometric and single-phase barium hexaferrite through a technique of oxalate precursor. Effects of different  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio and annealing temperature on the particle size, microstructure and magnetic properties of the resulting barium hexaferrite powders has been studied, and reported in the presented paper. The annealing temperature was controlled from 900 to 1200°C, while the  $\text{Fe}^{3+}/\text{Ba}^{2+}$  was controlled from 12 to 8.57. The resultant powders were investigated by differential thermal analyzer (DTA), X-ray diffractometer (XRD), scanning electron microscopy (SEM) and vibrating sample magnetometer (VSM). Single phase of well crystalline  $\text{BaFe}_{12}\text{O}_{19}$  was first obtained at  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio of 9.23 and 8.57 at annealing temperature 1100°C. Moreover, at annealing temperature 1200°C the single phase  $\text{BaFe}_{12}\text{O}_{19}$  appeared at all different  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio. The SEM results showed that the grains were regular hexagonal platelets. In addition, maximum saturation magnetization (70.25 emu/g) was observed at mole ratio 10 and annealing temperature 1200°C. However, it was found that the coercivity of the synthesized  $\text{BaFe}_{12}\text{O}_{19}$  samples were lower than the theoretical values. © 2010 Trade Science Inc. - INDIA

### KEYWORDS

Barium hexaferrite;  
Annealing temperature;  
Mole ratio;  
Magnetic properties;  
Oxalate precursor.

### INTRODUCTION

Ferromagnetic oxides, or ferrites as they are usually known, are of great importance as high-frequency magnetic materials due to their large resistivities. Ferrites have become available as practical magnetic materials over the course of the last twenty years. Ferrite is a class of ceramic materials with useful electromagnetic properties. Ferrites play an important role in the field of electronics industry because they are

relatively inexpensive, more stable and easily manufactured. Ferrites are widely used in microwave devices, permanent magnets, high density magnetic and magneto-optic recording media, and telecommunications devices. Permanent magnet materials are essential in devices for storing energy in a static magnetic field. As a result of its specific magnetic properties barium hexaferrite and its derivatives can be used for permanent magnets, magnetic recording media and microwave applications<sup>[1]</sup>.  $\text{BaFe}_{12}\text{O}_{19}$  (M-type hex-

## Full Paper

agonal ferrites) is currently magnetic material with great scientific and technological interest, because of its relatively high Curie temperature, high coercive force and high magnetic anisotropy field as well as an excellent chemical stability and corrosion resistivity<sup>[2]</sup>. Barium hexaferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) has a complex hexagonal unit cell and belongs to the magnetoplumbite structures<sup>[3]</sup>. Magnetoplumbite are of the type  $\text{A}^{2+}\text{O}_1.6\text{B}_2^{3+}\text{O}_3$ . The arrangement of the 12  $\text{Fe}^{3+}$  ions in the unit cell is as follows: two ions in the tetrahedral sites (four nearest  $\text{O}^{2-}$  neighbors), nine ions in the dodecahedral sites (six nearest  $\text{O}^{2-}$  neighbors) and one ion in the hexagonal site (five nearest  $\text{O}^{2-}$  neighbors). Materials of this type have a strong uniaxial magnetic direction, making as permanent magnets. This type of ferrites is termed as hard ferrite due to their high coercive force, high saturation magnetization, high curie temperature and chemically inert. The reported theoretical calculated coercive force, saturation magnetization and Curie temperature values for pure and single domain barium hexaferrite was 6700 Oe, 72 emu/g and 450°C, respectively<sup>[4,5]</sup>. It is difficult to obtain ultrafine and monodispersed particles by the commercial ceramic method (solid-state reaction) which involves the firing of stoichiometric mixture of barium carbonate and  $\alpha$ -iron oxide at high temperatures (about 1200°C)<sup>[6]</sup>. In this respect, several low-temperatures chemical methods were investigated for the formation of ultrafine  $\text{BaFe}_{12}\text{O}_{19}$  particles. These methods comprised coprecipitation<sup>[7-10]</sup>, hydrothermal<sup>[11-13]</sup>, sol-gel<sup>[14-16]</sup> microemulsion<sup>[17]</sup>, citrate precursor<sup>[18]</sup>, glass crystallization<sup>[19]</sup>, sonochemical<sup>[20]</sup> and mechano-chemical activation<sup>[21]</sup>. It has been reported that synthesis of barium ferrite by a hydrothermal reaction is preferable as it can be carried out at lower temperatures and fewer unwanted intermediates and/or impurities are obtained<sup>[22]</sup>. It is recognized that the magnetic properties of barium ferrite,  $\text{BaFe}_{12}\text{O}_{19}$  (BaM) can be optimized for particular purposes through doping. Solid state reaction method is used for the fabrication of barium based hexaferrites. The sites occupied by Fe ions can be replaced with Al, Co, Zr, Mn, Zn, Sm and Sn, or rare earth elements such as,  $\text{Ho}^{3+}$ ,  $\text{Nd}^{3+}$ <sup>[22-25]</sup>. In this study, the oxalate precursor technique was used to synthesize nanocrystalline barium ferrite with high saturation magnetization and narrow size distribution.

In this study, the oxalate precursor technique was used to synthesize nanocrystalline barium ferrite with high saturation magnetization. Effects of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios and the annealing temperature on the synthesis of ferrite powders were investigated. The annealing temperature was controlled from 900 to 1200°C, while  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios were controlled from 12 to 8.57.

## EXPERIMENTAL

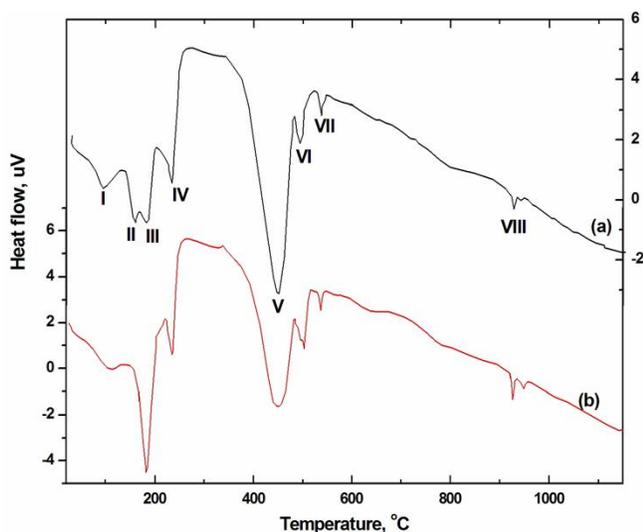
The oxalate precursor method was applied for the preparation of Barium hexaferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ). Chemically grade ferric chloride ( $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ), barium chloride ( $\text{BaCl}_2 \cdot \text{H}_2\text{O}$ ) and oxalic acid as source of organic were used as starting materials. A series of ferric chloride and barium chloride solution with various  $\text{Fe}^{3+}/\text{Ba}^{2+}$  molar ratios of (12, 10.9, 10.9, 23 and 8.57) and containing equivalent amount of oxalic acid were prepared. The mixtures of barium chloride and ferric chloride solution firstly prepared and then stirred for 15 min on a hot-plate magnetic stirrer, followed by addition of an aqueous solution, which was evaporated to 80°C with constant stirring until dry and then dried in a dryer at 100°C overnight. The dried powders obtained as barium ferrite precursors. Differential thermal analyzer (DTA) analysis of various un-annealed precursors was carried out. The rate of heating was kept at 10°C/min between room temperature and 1000°C. The measurements were carried out in a current of argon atmosphere.

For the formation of the barium ferrite phase, the dry precursors were annealed at the rate of 10°C/min in static air atmosphere up to different temperatures (900 - 1200°C and maintained at the temperature for annealed time (2h). The crystalline phases presented in the different annealed samples were identified by XRD on a Bruker axis D8 diffractometer using  $\text{Cu-K}\alpha$  ( $\lambda = 1.5406$ ) radiation and secondary monochromator in the range  $2\theta$  from 10 to 80°. The ferrites particles morphologies were observed by scanning electron microscope (SEM, JSM-5400).

The magnetic properties of the ferrites were measured at room temperature using a vibrating sample magnetometer (VSM; 9600-1 LDJ, USA) in a maximum applied field of 10 kOe. From the obtained hysteresis loops, the saturation magnetization ( $M_s$ ), remanence magnetization ( $M_r$ ) and coercivity ( $H_c$ ) were determined.

## RESULTS AND DISCUSSION

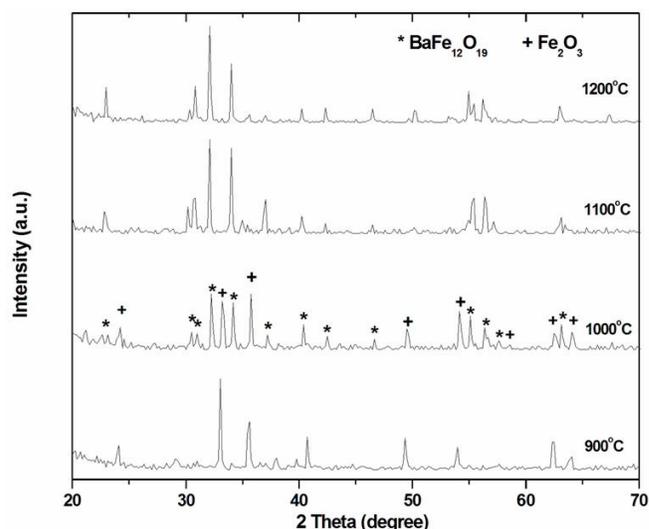
Figure 1 shows the differential thermal analysis (DTA) plot (a and b) of the synthesized mixture of barium - iron oxalates precursors at two different  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios 12 and 8.57 respectively. It can be seen in peaks I-IV that an endothermic reaction occurred at around (95.11, 158.52, 182.45 and 232.15 °C) which corresponds to the dehydration of iron and barium oxalates. This is consistent with earlier findings suggesting that, two different crystals hydrate types namely  $\text{MeC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$  and  $\text{MeC}_2\text{O}_4 \cdot 3\text{H}_2\text{O}$  (Me = metal ion)<sup>[26]</sup>. Thereafter, peaks V, VI and VII (448.02, 493.88 and 586.56°C) were significantly related to the anhydrous oxalate mixture decomposition into both metal oxide and gases ( $\text{CO}_2$  &  $\text{CO}$ ). Figure 1b shows that the VI peak has higher intensity in the case of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio 8.57 as compared with mole ratio 12. This is most likely due to the increase of barium oxalate amount. Peak VIII in the plot at (956.90 °C), showed the initial step to form  $\text{BaFe}_{12}\text{O}_{19}$ . Moreover, the intensity and sharpness of peak VIII in (plot b) was also increased, indicating that the stability of the formed barium ferrite will be increased with increasing content of barium oxalate ratio. Therefore, the DTA results indicate that barium ferrite cannot be formed before 956.90°C.



**Figure 1 :** Thermal profiles (DTA) of mixtures of barium-iron oxalates precursors at  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios (a) 12 and (b) 8.57.

Figure 2 shows the XRD patterns of the calcined powder of  $\text{BaFe}_{12}\text{O}_{19}$  obtained from barium – iron ox-

alate precursor solutions, with  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio 8.57 thermally treated at different temperatures (900 - 1200°C) for 2h. On the start of the annealing process at (900°C), a complete absence of M-type barium ferrite phase. Instead, the hematite  $\text{Fe}_2\text{O}_3$  phase appears as a major phase, which is consistent with DTA results. But at (1000°C) the concentration of the hematite phase decreases, and barium ferrite phase was detected. Increasing the annealing temperature to 1100°C, enhanced the formation of barium hexaferrite phase, and decreased the hematite  $\text{Fe}_2\text{O}_3$  phase. At the calcinations temperature (1200°C), single phase of barium hexaferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) evidently was formed.



**Figure 2 :** XRD patterns of  $\text{BaFe}_{12}\text{O}_{19}$  from barium-iron oxalate precursor with  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio 8.57 thermally treated at different temperatures (900-1200°C) for 2 h.

XRD analysis was carried out in this study to investigate the effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios of the powders thermally treated at different temperatures (900-1200°C) for 2h, and the results are presented in Figure 3-6. The results in Figure 3 indicate that the thermal calcination of barium – iron oxalate precursor at 900°C has not yielded barium ferrite phase  $\text{BaFe}_{12}\text{O}_{19}$  in any case. Instead iron oxide ( $\text{Fe}_2\text{O}_3$ ) phase has appeared clearly in all the  $\text{Fe}^{3+}/\text{Ba}^{2+}$  ratio. These results confirm the DTA results, which showed no sign of  $\text{BaFe}_{12}\text{O}_{19}$  formation at 900 °C. Figure 4 shows XRD patterns at annealing temperature 1000 °C, formation of barium ferrite was observed for all mole ratios. As expected, increasing the  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ration enhanced the formation of barium ferrite phase (i.e decreasing the amount of  $\text{Fe}_2\text{O}_3$ ). Figure 5

## Full Paper

showed the XRD patterns of  $\text{BaFe}_{12}\text{O}_{19}$  precursor powders at  $1100^\circ\text{C}$ . The formation of barium ferrite phase was highly enhanced for all mole ratios, while the  $\text{Fe}_2\text{O}_3$  formation diminished significantly. It can be also observed that single phase of well crystalline  $\text{BaFe}_{12}\text{O}_{19}$  was first obtained at  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio of 9.23 and 8.57 at annealing temperature  $1100^\circ\text{C}$ . Raising the calcination temperature for the precursor up to  $1200^\circ\text{C}$  (Figure 6), the single phase  $\text{BaFe}_{12}\text{O}_{19}$  appeared at all different  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio. At the same time no sign of iron oxide ( $\text{Fe}_2\text{O}_3$ ) appeared.

Figure 7 shows the effect of various mole ratios on the crystalline size of the obtained powders. It can be observed that increasing the annealing temperatures helps significantly agglomeration of the particles and grains growth during calcination course. This leads to the increase of grain size and formation of single phase barium hexaferrite powders. However, this observed crystalline size for the single phase barium hexaferrite produced via oxalate precursor route is much higher than that produced by other wet chemical methods. This finding was also observed by other authors<sup>[26,27]</sup>.

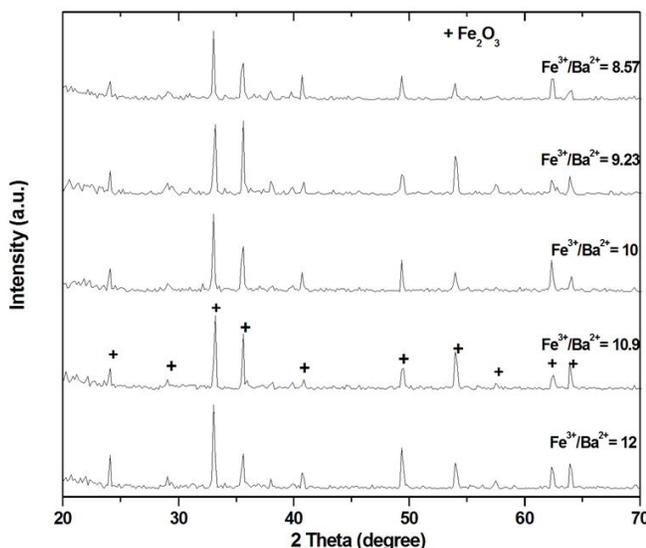


Figure 3 : XRD patterns of  $\text{BaFe}_{12}\text{O}_{19}$  from barium-iron oxalate precursor with different  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio and thermally treated at  $900^\circ\text{C}$  for 2 h.

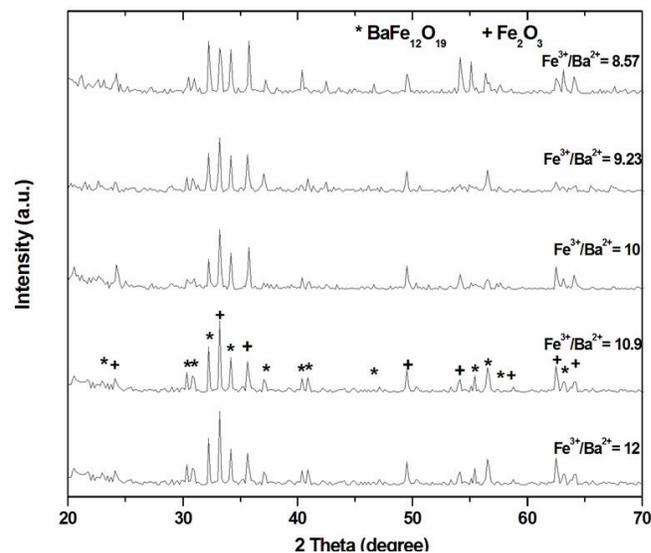


Figure 4 : XRD patterns of  $\text{BaFe}_{12}\text{O}_{19}$  from barium-iron oxalate precursor with different  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio and thermally treated at  $1000^\circ\text{C}$  for 2 h.

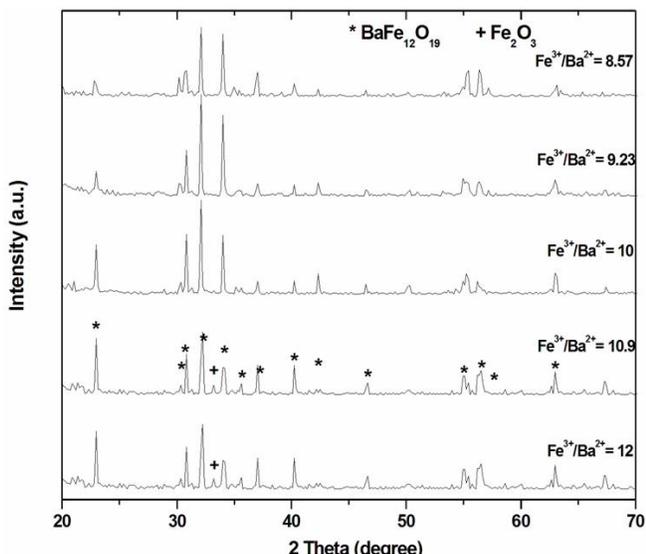


Figure 5 : XRD patterns of  $\text{BaFe}_{12}\text{O}_{19}$  from barium-iron oxalate precursor with different  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio and thermally treated at  $1100^\circ\text{C}$  for 2 h.

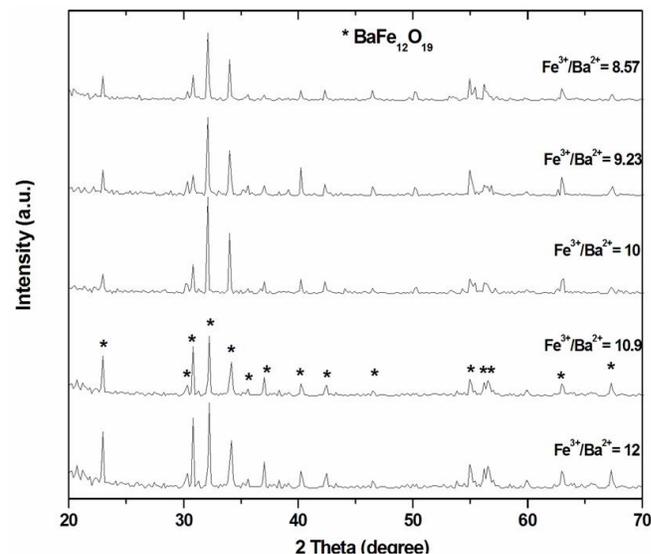
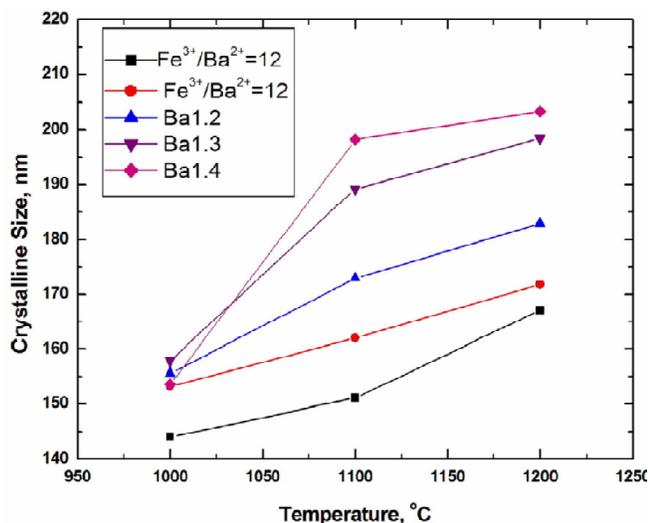


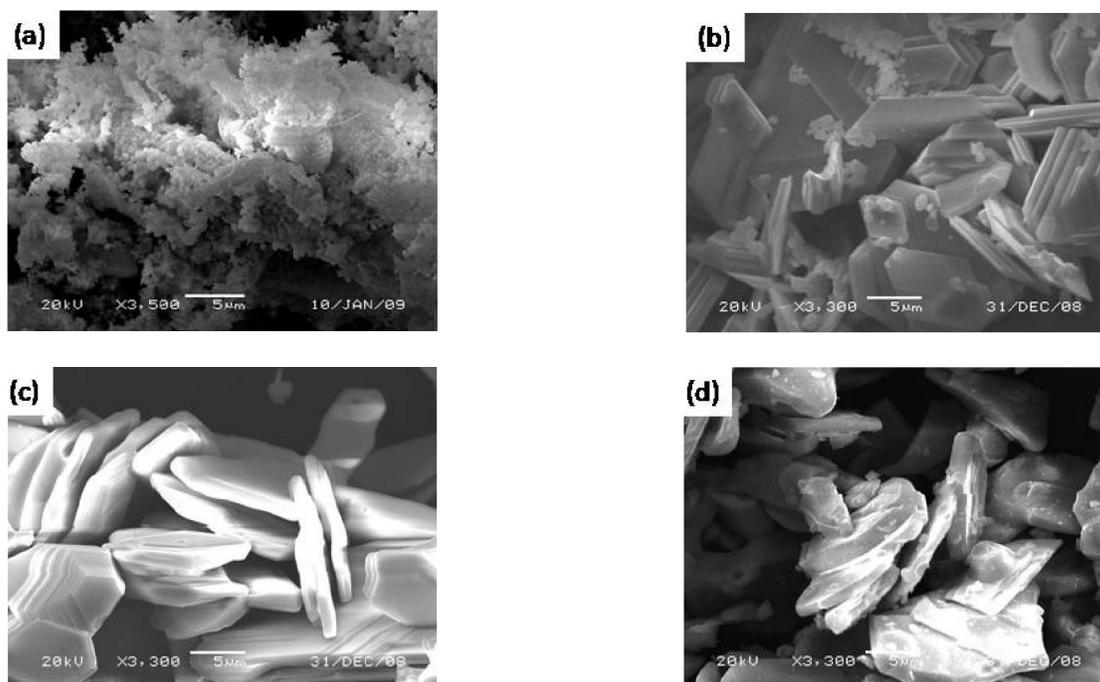
Figure 6 : XRD patterns of  $\text{BaFe}_{12}\text{O}_{19}$  from barium-iron oxalate precursor with different  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio and thermally treated at  $1200^\circ\text{C}$  for 2 h.



**Figure 7 :** Effect of annealing temperature and mole ratio on the crystalline size of the prepared barium hexaferrite

Figure 8 displays SEM micrographs of  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors with  $\text{Fe}^{3+}/$

$\text{Ba}^{2+}$  mole ratio of 9.23 and annealed for 2 hr. Clearly, it appears that increasing calcination temperature (900–1200°C) has a substantial effect on the microstructure of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders. In Figure 8(a), fine precipitated particles, with random grain orientation. This confirms the previous results of XRD and DTA, which showed no sign of  $\text{BaFe}_{12}\text{O}_{19}$  growth at 900°C. However, as the annealing temperatures increased to 1000°C Figure 8(b), individual particles possess a plate-like hexagonal shape containing a few numbers of spherical small particles. At annealing temperature 1100°C (Figure 8(c)), the ferrite powders showed uniform coarse structure with a well-clear hexagonal shape which is in line with XRD patterns in Figure 2, for where pure single crystal peaks of barium ferrite was very evident. The grains were then started to distort again at 1200°C (Figure 8(d)), which may lead to agglomeration of the particles at more higher annealing temperatures.

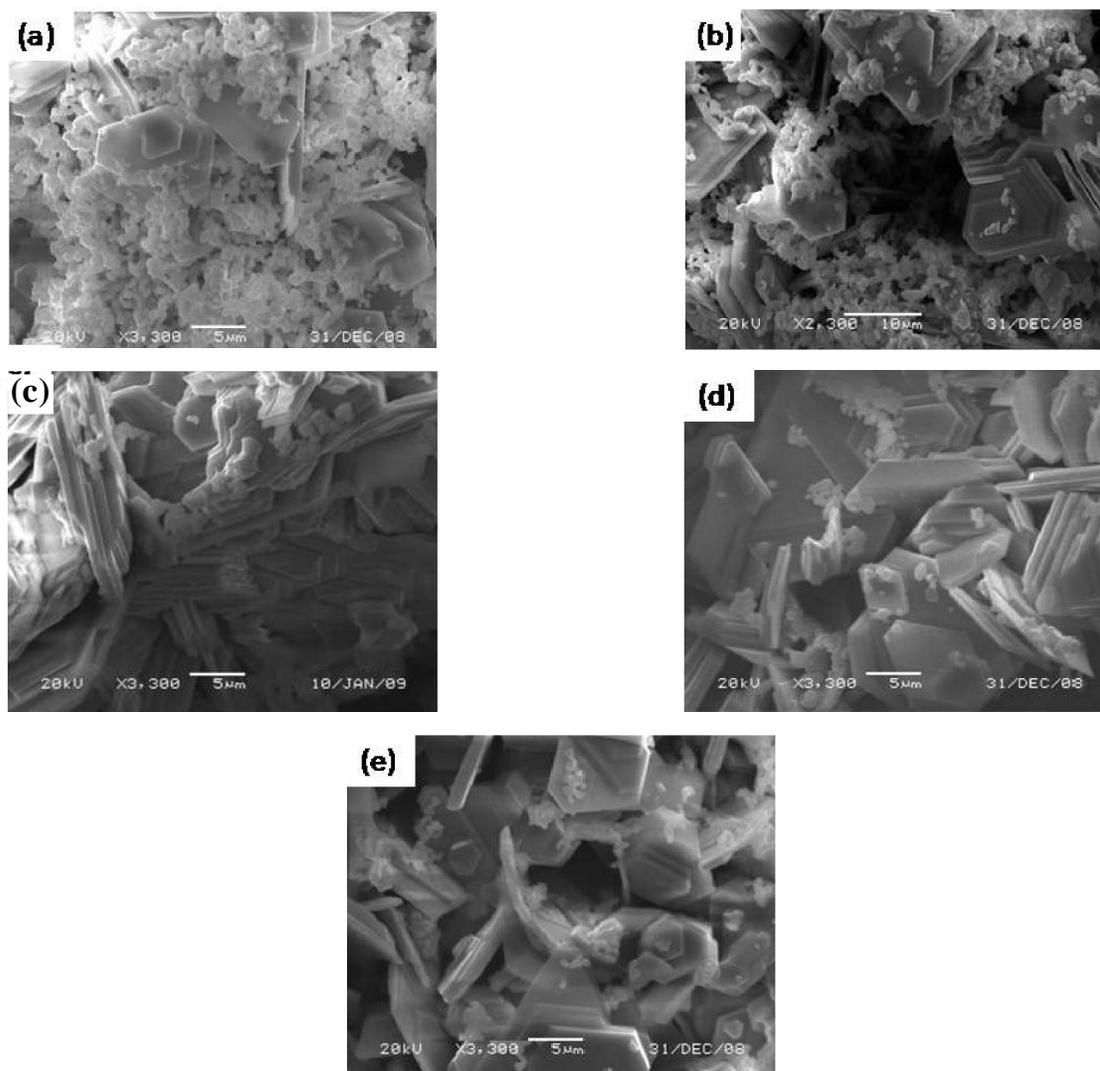


**Figure 8 :** Effect of annealing temperatures on the microstructure of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors with  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios of 9.23 and annealed for 2h. (a) 900 C, (b) 1000 °C, (c) 1100 °C, (d) 1200 °C

Figure 9(a-e) presented the SEM micrographs of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors and annealed at 1000°C for 2 hr. Effect of changing  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios on the microstructure was observed. In Figure 9(a and b), very fine particles of hematite powder started to agglomerate, where the mole ratios of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  were 12 and 10.9 respec-

tively. In addition, a few large crystal particles were formed, indicating that these ratios of the composition were insufficient for the complete formation of the structure. As the  $\text{Ba}^{2+}$  ion concentration increased in the composition of the samples (Figure 9 (c, d and e), uniform and coarse structure with clear homogeneous microstructure become more pronounced. Moreover, a well

## Full Paper



**Figure 9 :** Effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios on the microstructure of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors and annealed at  $1000^\circ\text{C}$  for 2 h. (a)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 12$ , (b)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 10.9$ , (c)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 10$ , (d)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 9.23$  and (e)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 8.57$

—clear crystalline micro-structure containing a fewer numbers of spherical small particles can be seen in these SEM micrographs. As the annealing temperature increased to  $1100^\circ\text{C}$  (Figure 10(a-e)), the produced powders of  $\text{BaFe}_{12}\text{O}_{19}$  possessed very well-defined plate-like hexagonal shape.

TABLE 1 and Figures (11-14), present the magnetic properties of the synthesized barium ferrite powders, which were obtained at room temperature under an applied field of 10 kOe. The results showed that the saturation magnetization of the produced powders increased by increasing the temperatures. Figure 11 display the effect of annealing temperature on the hysteresis loop of  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors at  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio 10. The results showed

that, the saturation magnetization of the produced powders increased by increasing the annealing temperature. It exhibited a maximum value of (70.25emu/g) at annealing temperature  $1200^\circ\text{C}$  for 2hr. This is likely due to the presence of single domain of  $\text{BaFe}_{12}\text{O}_{19}$  particles. In line with SEM results, the change in magnetic properties can be attributed to the presence of well crystalline  $\text{BaFe}_{12}\text{O}_{19}$  microstructures, as the annealing temperature of the powders was increasing gradually to reach optimum conditions. Figures (12-14) show the effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio on the M–H hysteresis loop of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors and annealed for 2h at 1000, 1100 and  $1200^\circ\text{C}$ , respectively. The maximum saturation magnetization value of 70.25 emu/g was obtained for

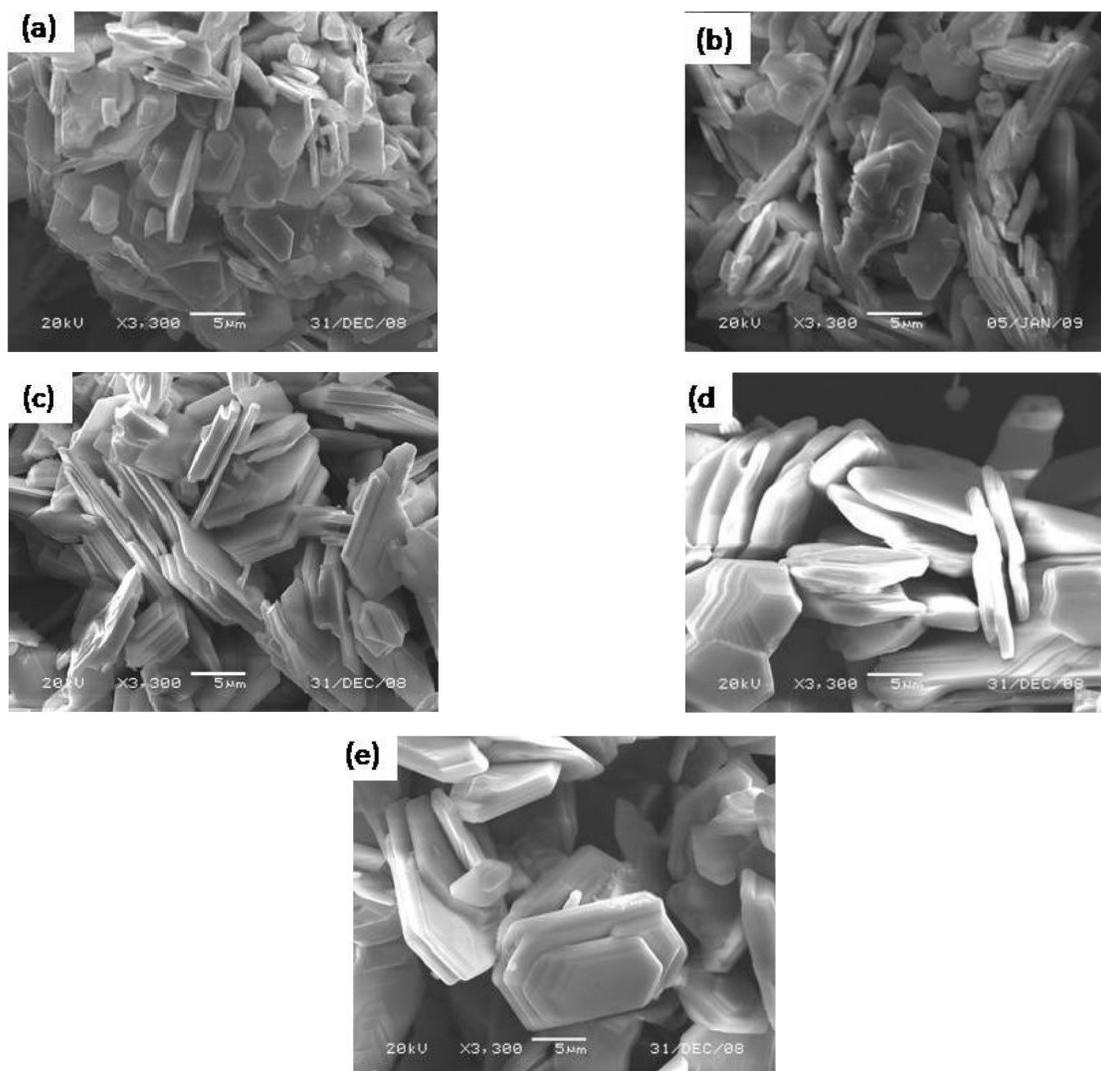


Figure 10 : Effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios on the microstructure of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors and annealed at  $1100^\circ\text{C}$  for 2 h. (a)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 12$ , (b)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 10.9$ , (c)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 10$ , (d)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 9.23$  and (e)  $\text{Fe}^{3+}/\text{Ba}^{2+} = 8.57$

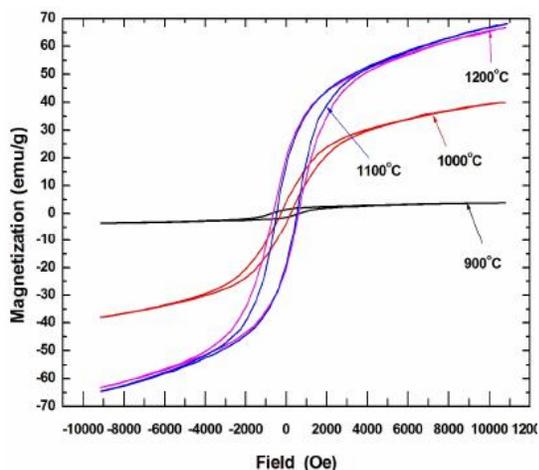


Figure 11 : Effect of annealing temperature on the M-H hysteresis loop of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors at  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio 10.

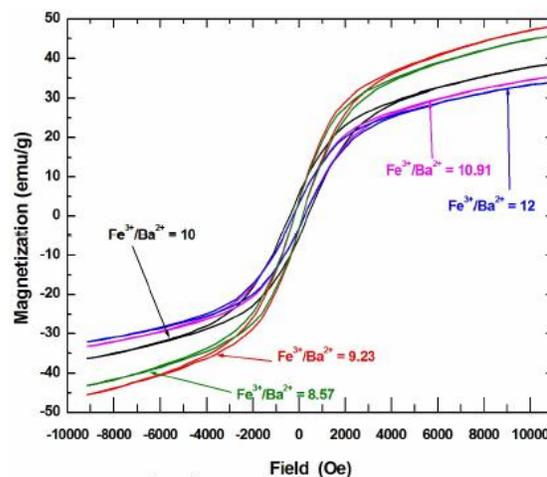


Figure 12 : Effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio on the M-H hysteresis loop of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors and annealed at  $1000^\circ\text{C}$  for 2 h.

## Full Paper

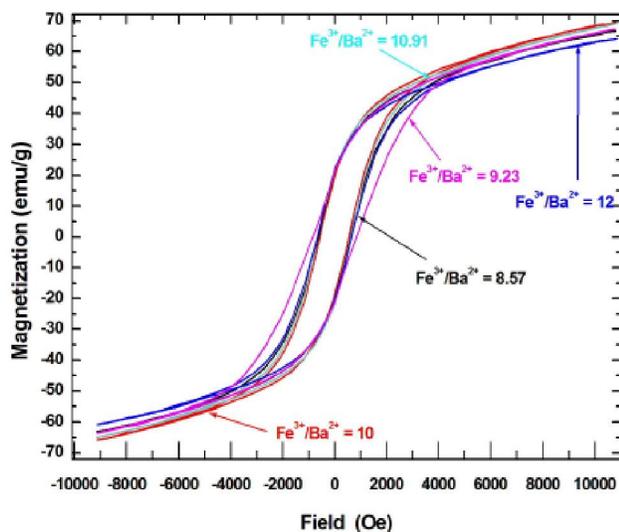


Figure 13 : Effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio on the M–H hysteresis loop of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors and annealed at  $1100\text{ }^{\circ}\text{C}$  for 2 h.

the precursor prepared at  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio 10 at  $1200\text{ }^{\circ}\text{C}$  for 2 h. This results shows that saturation magnetization values obtained from the oxalate precursor method was quite close to the theoretical limit of  $72\text{ emu/g}$ <sup>[7]</sup>. This was likely due to the presence of single domain of  $\text{BaFe}_{12}\text{O}_{19}$  particles. Such high saturation magnetization for barium ferrite at a  $1200\text{ }^{\circ}\text{C}$  can be attributed to the high  $\gamma$ -phase purity and well-defined crystalline structure of  $\text{BaFe}_{12}\text{O}_{19}$ . In contrast, the coercive force  $H_c$  results of  $\text{BaFe}_{12}\text{O}_{19}$  powders, which is produced by the oxalate precursor method were lower than the theoretical value of ( $6700\text{ Oe}$ ). In addition, these results might be related to the residual  $\text{Fe}_2\text{O}_3$  having a high intrinsic coercive force<sup>[3]</sup>. However, clearly this technique is a promise for providing  $\text{BaFe}_{12}\text{O}_{19}$  with the highest saturation magnetization value. Figure 11-13 and TABLE 1, display effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio on the M-H hysteresis loop of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained at different annealing temperatures.

Figure 12 showed that at annealing temperature  $1000\text{ }^{\circ}\text{C}$  decreasing the  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios from 12 to 8.57 increased the saturation magnetization of the formed  $\text{BaFe}_{12}\text{O}_{19}$  particles from  $33.01$  to  $44.8\text{ emu/g}$ . This is mainly due to increasing formation of well crystalline  $\text{BaFe}_{12}\text{O}_{19}$  powders, and decrease of the presence of non-magnetic species of  $\text{Fe}_2\text{O}_3$  as the mole ratio percentage went up. These results are in substantial agreement with the previous XRD and SEM results, which was shown in Figure 4 and 8. However,

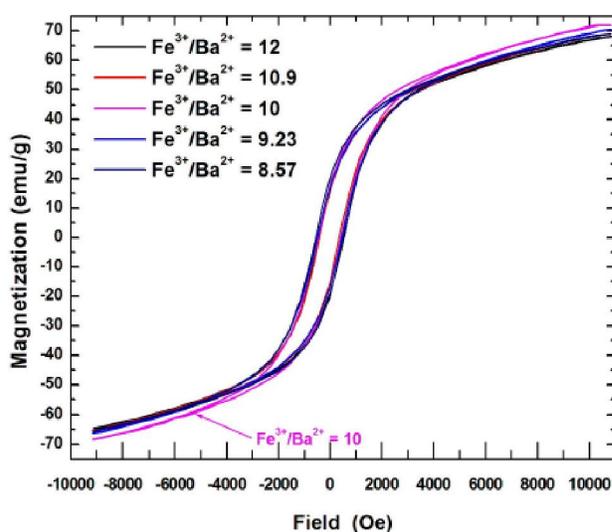


Figure 14 : Effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio on the M–H hysteresis loop of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained from oxalate precursors and annealed at  $1200\text{ }^{\circ}\text{C}$  for 2 h.

TABLE 1 : Effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratios and annealing temperature on the magnetic properties of Barium hexaferrite.

$\text{Fe}^{3+}/\text{Ba}^{2+}$ mole Ratio	Temp. ( $^{\circ}\text{C}$ )	Magnetic Properties		
		Ms (emu/g)	Mr (emu/g)	Hc (Oe)
12	900	3.667	1.371	666
	1000	33.01	3.264	250
	1100	62.44	21.41	662.6
	1200	66.36	17.5	503.8
10.91	900	2.074	0.8269	643.7
	1000	34.22	3.066	235.4
	1100	67.15	21.01	641.6
	1200	67.21	13.69	387.5
10	900	1.702	0.621	653
	1000	37.4	5.531	381.1
	1100	67.59	18.45	556.6
	1200	70.25	16.64	451.3
9.23	900	1.733	0.6292	653.3
	1000	46.82	3.471	172.6
	1100	65.44	20.83	837.8
	1200	68.45	15.11	480.6
8.57	900	2.728	2.728	808.7
	1000	44.8	3.507	240.8
	1100	64.96	19.12	620.8
	1200	67.23	18.77	536.7

the effect of  $\text{Fe}^{3+}/\text{Ba}^{2+}$  mole ratio on the M-H hysteresis loop of synthesized  $\text{BaFe}_{12}\text{O}_{19}$  powders obtained at  $1100\text{ }^{\circ}\text{C}$  and  $1200\text{ }^{\circ}\text{C}$  was less significant in Figure 13 and 14. This suggests that the  $\text{Fe}_2\text{O}_3$  particles di-

minished dramatically at temperature 1100 °C and 1200 °C, which lend support to the the XRD and SEM results corresponding to these temperatures in Figure 5 and 10, where the crystallinity of BaFe<sub>12</sub>O<sub>19</sub> powders were very evident.

## CONCLUSIONS

The structural and magnetic properties of newly prepared barium hexaferrite powders were studied in a comparative way. The results from DTA, XRD, SEM and VSM studies can be summarized as follows:

- Differential thermal analysis (DTA) plots of the synthesized mixture of barium- iron oxalates precursors showed that the initial step to form BaFe<sub>12</sub>O<sub>19</sub> started at (956.90 °C).
- Single phase of well crystalline BaFe<sub>12</sub>O<sub>19</sub> was first obtained at Fe<sup>3+</sup> /Ba<sup>2+</sup> mole ratio of 9.23 and 8.57 at annealing temperature 1100°C while at annealing temperature 1200°C the single phase BaFe<sub>12</sub>O<sub>19</sub> appeared at all different Fe<sup>3+</sup> /Ba<sup>2+</sup> mole ratio.
- The morphology of the particles at 1000 and 1100 °C are hexagonal platelet crystal. By increasing the temperature up to 1200 °C, grains have coalesced to form larger grains.
- The oxalate precursor route has proven to produce pure barium ferrite powders with good magnetic properties with maximum saturation magnetization value of (70.25 emu/g), and coercivity force (451.3 Oe).
- Regarding the particles size, it can be seen that, the minimum particle size appeared at (1000°C), and the maximum size was found at (1200°C), which most likely explained by the formation of the single phase of barium hexaferrite (BaFe<sub>12</sub>O<sub>19</sub>).

## REFERENCES

- D.Lisijak, M.Drofenik; J.of the European Ceramic Society, **27**, 4515 (2007).
- H.Kojimi, E.P.Wohlfarth (Ed.); Ferromagnetic Materials, North Holland, Amsterdam, **3**, 305 (1982).
- D.R.Richerson; 'Modern Ceramic Enginring', Marcel Dekker, Inc., NY, 294 (1992).
- M.M.Hessien, M.Radwan, M.M.Rashad; J.Anal. Appl.Pyrolisis, **78**, 282 (2007).
- V.Pillai, P.Kumar, M.S.Multani, D.O.Shah; Colloids Surf.A: Physi-Cochem.Eng.Aspects, **80**, 69 (1993).
- H.Sözeri; J.Alloys Compd., **486**, 809 (2009).
- S.E.Jacobo, L.Civale, C.Domingo-Pascual, R.Rodrigues-Clements, M.A.Blesa; J.Mater.Sci., **32**, 1025 (1997).
- O.Carp, R.Barjega, E.Segal, M.Brezeanu; Thermochim.Acta, **318**, 57 (1998).
- T.Ogasawara, M.A.S.Oliveira; J.Magn.Magn. Mater., **217**, 147 (2000).
- J.Matutes-Aquino, S.D'áz-Castañón, M.Mirabal-García, S.A.Palomares-Sánchez; Scripta Mater., **42**, 295 (2000).
- M.-L.Wang, Z.W.Shih, C.-H.Lin; J.Cryst.Growth, **130**, 153 (1993).
- X.Liu, J.Wang, L.-M.Gan, S.-C.Ng; J.Magn.Magn. Mater., **195**, 452 (1999).
- D.Mishra, S.Anand, R.K.Panda, R.P.Das; Mater.Chem.Phys., **86**, 132 (2004).
- C.Sürig, K.A.Hempel, Ch.Sauer; J.Magn.Magn. Mater., **157-158**, 268 (1996).
- W.Zhong, W.Ding, N.Zhang, J.Hong, Q.Yan, Y.Du; J.Magn.Magn.Mater., **168**, 196 (1997).
- R.M.Garcia, E.R.Ruiz, E.E.Rams, R.M.Sanchez; J.Magn.Magn.Mater., **223**, 133 (2001).
- V.Pillai, P.Kumar, M.S.Multani, D.O.Shah; Colloids Surf.A: Physicochem Eng.Aspects, **80**, 69 (1993).
- V.K.Sankaranarayanan, D.C.Khan; J.Magn.Magn. Mater., **153**, 337 (1996).
- M.El-Hilo, H.Pfeiffer, K.O'Grady, W.Schuppel, E.Sinn, P.Görnert, M.Rösler, D.P.E.Dickson, R.W.Chantrell; J.Magn.Magn.Mater., **129**, 339 (1994).
- K.V.P.M.Shafi, A.Gedanken; Nanostructured Mater., **12**, 29 (1999).
- O.Abe, M.Narita; Solid State Ionics, **101-103**, 103 (1997).
- Y.Liu, M.G.B.Drew, J.Wang, M.Zhang, Y.Liu; J.of Magn.Magn.Mater., **322**, 366 (2010).
- Y.Liu, M.G.B.Drew, J.Wang, M.Zhang, Y.Liu; J.Alloys Compd., **322**, 814 (2010).
- J.Xu, H.Zou, H.Li, G.Li, S.Gan, G.Hong; J.Alloys Compd., **490**, 552 (2010).
- L.Wang, J.Song, Q.Zhang, X.Huang, N.Xu; J.Alloys Compd., **481**, 863 (2009).
- M.M.Hessien; J.Magn.Magn.Mater., **320**, 2800 (2008).
- M.M.Hessien, M.M.Rashad, M.S.Hassan, K.El-Barawy; J.Alloys Compd., **476**, 373 (2009).