

Factors Affecting the Synthesis and Formation of Single-Phase Barium Hexaferrite by a Technique of Oxalate Precursor

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Abstract: Problem statement: 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. **Approach:** 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 research. The annealing temperature was controlled from 900-1200°C, while the $\text{Fe}^{3+}/\text{Ba}^{2+}$ was controlled from 12-8.57. **Results:** 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^{-1}) 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. **Conclusion:** The barium hexaferrite was synthesized at annealing temperature 1200°C with a single phase using oxalate as precursor route.

Key words: Barium hexaferrite, annealing temperature, mole ratio, magnetic properties, oxalate precursor

INTRODUCTION

Permanent magnet materials are essential in devices for storing energy in a static magnetic field. Ferrite is a class of ceramic materials with useful electromagnetic properties and the electromagnetic properties for ferrite materials is affected by operating conditions such as temperature, pressure, field strength, frequency and time. 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. Hexagonal ferrites, $\text{MFe}_{12}\text{O}_{19}$, (where M = Ba, Sr and/or Pb) are a group of magnetic compounds, which all have high resistivity, magneto-crystalline anisotropy and saturation magnetization, low dielectric losses and are thermally stable well above their Curie temperature (Kojimi and Wohlfarth, 1982; Smit and Wijn, 1959). The hexagonal M-type hard ferrites have attracted much attention as the most widely used permanent magnets, which account for about 90% of the annual production of the

permanent magnets due to the good combination of high magnetic properties, chemical stability and low cost. Moreover, M-type hexaferrites have widely used in telecommunication, magnetic recording media, magneto-optics and microwave devices (Kojima, 1982; Smit and Wijn, 1959; Ogasawara and Oliveira, 2000). 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 (Hessien *et al.*, 2008). $\text{BaFe}_{12}\text{O}_{19}$ (BaM) and its derivatives are currently magnetic material with great scientific and technological interest, due to its relatively high curie temperature, high coercive force and high magnetic anisotropy field as well as an excellent chemical stability and corrosion resistivity (Kojimi and Wohlfarth, 1982). Barium hexaferrite ($\text{BaFe}_{12}\text{O}_{19}$) has a complex hexagonal unit cell and belonging to the magnetoplumbite structures (Richerson, 1992). Magnetoplumbite are of the type $\text{A}^{2+}\text{O}_1.6\text{B}_2^{3+}\text{O}_3$. The arrangement of the 12 Fe^{3+} ions in the unit cell is as follows: Two ions in the tetrahedral sites (four nearest O^{2-} neighbors), nine ions in the octahedral sites (six nearest O^{2-} neighbors) and one ion in the hexagonal site

(five nearest O²⁻ neighbors). Materials of this type have a strong uniaxial magnetic direction, making as permanent magnets. 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⁻¹ and 450°C, respectively (Miller and Drillon, 2002; Pillai *et al.*, 1993). 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 iron oxide at high temperatures (about 1200°C) (Cabaoas and Gonzalez-Calbet, 1993). In this respect, several low-temperatures chemical methods were investigated for the formation of ultrafine BaFe₁₂O₁₉ particles. These methods comprised coprecipitation (Hessien *et al.*, 2008; Jacobo *et al.*, 1997; Matutes-Aquino *et al.*, 2000), hydrothermal (Wang *et al.*, 1993; Liu *et al.*, 1999; Mishra *et al.*, 2004), sol-gel (Surig *et al.*, 1996; Zhong *et al.*, 1997; Garcia *et al.*, 2001; Jacobo *et al.*, 1997), microemulsion (Pillai *et al.*, 1993), oxalate precursor (Sankaranarayanan and Khan, 1996), glass crystallization (El-Hilo *et al.*, 1994), sonochemical (Shafi and Gedanken, 1999) and mechano-chemical activation (Abe and Narita, 1997). The oxalate precursor technique was found to be more suitable for synthesis of barium ferrite with single phase powder. In addition, this technique needs relatively low-processing temperature to produce homogenous microstructure with narrow size distribution and uniform shape. In this study, the oxalate precursor technique was used to synthesize nanocrystalline barium ferrite with high saturation magnetization. Effects of Fe³⁺/Ba²⁺ mole ratios and the annealing temperature on the synthesis of ferrite powders were investigated. The annealing temperature was controlled from 900-1200°C, while Fe³⁺/Ba²⁺ mole ratios were controlled from 12-8.57.

MATERIALS AND METHODS

Preparation: The oxalate precursor method was applied for the preparation of Barium hexaferrite (BaFe₁₂O₁₉). Chemically grade ferric chloride (FeCl₃·6H₂O), barium chloride (BaCl₂·H₂O) and oxalic acid as source of organic were used as starting materials. A series of ferric chloride and barium chloride solution with various Fe³⁺/Ba²⁺ 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.

Measurements: Differential Thermal Analyzer (DTA) analysis of various un-annealed precursors was carried out. The rate of heating was kept at 10°C min⁻¹ 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⁻¹ in static air atmosphere up to different temperatures (900-1200°C and maintained at the temperature for annealed time (2 h). The crystalline phases presented in the different annealed samples were identified by XRD on a Bruker axis D8 diffractometer using Cu-Kα (λ = 1.5406) radiation and secondary monochromator in the range 2θ from 10-80°. 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

Figure 1 shows the Differential Thermal Analysis (DTA) plot (a and b) of the synthesized mixture of barium-iron oxalates precursors at two different Fe³⁺/Ba²⁺ 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 MeC₂O₄·2H₂O and MeC₂O₄·3H₂O (Me = metal ion) (Shafi and Gedanken, 1999; Abe and Narita, 1997; Hessien, 2008). 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 (CO₂ and CO). Figure 1b shows that the VI peak has higher intensity in the case of Fe³⁺/Ba²⁺ 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 BaFe₁₂O₁₉. 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 form before 956.90°C (Carp *et al.*, 1998).

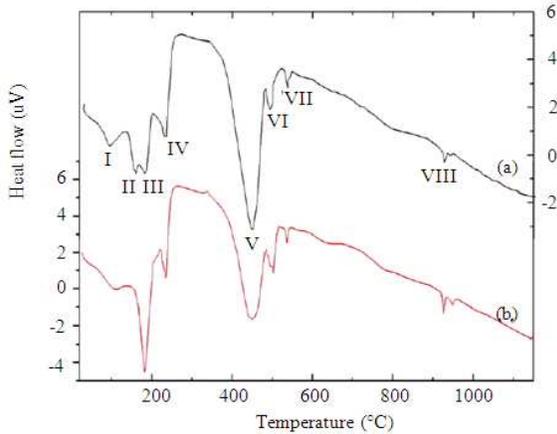


Fig. 1: Thermal profiles (DTA) of mixtures of barium-iron oxalates precursors at $\text{Fe}^{3+}/\text{Ba}^{2+}$ mole ratios (a) 12 and (b) 8.57

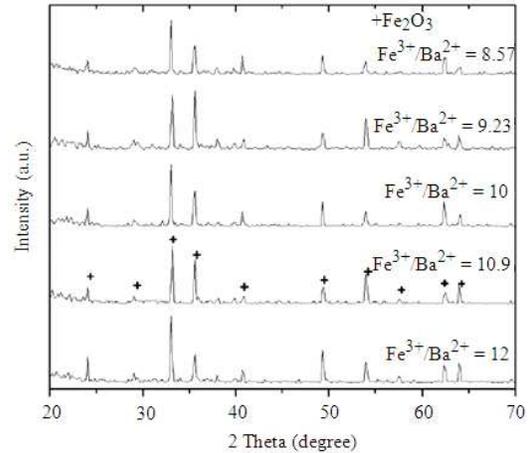


Fig. 3: XRD patterns of $\text{BaFe}_{12}\text{O}_{19}$ from barium-iron oxalate precursor with different $\text{Fe}^{3+}/\text{Ba}^{2+}$ mole ratio and thermally treated at 900°C for 2 h

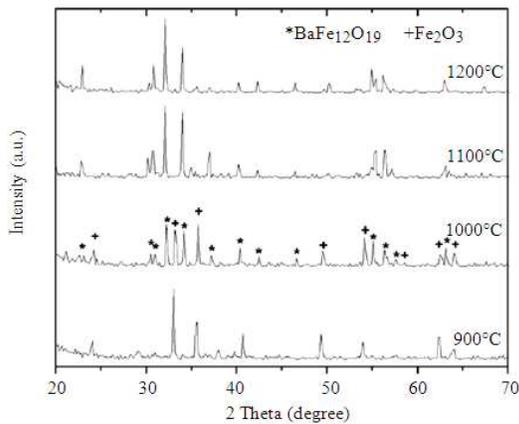


Fig. 2: XRD patterns of $\text{BaFe}_{12}\text{O}_{19}$ from barium-iron oxalate precursor with $\text{Fe}^{3+}/\text{Ba}^{2+}$ mole ratio 8.57 thermally treated at different temperatures (900 - 1200°C) for 2 h

Figure 2 shows the XRD patterns of the calcined powder of $\text{BaFe}_{12}\text{O}_{19}$ obtained from barium-iron oxalate precursor solutions, with $\text{Fe}^{3+}/\text{Ba}^{2+}$ mole ratio 8.57 thermally treated at different temperatures (900 - 1200°C) for 2 h. On the start of the annealing process at (900°C), a complete absence of M-type barium ferrite phase. Instead, the hematite Fe_2O_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 Fe_2O_3 phase. At the calcinations temperature (1200°C), single phase of barium hexaferrite ($\text{BaFe}_{12}\text{O}_{19}$) evidently was formed.

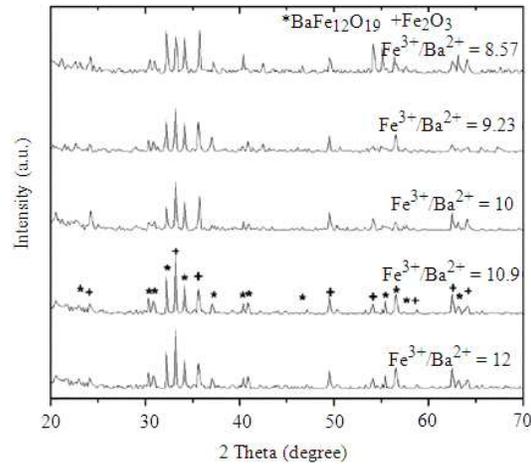


Fig. 4: XRD patterns of $\text{BaFe}_{12}\text{O}_{19}$ from barium-iron oxalate precursor with different $\text{Fe}^{3+}/\text{Ba}^{2+}$ mole ratio and thermally treated at 1000°C for 2 h

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 2 h and the results are presented in Fig. 3-6. The results in Fig. 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 (Fe_2O_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.

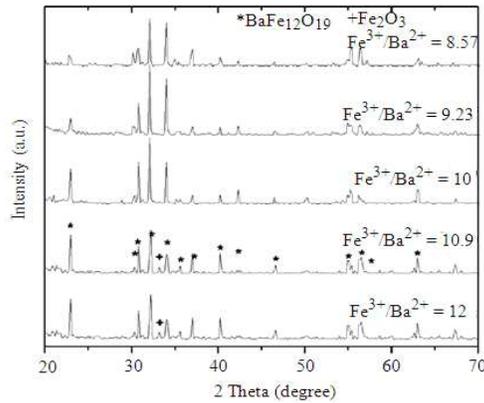


Fig. 5: XRD patterns of BaFe₁₂O₁₉ from barium-iron oxalate precursor with different Fe³⁺/Ba²⁺ mole ratio and thermally treated at 1100°C for 2 h

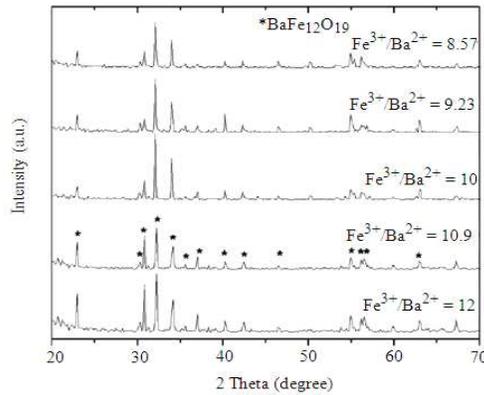


Fig. 6: XRD patterns of BaFe₁₂O₁₉ from barium-iron oxalate precursor with different Fe³⁺/Ba²⁺ mole ratio and thermally treated at 1200°C for 2 h

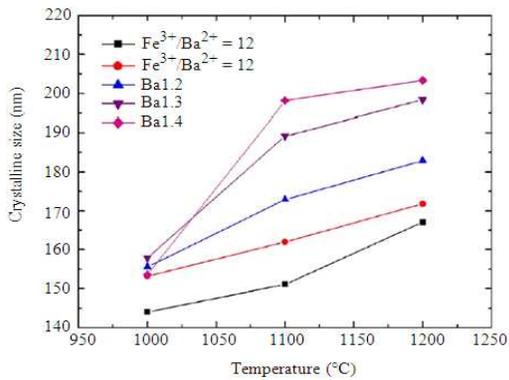


Fig 7: Effect of annealing temperature and mole ratio on the crystalline size of the prepared barium hexaferrite

As expected, increasing the Fe³⁺/Ba²⁺ mole ratio enhanced the formation of barium ferrite phase (i.e., decreasing the amount of Fe₂O₃). Figure 5 showed the XRD patterns of BaFe₁₂O₁₉ precursor powders at 1100°C. The formation of barium ferrite phase was highly enhanced for all mole ratios, while the Fe₂O₃ formation diminished significantly. It can be also observed that single phase of well crystalline BaFe₁₂O₁₉ was first obtained at Fe³⁺/Ba²⁺ mole ratio of 9.23 and 8.57 at annealing temperature 1100°C. Raising the calcination temperature for the precursor up to 1200°C (Fig. 6), the single phase BaFe₁₂O₁₉ appeared at all different Fe³⁺/Ba²⁺ mole ratio. At the same time no sign of iron oxide (Fe₂O₃) 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.

Figure 8 displays SEM micrographs of BaFe₁₂O₁₉ powders obtained from oxalate precursors with Fe³⁺/Ba²⁺ mole ratio of 9.23 and annealed for 2 h.

Clearly, it appears that increasing calcination temperature (900-1200°C) has a substantial effect on the microstructure of synthesized BaFe₁₂O₁₉ powders.

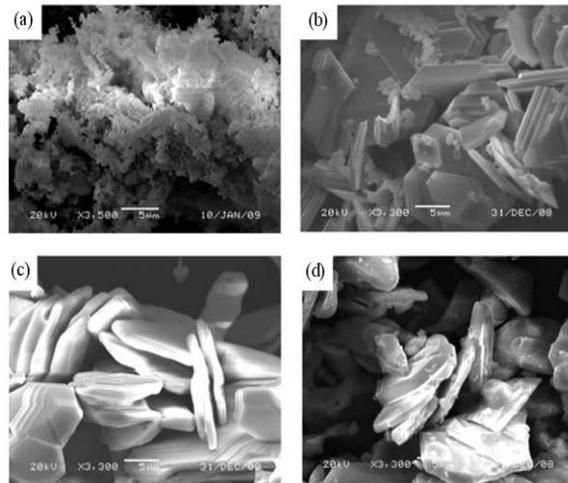


Fig. 8: Effect of annealing temperatures on the microstructure of synthesized BaFe₁₂O₁₉ powders obtained from oxalate precursors with Fe³⁺/Ba²⁺ mole ratios of 9.23 and annealed for 2 h. (a) 900°C; (b) 1000°C; (c) 1100°C; (d) 1200°C

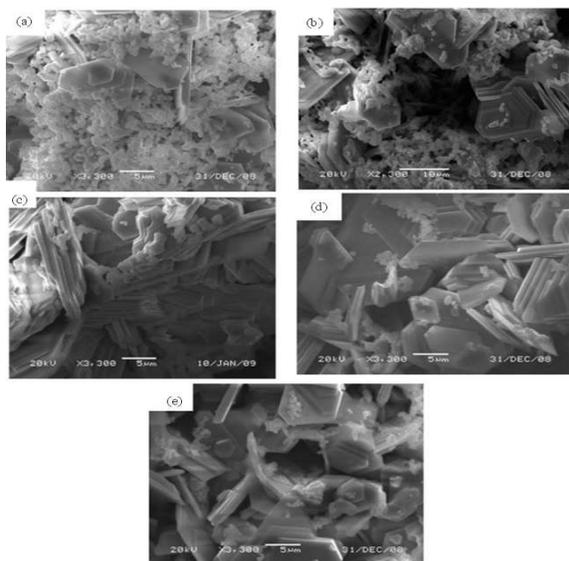


Fig. 9: Effect of Fe³⁺/Ba²⁺ mole ratios on the microstructure of synthesized BaFe₁₂O₁₉ powders obtained from oxalate precursors and annealed at 1000°C for 2 h. (a) Fe³⁺/Ba²⁺ = 12; (b) Fe³⁺/Ba²⁺ = 10.9; (c) Fe³⁺/Ba²⁺ = 10; (d) Fe³⁺/Ba²⁺ = 9.23 and (e) Fe³⁺/Ba²⁺ = 8.57

In Fig. 8a, fine precipitated particles, with random grain orientation. This confirms the previous results of XRD and DTA, which showed no sign of BaFe₁₂O₁₉ growth at 900°C. However, as the annealing temperatures increased to 1000°C Fig. 8b, individual particles possess a plate-like hexagonal shape containing a fewer numbers of spherical small particles. At annealing temperature 1100°C (Fig. 8c), the ferrite powders showed uniform coarse structure with a well-clear hexagonal shape which is in line with XRD patterns in Fig. 2, for where pure single crystal peaks of barium ferrite was very evident. The grains were then started to distort again at 1200°C (Fig. 8d), which may lead to agglomeration of the particles at more higher annealing temperatures.

Figure 9a-e presented the SEM micrographs of synthesized BaFe₁₂O₁₉ powders obtained from oxalate precursors and annealed at 1000°C for 2 h. Effect of changing Fe³⁺/Ba²⁺ mole ratios on the microstructure was observed. In Fig. 9a and b, very fine particles of hematite powder started to agglomerate, where the mole ratios of Fe³⁺/Ba²⁺ were 12 and 10.9 respectively. 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.

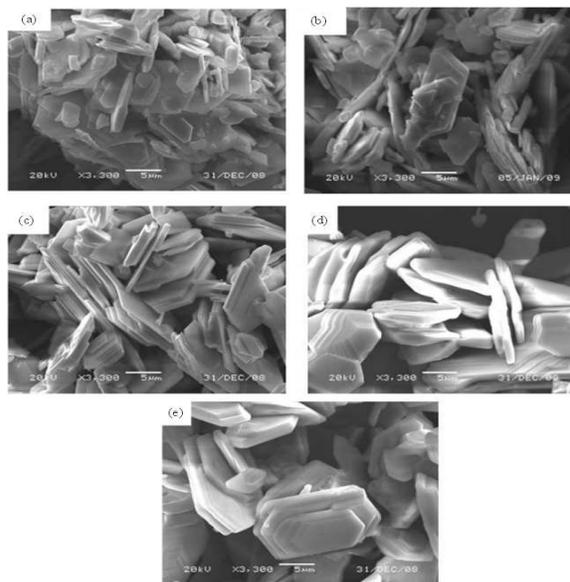


Fig. 10: Effect of Fe³⁺/Ba²⁺ mole ratios on the microstructure of synthesized BaFe₁₂O₁₉ powders obtained from oxalate precursors and annealed at 1100°C for 2 h. (a) Fe³⁺/Ba²⁺ = 12; (b) Fe³⁺/Ba²⁺ = 10.9; (c) Fe³⁺/Ba²⁺ = 10; (d) Fe³⁺/Ba²⁺ = 9.23 and (e) Fe³⁺/Ba²⁺ = 8.57

As the Ba²⁺ ion concentration increased in the composition of the samples (Fig. 9c-e), uniform and coarse structure with clear homogeneous microstructure become more pronounced. Moreover, a well-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°C (Fig. 10a-e), the produced powders of BaFe₁₂O₁₉ possessed very well-defined plate-like hexagonal shape.

DISCUSSION

Table 1 and Fig. 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 BaFe₁₂O₁₉ powders obtained from oxalate precursors at Fe³⁺/Ba²⁺ mole ratio 10. This is likely due to the presence of single domain of BaFe₁₂O₁₉ particles. In line with SEM results, the change in magnetic properties can be attributed to the presence of well crystalline BaFe₁₂O₁₉ microstructures, as the annealing temperature of the powders was increasing gradually to reach optimum conditions.

Table 1: Effect of Fe³⁺/Ba²⁺ mole ratios and annealing temperature on the magnetic properties of Barium hexaferrite

Fe ³⁺ /Ba ²⁺ mole ratio	Temp. (°C)	Magnetic properties		
		Ms (emu g ⁻¹)	Mr (emu g ⁻¹)	Hc (Oe)
12	900	3.667	1.3710	666.0
	1000	33.010	3.2640	250.0
	1100	62.440	21.4100	662.6
	1200	66.360	17.5000	503.8
10.91	900	2.074	0.8269	643.7
	1000	34.220	3.0660	235.4
	1100	67.150	21.0100	641.6
	1200	67.210	13.6900	387.5
10	900	1.702	0.6210	653.0
	1000	37.400	5.5310	381.1
	1100	67.590	18.4500	556.6
	1200	70.250	16.6400	451.3
9.23	900	1.733	0.6292	653.3
	1000	46.820	3.4710	172.6
	1100	65.440	20.8300	837.8
	1200	68.450	15.1100	480.6
8.57	900	2.728	2.7280	808.7
	1000	44.800	3.5070	240.8
	1100	64.960	19.1200	620.8
	1200	67.230	18.7700	536.7

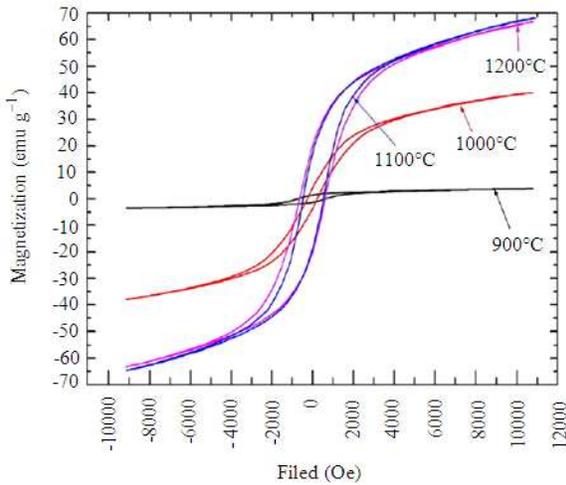


Fig. 11: Effect of annealing temperature on the M-H hysteresis loop of synthesized BaFe₁₂O₁₉ powders obtained from oxalate precursors at Fe³⁺/Ba²⁺ mole ratio 10

Figure 12-14 show the effect of Fe³⁺/Ba²⁺ mole ratio on the M-H hysteresis loop of synthesized BaFe₁₂O₁₉ powders obtained from oxalate precursors and annealed for 2 h at 1000, 1100 and 1200°C, respectively. The maximum saturation magnetization value of 70.25 emu g⁻¹ was obtained for the precursor prepared at Fe³⁺/Ba²⁺ mole ratio 10 at 1200°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 emu g⁻¹ (Pillai *et al.*, 1993).

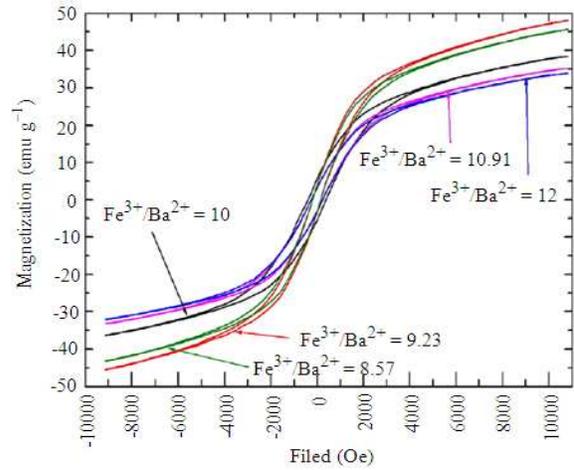


Fig. 12: Effect of Fe³⁺/Ba²⁺ mole ratio on the M-H hysteresis loop of synthesized BaFe₁₂O₁₉ powders obtained from oxalate precursors and annealed at 1000°C for 2 h

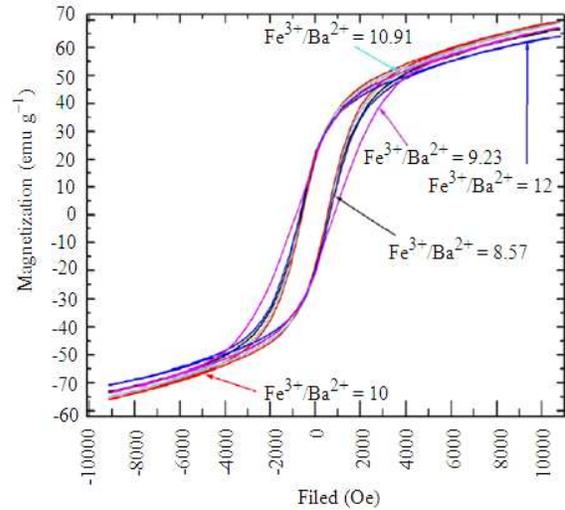


Fig. 13: Effect of Fe³⁺/Ba²⁺ mole ratio on the M-H hysteresis loop of synthesized BaFe₁₂O₁₉ powders obtained from oxalate precursors and annealed at 1100°C for 2 h

This was likely due to the presence of single domain of BaFe₁₂O₁₉ particles. Such high saturation magnetization for barium ferrite at a1200°C can be attributed to the high-phase purity and well-defined crystalline structure of BaFe₁₂O₁₉. In contrast, the coercive force Hc results of BaFe₁₂O₁₉ powders, which is produced by the oxalate precursor method were lower than the theoretical value of (6700 Oe). In addition, these results might be related to the residual Fe₂O₃ having a high intrinsic coercive force (Hessien *et al.*, 2008).

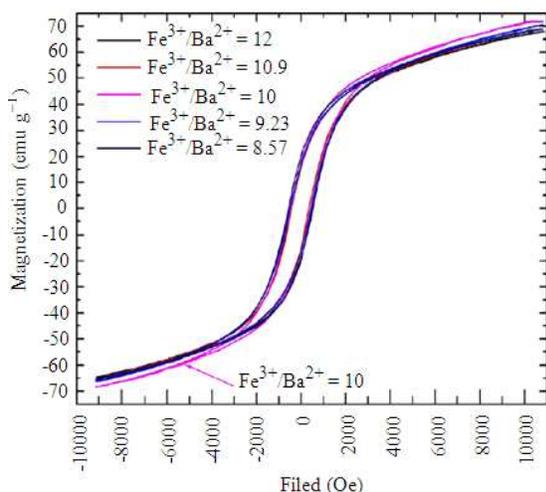


Fig. 14: Effect of $\text{Fe}^{3+}/\text{Ba}^{2+}$ mole ratio on the M-H hysteresis loop of synthesized $\text{BaFe}_{12}\text{O}_{19}$ powders obtained from oxalate precursors and annealed at 1200°C for 2 h

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°C decreasing the $\text{Fe}^{3+}/\text{Ba}^{2+}$ mole ratios from 12-8.57 increased the saturation magnetization of the formed $\text{BaFe}_{12}\text{O}_{19}$ particles from 33.01-44.8 emu g^{-1} . 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 Fe_2O_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 Fig. 4 and 8. However, 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°C and 1200°C was less significant in Fig. 13 and 14. This suggests that the Fe_2O_3 particles diminished dramatically at temperature 1100°C and 1200°C , which lend support to the XRD and SEM results corresponding to these temperatures in Fig. 5 and 10, where the crystallinity of $\text{BaFe}_{12}\text{O}_{19}$ powders were very evident.

CONCLUSION

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 $\text{BaFe}_{12}\text{O}_{19}$ started at (956.90°C)
- 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 while 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 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^{-1}) 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 ($\text{BaFe}_{12}\text{O}_{19}$)

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