Effect of polyethylene glycol 6000 on the microstructure and magnetic properties of BaFe\textsubscript{10.4}Al\textsubscript{1.6}O\textsubscript{19}

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Abstract

Aluminum-substituted barium hexaferrite (BaFe\textsubscript{10.4}Al\textsubscript{1.6}O\textsubscript{19}) powders were prepared using sol-gel auto-combustion and the calcination method. The effects of adding 1 g, 1.5 g, and 2 g of polyethylene glycol (PEG 6000) to the sol solution on the microstructure and magnetic properties of calcined BaFe\textsubscript{10.4}Al\textsubscript{1.6}O\textsubscript{19} were investigated. The x-ray diffraction analysis of the synthesized sample without PEG produced a single-phase BaFe\textsubscript{10.4}Al\textsubscript{1.6}O\textsubscript{19}; furthermore, BaFe\textsubscript{10.4}Al\textsubscript{1.6}O\textsubscript{19} and hematite (α-Fe\textsubscript{2}O\textsubscript{3}) coexisted with the addition of 1–2 g of PEG. Scanning electron microscope analysis revealed the transformation of the morphology of BaFe\textsubscript{10.4}Al\textsubscript{1.6}O\textsubscript{19} from nanorod to hexagonal platelet in shape and growth in particle sizes as increasing amounts of PEG were added. The prepared samples’ magnetic data revealed an increase in the coercive field with PEG’s addition, while the saturation magnetization increased for PEG additions of up to 1.5 g (1 g and 1.5 g) and decreased for 2 g of PEG. The particle sizes, coercive field, and saturation magnetization confirmed the formation of single-domain BaFe\textsubscript{10.4}Al\textsubscript{1.6}O\textsubscript{19} for PEG additions of up to 1.5 g and multi-domain formation BaFe\textsubscript{10.4}Al\textsubscript{1.6}O\textsubscript{19} for a PEG addition of 2 g.

1. Introduction

Hexaferrite has been known as a magnetic metal oxide class that emerged as one of the most widely used permanent magnet materials for practical applications. The main advantages of hexaferrite are low cost, ease of mass production, chemical stability, and electrical and magnetic properties suited to specific device requirements [1]. Among these ferrites, the M-type hexaferrite has been produced in large quantities and used as part of the vital component in modern devices, such as motors, generators, transformers, sensors and actuators, computers, cellular phones, radios, loudspeakers, microwaves, relays, and disc drives [2].

The M-type (M = Ba, Pb, Sr) hexaferrite has attracted technological applications due to its unique magnetic and electrical properties such as high magnetocrystalline anisotropy and magnetic anisotropy field, large electrical resistivity [3]. Such unique material properties provide the basic requirement for producing high-density recording media [4] and good electromagnetic absorber materials used in telecommunication and defense industries [5].

Various methods have been used to prepare hexaferrite, such as the conventional ceramic method [6] and wet a variety of chemical methods, including co-precipitation method [7], hydrothermal synthesis [8], a sol-gel auto-combustion method [9]. The ceramic solid-state reaction method involves mixing and grinding the reaction product, resulting in an irregular shape, size and stoichiometry of the final product. On the other hand, the wet chemical methods offer a simple and safe process conducted at relatively low temperatures, producing high purity and narrow size distribution of fine particles [10].
Barium hexaferrite (BaFe$_{12}$O$_{19}$) is hard magnetic material with a magnetoplumbite structure belonging to the hexagonal space group of $P6/mmc$. The hexagonal structure has five different Fe crystallographic sites, i.e., tetrahedral (4f1), octahedral (2k, 12a and 4f2), and trigonal bipyramid (2b) coupled with the Fe$^{3+}$-O-Fe$^{3+}$ superexchange interaction, which built-in ferromagnetic structure [3]. These sites can be substituted by metal ions from different elements and change the electrical and magnetic properties are sensitive to the microstructure and particle size, which can be modified by substitution of Fe in hexaferrite using ions from different elements and their concentrations [11]. There have been reported of various attempts to improve its magnetic properties of barium hexaferrite by replacing Fe$^{3+}$ with single metal ions such as Al$^{3+}$ [12], Ce$^{3+}$ [13], Dy$^{3+}$ [14], Cr$^{3+}$ [15], Zn$^{2+}$ [16], Co$^{2+}$ [17], Ga$^{3+}$ [18], and Ti$^{4+}$ [19]. They commonly discovered that doping metal ions in hexaferrite enhanced the coercivity. However, the coercivity improvement was made at the expense of reducing the saturation magnetization, limiting its use in various applications. The decreased saturation magnetization was attributed to the substitution of Fe$^{3+}$ ions by doped ions with non-magnetic or less magnetic moments than Fe$^{3+}$. The substitution partially obstructs and weakens the Fe$^{3+}$-O-Fe$^{3+}$ superexchange interaction, which decreases the magnetic moment in hexaferrite. Interestingly, aluminum ion-doped barium hexaferrite is the most intensively investigated in the last two decades among all metal ions. The Al$^{3+}$ doped barium hexaferrate at low concentration (x < 2) consistently decreases the saturation magnetization but could decrease [20] or increase [21–25] the coercivity. Doping Al$^{3+}$ at low concentrations tend to decrease the particle size of polycrystalline BaFe$_{12-x}$Al$_x$O$_{19}$, while high Al$^{3+}$ doping concentrations (x > 2) yield an amorphous structure for BaFe$_{12-x}$Al$_x$O$_{19}$ [26].

Many polymers with long hydrocarbon chain structures, such as polymethylmethacrylate (PMMA), ethylene glycol (EG), and polyethylene glycol (PEG), have been used as a surfactant, stabilizer, or dispersant to control the morphology, crystallinity, and properties of the nano hexaferrite [27–33]. It is believed that polymers with functional groups of carboxyl and hydroxyl easily interact with the free hydroxyl of metal oxide, facilitating nucleation and growth of hexaferrite. However, the final product’s microstructure and properties depend on the polymer’s type and concentration and other synthesis parameters. Xu et al prepared a hollow sphere of barium hexaferrite by ethylene glycol-assisted spray pyrolysis. EG addition to the sol solution enlarged the particle size, increased the coercivity and decrease the saturation magnetization. The decrease in saturation magnetization has been related to incomplete combustion of organics leaving behind carbon non-magnetic in the sample [27]. Adding PMMA as a template in the sol-gel synthesis of barium ferrite modified the morphology and particle size from irregular platelet shape to rod-like shape, enhancing both saturation magnetization and coercivity [28]. Lu et al prepared co-precipitated strontium ferrite by adding PEG 6000, which resulted in smaller particle size and better dispersibility [28]. The decrease in the particle size reduced the coercive field and saturation magnetization [29]. Han et al synthesized hexagonal ferrite using a sol-gel method with different weights of PEG 2000. They found that PEG addition up to 2 g reduces both coercivity and saturation magnetization. The prepared PEG samples were agglomerated to form a larger particle size than a single domain barium hexaferrite’s critical size value of 460 nm [30]. The decrease in saturation magnetization upon PEG addition was believed due to the samples containing hematite [31]. Durmus investigated the effect of PEG with a different molecular weight in the formation of barium hexaferrite by the sol-gel combustion method. He found a small increase in the saturation magnetization but considerably decreases in the coercive field for increasing PEG molecular weight from 400 to 10000 [32]. Cao et al prepared barium hexaferrite using a one-step hydrothermal process, mediated by PEG 10000 with different weights at 220 °C. Increasing the PEG weight in the precursor solution produced a well-defined hexagonal platelet barium hexaferrite and significantly enhanced the saturation magnetization with slightly decreased coercivity [33]. The synthesis of hexaferrite mediated PEG with higher molecular weight tends to produce better particle crystallinity and improve the saturation magnetization. Thus, PEG addition in the precursor solutions played an important role in controlling the surface morphology and facilitating the development of crystalline nucleation and growth to improve the hexaferrite properties.

In this work, sol-gel auto combustion and the calcination method were used to prepare BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ powders from metal nitrates with citric acid as the chelating agent and adding 1–2 g of PEG 6000 to the precursor solution. The effects of different PEG molar weights on the microstructure, particle nucleation, growth, and the magnetic properties of BaFe$_{10.4}$Al$_{1.6}$O$_{19}$, were discussed.

2. Experiment

2.1. Materials

Reagent-grade of barium nitrate Ba(NO$_3$)$_2$ (99.95%), ferric nitrate Fe(NO$_3$)$_3$,9H$_2$O (99.95%), aluminum nitrate Al(NO$_3$)$_3$,9H$_2$O (99.997%), citric acid C$_6$H$_8$O$_7$ (99.95%), and polyethylene glycol (PEG 6000) were purchased from Sigma-Aldrich. Double-distilled water was obtained from a local chemical store.
2.2. Method
Aluminum-substituted barium hexaferrite samples were synthesized via the sol-gel auto combustion method. According to the stoichiometric calculation, reagent-grade barium nitrate, ferric nitrate, and aluminum nitrate were suspended in double-distilled water. The citric acid was then added to the prepared aqueous solution to chelate the Ba$^{2+}$ and Fe$^{3+}$ ions. The molar ratio of nitrates to citric acid was set at 1:1. An appropriate amount of ammonia hydroxide solution was added to adjust the pH value to 8. A homogenous, transparent solution was achieved within a few minutes. Then polyethylene glycol in the amounts of 1 g, 1.5 g, and 2 g was added to the mixed precursor under continuous stirring. The mixed solutions were aged 24 hours at room temperature and then heated at 100 $^\circ$C on a hotplate until evaporation was complete. The brown gel formed was burned out by increasing the temperature to 150 $^\circ$C, resulting in loose powder. The last step was to calcine the loose powder in a furnace at 900 $^\circ$C for 2 h.

2.3. Characterizations
Crystalline phase analysis of the calcined powder was conducted using an x-ray diffractometer (XRD, Shimadzu Maxima-7000) using Cu Kα radiation ($\lambda = 1.5405$ Å). The morphology of the particles was observed via a scanning electron microscope (SEM, JEOL-JSM-6510LA). The magnetic properties were examined at room temperature in an applied field of up to 10 kOe using a vibrating sample magnetometer (VSM, OXFORD 1.2H). Energy dispersive spectroscopy (EDS, Hitachi S3000N Series) was used to observe chemical elements.

3. Results and discussion
Figure 1 depicts the XRD pattern of aluminum-substituted barium ferrite (BaFe$_{12-x}$Al$_x$O$_{19}$) synthesized without and with the addition of PEG and calcined at 900$^\circ$C for 2 h. The diffraction peaks are well indexed to the $P6_3/mmc$ space group, corresponding to the standard (ICPDS 51-1867) diffraction pattern for barium ferrite [34]. Furthermore, their relative intensities display a strong dependence on the PEG content. The sample synthesized without PEG produced a diffraction pattern resembling that of the single-crystal phase of barium hexaferrite, while the samples with PEG addition exhibited barium ferrite diffraction peaks, plus additional peaks corresponding to hematite, an intermediate compound ($\alpha$-Fe$_2$O$_3$, JCPDS 33-0664). Figure 1(a) displays the XRD pattern of the sample synthesized without the addition of PEG. No characteristic diffraction pattern of

![Figure 1. X-ray diffraction pattern of aluminum-substituted barium ferrite synthesized with PEG in the amount of (a) 0 g, (b) 1 g, (c) 1.5 g, and (d) 2 g.](image)
Al$^{3+}$ planes was observed at 2\(\theta\) = 45°, 58°, and 64°, indicating that Al$^{3+}$ ions entered the barium ferrite-substituted Fe$^{3+}$ lattice, forming a single-phase barium ferrite [35]. The confirmation of the aluminum content in the sample was examined using energy dispersive spectroscopy (EDS). Figure 2 shows the aluminum-substituted barium ferrite’s EDS spectrum synthesized without PEG and the corresponding elemental analysis. The results clearly show that the atomic percentages of elements in the BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ match the designated composition.

Figures 1(b)–(d) shows the samples’ diffraction patterns with PEG additions. Introducing PEG to the solution during synthesis yielded BaFe$_{12-x}$Al$_x$O$_{19}$ with hematite as a secondary crystalline phase. As the PEG content increased, the peak intensities of BaFe$_{12-x}$Al$_x$O$_{19}$ were enhanced, while the peak intensity of \(\alpha\)-Fe$_2$O$_3$ strengthened and then weakened, indicating that the PEG strongly affected crystalline formation and growth. Without PEG, citric acid in an equal molar ratio to the nitrates provided optimum fuel for producing single-phase BaFe$_{12-x}$Al$_x$O$_{19}$. Adding PEG provided extra fuel for the auto-combustion process, which generated additional heat to accelerate the formation of \(\alpha\)-Fe$_2$O$_3$ during calcination. At this stage, there were competing processes for crystalline formation and the grain growth in barium ferrite and hematite. Adding more PEG supplies more heat for crystallization and grain growth \(\alpha\)-Fe$_2$O$_3$ and BaFe$_{12-x}$Al$_x$O$_{19}$.

Figure 3 shows the SEM images of samples synthesized from solution with and without PEG additions using sol-gel auto combustion and calcination at 900 °C for 2 h. There is a clear microscopic evolution in crystalline shape and size, as increasing amounts of PEG were added to the solution. The solution containing no PEG and 1 g of PEG yielded similar rod-shaped microstructures. The addition of more PEG (1.5–2 g) produced irregularly-sized, plate-like microstructures. In all cases, the particle sizes increased as increasing amounts of PEG were added.

The morphology of the BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ particles without PEG added is shown in figure 3(a). Most of the particles are aggregated and interlaced in scattered positions. These rod-like particles had an average length and diameter of 147 nm and 34, respectively. For comparison, Ozah et al using the co-precipitation method, synthesized elongated hexagonal-shaped BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ with an average of about 90 nm in length and 53 nm in width [36]. Using the mechanochemical milling method, produced equiaxed BaFe$_{10.3}$Al$_{1.5}$O$_{19}$ with an average diameter particle size of about 50–150 nm [37]. Li et al used the sol-gel method, synthesized hexagonal, plate-like BaFe$_{10.5}$Al$_{1.5}$O$_{19}$ with an average diameter particle size of 200 nm [21]. Wang et al using the mechanical milling method produced a platelet-shaped BaFe$_{10}$Al$_{2}$O$_{19}$ with a typical size of 50 nm \(\times\) 20 nm [38]. Employing a similar auto-combustion process with a large amount of citric acid and a different pH produced nanorod-shaped BaFe$_{12-x}$Al$_x$O$_{19}$ with an average size of about 200 nm \(\times\) 58 nm for \(x = 1.5\), while, for \(x = 3\), disk-shaped BaFe$_{12-x}$Al$_x$O$_{19}$ was produced [39]. There is no unique method for producing specific particle shapes and sizes in the case of BaFe$_{12-x}$Al$_x$O$_{19}$. The chemical composition, heat treatment, and other parameters during preparation could affect the final product’s morphology. Figure 3(b) showed the calcined sample’s morphology when 1 g of PEG was added. BaFe$_{10.6}$Al$_{1.6}$O$_{19}$ has a rod-like shape with an average length and diameter of 318 nm and 57 nm. Increasing particle size with PEG in the solution is attributed to PEG’s role as a template and surfactant for the rapid nucleation and growth of BaFe$_{12-x}$Al$_x$O$_{19}$ [40]. However, increasing the amount of PEG to 1.5 g, hexagonal platelets of BaFe$_{12-x}$Al$_x$O$_{19}$ with an average diameter of about 387 nm were formed. As shown in the XRD data results, auto combustion using 1.5 g of PEG produces \(\alpha\)-Fe$_2$O$_3$ and BaFe$_{12-x}$Al$_x$O$_{19}$. When a sufficient amount of \(\alpha\)-Fe$_2$O$_3$ is present, it acts as an effective surfactant for the nucleation and growth of...
hexagonal platelets of $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{19}$, as shown in figure 3(c) \cite{41}. It has been reported that the critical size of single-domain, spherical barium hexaferrite is about 460 nm, above which the particle has multiple domains \cite{30}. Thus, the samples synthesized without and with 1–1.5 g of PEG having grain sizes smaller than 460 nm are single-domain barium ferrite particles. When PEG was increased to 2 g, the platelets of $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{19}$ grew to various sizes and were distributed in an inhomogeneous fashion, as shown in figure 3(d). The largest grains reached an average diameter of about 800 nm, perhaps forming multi-domain particles.

Figure 4 shows the typical hysteresis loops of the synthesized samples without and with PEG obtained at room temperature. The hysteresis loop of the synthesized sample without PEG has a smooth shape, exhibiting the typical signature of single-phase magnetic materials, while PEG additions produced wasp-waisted hysteresis loops around $H = 0$ due to the coexistence of $\alpha$-$\text{Fe}_2\text{O}_3$ and $\text{BaFe}_{10.4}\text{Al}_{1.6}\text{O}_{19}$ in the samples, as indicated by the XRD spectra in figure 1. Increasing the amount of PEG in the sol solution up to 1.5 g enlarged the hysteresis loop. The addition of 2 g of PEG shrunk the hysteresis loop, but it still more prominent than the hysteresis loop produced without the PEG addition. Since the magnetizations of all samples at 10 kOe did not achieve saturation, the saturation magnetizations, $M_s$, were calculated from the law of approach to saturation (LAS) for the high applied field sections of the hysteresis loops \cite{42,43}. According to the LAS, the magnetization’s dependence on the applied field is written as

$$M(H) = M_s(1 - \frac{A}{H} - \frac{B}{H^2} + CH)$$

(1)

where $M_s$ is the spontaneous magnetization and $H$ is the applied field. $A$ corresponds to the inhomogeneity in the microcrystals and can be ignored for high field regions. $B$ contributes to the overall anisotropy of the sample. For hexagonal ferrites, $B$ is proportional to the magneto-anisotropy constant $K_1$ and is given by \cite{44}

$$B = \frac{4}{15}\left(\frac{K_1}{M_s}\right)^2$$

(2)

$C$ is the high-field magnetic susceptibility and can be ignored at temperatures much lower than the Curie temperature. Linear fits of $M$ versus $1/H^2$ plot in the applied field region between 8 kOe and 10 kOe for each sample gave a straight line \cite{45}. This straight-line intercept with the M axis determines the saturation magnetization ($M_s$), and the slope determines the value of $B$. Using equation (2), the magneto-anisotropy
constant $K_1$ can be determined. The magnetic properties drawn from the hysteresis loops and the calculated data are tabulated in Table 1.

The values of $M_s$, $M_r$, and $H_c$ for the sample (BaFe$_{10.4}$Al$_{1.6}$O$_{19}$) synthesized without PEG in the sol solution are 13.81 emu g$^{-1}$, 7.63 emu g$^{-1}$, and 3.07 kOe, respectively, which are smaller than those for BaFe$_{10.5}$Al$_{1.5}$O$_{19}$ prepared using a similar method with sodium citrate as the chelating agent [46]. Lower values of the sample’s magnetic properties are attributed to citric acid’s ineffectiveness in chelating the metal ions compared to sodium citrate. Adding PEG up to 1.5 g, the $M_s$ and $M_r$ increased and reached their maximum values of 25.06 emu g$^{-1}$ and 13.32 emu g$^{-1}$, respectively, when 1.5 g of PEG was added. The increases in $M_s$ and $M_r$ are attributed to the increasing grain sizes of BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ [47]. As shown in figures 3(a)–(c), the samples’ grain sizes with 0–1.5 g of PEG were less than 460 nm in diameter, suggesting that they were in the single domain. In this region, the variation in $M_s$, independent of grain size and related to rotational processes only. When 2 g of PEG was added, the values of $M_s$ and $M_r$ decreased. The decrease in magnetization could be related to the homogeneity in grain size in the sample and domain wall motion since the sample has larger grain sizes within the multi-domain region. As shown in figure 3(d), the grains have various sizes. The inhomogeneity in grain size leads to a lower density for the sample, which reduces the magnetization [48].

When 1 g of PEG was added, the $H_c$ increased and reached a maximum value of 4.79 kOe, then continually decreased as PEG’s amount increased. This trend agrees with previous work on the relation between magnetization and domain states [49]. The increase in $H_c$ is correlated with the enlargement in the grain size the number of single-domain grains. As the PEG increased to 1.5 g, the $H_c$ decreased to the point where the grain size reached a single domain’s critical size. The decrease in $H_c$ continued through the multi-domain region as the domain wall appeared. The transition from a single-domain state to a multi-domain state resulted in a decrease in $H_c$ since the magnetization process for removing the domain walls became more energetically favorable than the individual atomic spins. Stoner and Wohlfarth stated that the coercive field ($H_c$) could be related to the saturation magnetization ($M_s$) and magnetocrystalline anisotropy constant ($K_1$) with $H_c \approx K_1/M_s$ [50]. Using the measured data for $H_c$ and $M_s$, the values of $K_1$ can be calculated, which are proportional to the values of $M_s$. The smaller value of $K_1$ observed with the 2 g PEG addition is attributed to the reduction in $M_s$ due to multi-domain formation of BaFe$_{10.4}$Al$_{1.6}$O$_{19}$. The squareness of the hysteresis loop ascribed to $M_s/M_r$ that can be related to the domain states. A $M_s/M_r$ ratio higher than 0.5 is a common sign of a single-domain state, and a $M_s/M_r$ ratio less than 0.5 is associated with a multi-domain state [51].

![Hysteresis loops of the samples synthesized with (a) no PEG and with PEG in the amounts of (b) 1 g, (c) 1.5 g, and (d) 2 g.](image)

Table 1. Magnetic properties of BaFe$_{10.4}$Al$_{1.6}$O$_{19}$.

| PEG (g) | $M_s$ (emu g$^{-1}$) | $H_c$ (kOe) | $M_r$ (emu g$^{-1}$) | $K_1 \times 10^5$ (erg cm$^{-3}$) | $M_s/M_r$ |
|---------|---------------------|------------|---------------------|--------------------------------|----------|
| 0       | 7.63 ± 0.2          | 3.07 ± 0.2 | 13.81 ± 0.2         | 4.2 ± 0.1                      | 0.55     |
| 1.0     | 10.81 ± 0.2         | 4.79 ± 0.2 | 19.17 ± 0.2         | 6.1 ± 0.1                      | 0.56     |
| 1.5     | 13.32 ± 0.2         | 3.77 ± 0.2 | 25.06 ± 0.2         | 8.7 ± 0.1                      | 0.53     |
| 2.0     | 11.64 ± 0.2         | 3.55 ± 0.2 | 23.71 ± 0.2         | 8.6 ± 0.1                      | 0.49     |
4. Conclusion

BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ powders were successfully prepared by the sol-gel auto-combustion method using PEG 6000. X-ray diffraction shows a single-phase of barium hexaferrite structure for BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ synthesized without PEG. PEG additions of 1.0 g in the solution lead to the formation of rod-like particles containing α-Fe$_2$O$_3$ and BaFe$_{10.4}$Al$_{1.6}$O$_{19}$. The rod-like particle size increases within the single domain region, enhancing the magnetization and the coercive field. The presence of α-Fe$_2$O$_3$ in the sample acts as a surfactant, accelerating the nucleation and growth of BaFe$_{10.4}$Al$_{1.6}$O$_{19}$. Adding more PEG of 1.5 g provides an excess heat source for growing the grain of BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ and thus increases the saturation magnetization. However, the grain size of BaFe$_{10.4}$Al$_{1.6}$O$_{19}$ reaches the boundary of single and multi-domain critical sizes, which decreases the coercivity. An excessive PEG addition of 2 g affected particle growth in the formation of multi-domains of BaFe$_{10.4}$Al$_{1.6}$O$_{19}$, leading to a decrease in its magnetic properties. Thus, adding PEG to the growth precursor can open up the possibility to fine-tune the microstructure and improving the magnetic properties of aluminum-doped barium hexaferrite.

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