Synthesis and characterization of Ba-Ferrite with variation of Nd$_2$O$_3$ additive by powder metallurgy method

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Abstract. Ba –ferrite is a material permanent magnet (PM) with formula stoichiometry BaFe$_{12}$O$_{19}$. In this experiment, that Ba -ferrite with additive 0%, 0.5% and 1,0 %wt. Nd$_2$O$_3$ have been made by using raw materials: BaCO$_3$ (p.a) and Fe$_2$O$_3$ (p.a). The raw materials and additive were mixed and milled by using a ball mill for 24 hours and milling media is aquadest. After that the samples were dried at 100°C for 6 hours and then the samples were calcined at temperature 1000°C for 2 hours by using electrical furnace. The calcined samples were milled by using a ball mill for 6 hours and sieved until passing 400 mesh. That after all the powders were formed by using hydraulic pressure machine at pressure 40 MPa to obtain a pellet. The pellet was sintered with heating rate 10°C / minute and at temperature 1150°C and holding time was 1 hour. According the characterization results show that the optimum of additive composition is 0.5%wt.Nd$_2$O$_3$, at this condition, the magnetic properties can increase about 40%, where remanence value (mr) is 33.72 emu/g, the coercivity value (Hc) is 2.907 kOe and flux magnetic value is 650 Gauss. According the xrd results shows that the peak of Nd$_2$O$_3$ phase does not appear at sample with 0.5 % Nd$_2$O$_3$, but it appears at sample with 1.0 % Nd$_2$O$_3$.

1. Introduction
The Ba- ferrite has attracted great interest in the magnetic material field and has been widely used as a permanent magnet because of its fairly large magnetocrystalline anisotropy and high Curie temperature[1,2]. The Ba-ferrite permanent magnets of general formula BaFe$_{12}$O$_{19}$ contain approximately 80% of iron oxide and 20% of Sr or Ba [3,4]. Good chemical stability, low specific weight, suitable magnetic characteristics, low price and chemical and mechanical resistance are the main benefits of these materials making them important materials as media for magnetic and magnetic–optical recording and components for reproducers, engines and generators in automobile industry [3,5]. The permanent magnet Ba -ferrite has been synthesized through various techniques i.e. co-precipitation, including sol–gel synthesis, the use of organic precursors or glass ceramic, citrate nitrate synthesis and ceramic processing methods [6]. Furthermore, Ba -ferrite was made from a standard ceramic routed and sintered between 1100 to 1350 °C to give a high coercivity [7,8]. Powder metallurgy is a means of manufacturing, without full melting, objects have properties which cannot be obtained by any other process. It is a technique that is applied for consolidation and formation of materials into useful products with different shapes, starting with powder mass. Powder metallurgy has played a vital role in solving many production problems, especially with those materials having
very high melting points [7]. In any case, the magnetic properties of these materials depend largely on the microstructure, and the microstructure depends on the preparation method. There are many methods to increase of magnetic properties, firstly by modification of grain size and microstructure and secondly by using additive [4,9]. It is well known that additives play an important role in improving the magnetic characteristics of ferrites. For instance, the role of small amounts of Ca, Si, Bi, La in improving the residual magnetization and the coercive force of Ba ferrite were investigated [10, 11, 12]. Moreover, magnetic properties of various oxides with spinel structures were investigated. It is possible to expect many effects from various additives such as Fe$^{3+}$ and Nd$^{3+}$ in ferrites [9]. Suriya Ounnunkad has reported that Magnetization (Ms) and coercive field (Hc) could be improved by substitutions of La or Pr ions on Ba ion basis sites, where, the Hc increases remarkably with increasing La or Pr ions content [13]. The magnetic properties of Ba-ferrite can be improved by the substitution of Ba$^{2+}$ and/or Fe$^{3+}$. In order to achieve this purpose, element substitution was widely used for the synthesis of magnetic materials with appropriate saturation magnetization and coercivity [1,9]. The purpose of this research is to synthesize and characterize Ba-ferrite material using Nd$_2$O$_3$ additive through powder metallurgy method and to know the effect of Nd$_2$O$_3$ composition on magnetic properties and Ba-ferrite crystal structure.

2. Experiment works

The raw materials used for the manufacture of Ba-ferrite with the stochiometric formula BaFe$_{12}$O$_{19}$ are pure grade powder (pa): BaCO$_3$, Fe$_2$O$_3$, and Nd$_2$O$_3$. Additive composition is varied: 0%, 0.5% and 1.0% wt. Nd$_2$O$_3$. The raw materials and additive were mixed and milled by using a ball mill for 24 hours and milling media is aquadest. After that the samples were dried at 100°C for 6 hours and then the samples were calcined at temperature 1000°C for 2 hours by using electrical furnace. The calcined samples were milled by using a ball mill for 6 hours and sieved until passing 400 mesh. That after all the powders were formed by using hydraulic pressure machine at pressure 40 MPa to obtain a pellet with diameter 18 mm and thickness 5 mm. The pellet was sintered with heating rate 10 °C / minute and at temperature 1150°C and holding time was 1 hour. After the sintering process, the pellet sample was magnetized using an impulse magnetizer with a voltage of 1300 Volt DC. Then Flux magnetic value was measured by using Gaussmeter and the hysteresis curve was measured by using Vibrating Sample Magnetometer (VSM) to know remanence, coercivity and energy product. The change in crystal structure was analyzed by using X Ray Diffractometer (XRD) -Rigaku with Cu-alpha target.

3. Results and Discussion

The sintered Ba-ferrite with variation additive Nd$_2$O$_3$ samples were magnetized at 1300 volt DC by using Impulse Magnetizer. Then Flux magnetic value was measured by using Gaussmeter and the hysteresis curve was measured by using VSM. The flux density measurement results can be seen in figure 1.

![Figure 1. Flux Magnetic curve of sintered Ba-ferrite as function % wt ratio of Nd$_2$O$_3$](image-url)
The flux magnetic value increases at 0% to 0.5% of additive Nd$_2$O$_3$ and decreases 0.5% to 1.0% of additive Nd$_2$O$_3$. The increasing or decreasing of flux magnetic value occurred due to possibility of changes in crystal size or in crystal lattice parameters. The maximum flux density value achieved from this experiment is 650 Gauss at sample with additive of 0.5% wt. Nd$_2$O$_3$. The figure 2 shows hysteresis curves of the sintered Ba-ferrite with variation additive Nd$_2$O$_3$ samples.

![Figure 2. Hysteresis Curves of Ba-ferrite with additive 0% Nd$_2$O$_3$, 0.5% Nd$_2$O$_3$ and 1.0% Nd$_2$O$_3$ after sintering at 1150°C.](image)

According to hysteresis curves at figure 2 show that the magnetic properties (mr and Hc) of sintered Ba-ferrite are increased due to presence of 0.5 wt.% of Nd$_2$O$_3$ and decreased against due to presence of 1.0 wt.% of Nd$_2$O$_3$. The highest magnetic properties are achieved at sample with 0.5 wt.% of Nd$_2$O$_3$, Where it has remanence value (mr) about 36 emu/g and coercivity value (Hc) about 3000 Oe. The analysis of crystall structure of sintered Ba-ferrite with variation additive Nd$_2$O$_3$ samples have been done by using XRD. The XRD patterns of all samples are shown at figure 3.

![Figure 3. The XRD patterns of sample Ba-Ferrite with additive: 0%Nd$_2$O$_3$, 0.5% Nd$_2$O$_3$ and 1.0% Nd$_2$O$_3$ after sintering at 1150°C.](image)
Based on XRD curve analysis as shown in figure 3 shows that all samples showed almost identical pattern, by matching experimental data with JCPDS card data, it was found only Ba-ferrite phase (BaFe$_{12}$O$_{19}$), no presence of other phase like Nd$_2$O$_3$. It is possible that the Nd atom diffuses into the structure BaFe$_{12}$O$_{19}$, so that there will be changes in crystal size and there is a shift in the diffraction angle at the highest peak, causing a change of crystal lattice size. This can lead to changes in magnetic properties.

The crystallite size ($L$ in nm) of Ba-ferrite phase were calculated from most intense peaks in XRD patterns using the Scherrer equation

$$ L = \frac{k \lambda}{B \cos \theta} $$

Here, “k” is Scherrer constant and its value is ‘0.94’ for our material, ‘B’ is full width at half maximum (FWHM) of respective peaks, ‘$\theta$’ is Bragg’s angle and ‘$\lambda$’ is wavelength of Cu-k $\alpha$ radiation (1.5406 Å) used during the XRD analysis of the samples [9]. The calculated value of crystallite size of Ba-ferrite samples with different percentage of Nd$_2$O$_3$ are given in table 1.

| Name sample            | 2 theta at highest peak (deg) | FWHM (deg) | Crystal; size (nm) |
|------------------------|-------------------------------|------------|--------------------|
| Sample with 0 % Ba-ferrite | 35.372                       | 0.107      | 815                |
| Sample with 0.5 % Ba-ferrite | 35.349                       | 0.127      | 697                |
| Sample with 1.0 % Ba-ferrite | 35.361                       | 0.108      | 806                |

The additive effect of Nd$_2$O$_3$ on the preparation of Ba-ferrite can affect the size of the crystal, which by adding 0.5% by weight can decrease the crystallite size from 815 nm to 697 nm. But the size of the crystal enlarges back to 806 nm with the addition of 1.0 wt.%. The presence of changes in crystal size due to the addition of Nd$_2$O$_3$ may contribute to changes in magnetic properties. By decreasing the crystal size of Ba-ferrite from 815 nm to 697 nm, magnetic properties, especially magnetic remanence (mr), will increase from 28 emu/g to 36 emu/g. If the crystal size of Ba-ferrite approaches the size of a single domain crystal Ba-ferrite (460nm) [14] then there is a tendency to increase the magnetic properties. From this research it is found that with the addition of Nd2O3 as much as 1%, then there is an increase of Ba-ferrite crystal size so that it has an effect on the decreasing of its magnetic properties.

4. Conclusions

Permanent magnet Ba-ferrite with additive Nd$_2$O$_3$ has been successfully made through powder metallurgy technique with 1150ºC sintering temperature. The percentage addition of Nd$_2$O$_3$ can have a significant effect on magnetic properties, crystal structure and crystal size. In particular, the addition of 0.5 % wt. of Nd$_2$O$_3$ to Ba-ferrite had a significant role in reduction of crystal size , but if the amount of Nd$_2$O$_3$ additive exceeds 0.5%, then it will increase the crystal size of Ba-ferrit. This causes a decrease in the magnetic properties of Ba-ferrite. The optimum composition of the Nd$_2$O$_3$ additive is 0.5% by weight, in this composition the permanent magnet is obtained with the properties of: flux magnetic value = 650 Gauss, magnetic remanence (mr) = 36 emu/g, coercivity = 3000 Oe and crystal size = 697nm.
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