The Effect of Composition and Sintering Temperature on Magnetic Properties of Ba-Hexaferrite

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Abstract

The hard magnetic Barium Hexaferrite was made from BaCO\textsubscript{3} and Fe\textsubscript{3}O\textsubscript{4} powders at various compositions of BaCO\textsubscript{3} and Fe\textsubscript{3}O\textsubscript{4} with stoichiometric ratio of 1:6 and non-stoichiometric ratio of 1:6:5. The preparation was done by wet mixing and grinding using PBM. The powders were calcined at 1000°C for 2 hours and subsequently being analyzed using XRD to observe the structure of BaO.6Fe\textsubscript{3}O\textsubscript{4}. The powder was then sieved to pass 400 mesh (38 \mu m) followed by the addition of 3wt% printing Seluna as an adhesive. The printing was done by using a magnetic anisotropy field press and pressed at 5 tons at a temperature of 1120°C, 1150°C, and 1170°C for 2 hours respectively. The results showed that the non stoichiometric ratio was better than a stoichiometric ratio. A magnetic field strength of 544.2 Gauss with a density = 4.0 g/cm\textsuperscript{3}, porosity = 4.06%, Remanence (Br) = 1.72 kG, Coercivity (Hc) = 2.41 kOe, and Energy product (BH max) = 0.63 MGOe were achieved when the magnet sintered at 1170°C. It was found that the addition of 0.5-mole\% Fe\textsubscript{3}O\textsubscript{4} enhanced the density of the magnet.

Keywords: Hard Magnetic, BaO.6Fe\textsubscript{3}O\textsubscript{4}, XRD, calcined.

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1. Introduction

A permanent magnet should be able to generate a high magnetic flux of a given magnet volume, good magnetic stability against the effects of temperature and time, and high resistance to demagnetization effects. In principle, permanent magnet must have a minimum of the magnetic characteristics: high remanence (Br), high intrinsic coercivity (Hc) and high Curie temperature (Tc) [1]. A composite magnet based on ferrite has a wide application [2,3]. It is known that Barium hexaferrite is one of the most promising materials for high-density magnetic material due to their unique magnetic characteristics, namely, high coercivity, moderate magnetic moment, low or positive temperature coefficient of coercivity, and high chemical stability.

Barium Hexaferrite of type-M, which is better known as hexagonal barium ferrite (BAM) is a ceramic oxide and most widely used commercially and has been chosen as a material research to develop in terms of practical use until today. Barium Hexaferrite is an anisotropic magnetic material with a high field [4]. Hence, it can be used at a higher frequency than the ferrite spinel or garnet (above 30 GHz). Anisotropic magnetic crystals were obtained from crystal with high anisotropic structure. The crystal structure of the grain growth is also anisotropic, with hexagonal morphology as areas that provide improved anisotropic. As a result, BAM produces high coercivity. Many techniques [5-7] have been developed as well as the addition of doping have been added in [8-9] to the precursor. In this research, we tried to investigate the effect of precursor composition. In the first composition, we made a normal ratio of 1:6, whilst another composition we made with a ratio of 1:6.5. By addition of 0.5 mole% Fe$_2$O$_3$, we hope an enhancement either in physical properties or magnetic properties.

2. Experiment procedure

In this study, a permanent magnet Barium Hexaferrite was made by using the powder metallurgy method by combination of some oxide in powder form (milling process), calcinations, compaction and sintering. The raw materials used were BaCO$_3$ and Fe$_2$O$_3$ (technically raw). In the first route we combine: BaCO$_3$ + 6Fe$_2$O$_3$ to produce BaFe$_{12}$O$_{19}$, whereas in the second route we combine: BaCO$_3$ + 6.5Fe$_2$O$_3$ to produce BaFe$_{12}$O$_{19}$. The powder was then mixed by using a Planetary Ball Milling (PBM) in wet milling for 20 hours. The powder was then dried at 100°C for 24 hours. The dried powder had then sintered at 1000°C before grinding by using a mortar to pass a particle size of 400 mesh (38 μm). A pressure of 5 ton with anisotropic compaction was applied to form a pellet (by adding 3% Seluna WE - 518 of Barium Hexaferrite) with diameter of 5 cm and 2 mm in thickness. The pellet was then sintered at: 1120°C, 1150°C and 1170°C in an electrical furnace for 2 hours respectively. The sintered pellet was then characterized in order to determine the physical properties. Phase identification was performed by X-ray powder diffraction (XRD) method with Cu Kα radiation with 2θ = 10° to 80°. The pellet flux density was measured by using a Gaussmeter whereas BH curve was obtained by using Permagraph.

3. Results and discussion

The XRD pattern as shown in figure 1 was used in the determination of the major and minor phase occurrence after being sintered at 1000°C and retained for 2 hours.
Figure 1: X-ray diffraction pattern of permanent magnet Barium Hexaferrite

From Figure 1, it can be seen that the major phase is Barium Hexaferrite (BaO.6Fe₂O₃) and the minor phase is Hematite (Fe₂O₃). In the composition of 1:6, it was found that 90.9% of (BaO.6Fe₂O₃) phase occurred at 816°C, whilst 9.1% of Hematite (Fe₂O₃) was observed. On the other hand, the composition of 1:6.5 indicates that 80.8% major phase was Barium Hexaferrite (at 810°C) with 18.2% Hematite phase. The graph shows the first four maximum peaks occur at 30° ≤ 2θ ≤ 40°. This implies that the characteristic peaks corresponding to the barium hexaferrite structure as a major phase. This confirms the complete conversion of the precursor powder into BaFe₁₂O₁₉ [10].

Figure 2 below shows that the increasing in sintering temperature causes the density values (bulk density) tend to increase due to diffusion process, and sintering temperature may result in enhancement of grain growth so that the pores among the grains can be reduced more and more. The greater shrinkage occurs when the sintering temperature increases causing the flux density increases as the bonding among the particles are getting stronger.

Figure 2: The effect of sintering temperature on the density and porosity of the magnet
From Figure 3 above, it can be seen that the addition of raw materials of 0.5% Fe₂O₃ (% mol) of stoichiometric calculations can increase the magnetic field strength. A value of 508.10 gauss in magnetic field strength (stoichiometric) and 544.20 gauss in non-stoichiometric were obtained at 1170°C. This is due to fact that only the Fe⁺⁺ ions have a magnetic property and a densification of the magnet at this temperature. However, the obtained magnetic field strength is less than previous finding. Sudirman et al. [11] reported a range of (765-1072) gauss can be achieved by addition of coupling agent 3-APE. The difference might be due to the sample preparation as well as the addition of the coupling agent. The hysteresis curva of the magnet as can be presented in below Figure 4.

Figure 4: Hysteresis curve of 1 : 6 composition (a) and a composition of 1 : 6.5 (b)
From Figure 4 it can be seen that the BH curve has a significant narrowing of the curve that indicates the decreasing of coercivity and increasing of magnetic remanence. The result of physical and magnetic properties is shown in below Table 1 and Table 2 respectively.

**Table 1: Physical properties of BaO.6Fe$_2$O$_3$**

<table>
<thead>
<tr>
<th>Sintering Temp. ($^\circ$C)</th>
<th>Composition</th>
<th>1 : 6</th>
<th>1 : 6.5</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\rho$ (g/cm$^3$)</td>
<td>Porosity (%)</td>
<td>$\rho$ (g/cm$^3$)</td>
</tr>
<tr>
<td>1120</td>
<td>3.13</td>
<td>10.50</td>
<td>3.01</td>
</tr>
<tr>
<td>1150</td>
<td>3.64</td>
<td>7.62</td>
<td>3.9</td>
</tr>
<tr>
<td>1170</td>
<td>3.97</td>
<td>7.25</td>
<td>4.00</td>
</tr>
</tbody>
</table>

From Table 1, it can be seen that a little enhancement in porosity as well as density was achieved with composition of 1 : 6.5. If the commercial Ba-ferrite has a density of 5.1 g/cm$^3$ [12], then this ratio at sintering temperature of 1170$^\circ$C is reasonable to be developed to a specific purpose.

**Table 2: Magnetic properties of BaO.6Fe$_2$O$_3$**

<table>
<thead>
<tr>
<th>Sintering Temp. ($^\circ$C)</th>
<th>Composition</th>
<th>1 : 6</th>
<th>1 : 6.5</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Br (kG)</td>
<td>HcJ (kOe)</td>
<td>$BH_{max}$ (MGOe)</td>
</tr>
<tr>
<td>1120</td>
<td>1.19</td>
<td>5.257</td>
<td>0.30</td>
</tr>
<tr>
<td>1150</td>
<td>1.57</td>
<td>3.104</td>
<td>0.54</td>
</tr>
<tr>
<td>1170</td>
<td>1.69</td>
<td>2.577</td>
<td>0.61</td>
</tr>
</tbody>
</table>

From Table 2 above, it can be seen that a maximum remanence value was obtained when the magnet sintered at temperature 1700$^\circ$C (1 : 6.5) is around 1.72 kG with a coercivity value of 2.418 kOe and energy product value of 0.63 MGOe. According to Prijo Sarjono et al. [13], a permanent magnet in water flow meter application should have a value of remanence (Br) = 2.45 kG, coercivity (Hc) = 135.2 kA/m (1.7 kOe) and energy products ($BH_{max}$) = 1.13 MGOe. Thus, this composition is still reasonable to be explored in the future since its remanence is 70.2% of applied permanent magnet. However, all the magnetic values are in the range of
commercial Ba-ferrite [12].

4. Conclusion

We compared the magnetic properties of Barium Hexaferrite by means of the precursor ratio. The phase occurrence has been formed at lower temperature with a composition of 1:6.5. The addition of 0.5-mole% Fe₂O₃ in this composition can be used as a densification agent. The optimum physical properties as well as optimum magnetic properties were achieved when this composition had been sintered at 1170°C.

5. Recommendation

The composition of 1:6.5 can be referred as a nominal starting composition in the future work. The enhancement in physical as well as magnetic properties would be achieved by using nanosized Barium Hexaferrite powders and will be presented elsewhere.

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