Effect of Al2O3 addition with concentration (x=0.1%, 0.2%, 0.4%) mol on the crystal structure and physical properties of permanent magnets barium hexaferrite (BaO6Fe2O3)

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Abstract. The effects of Al2O3 addition with various concentration (x=0.1%, 0.2%, 0.4%) mol on the change of crystal structure and magnetic properties in Barium Hexaferrite (BaO6Fe2O3) has been made and analyzed by bulk metallurgy processes with raw material element BaCO3, Fe2O3 and Al2O3. Raw materials are mixed and calcined at 1150°C for 1 hour. After calcinations, the powder is mixed with 3% seluna, then press with a compressive force of 8000 kgf. Physical properties analysis is obtained from measurements of density and porosity. While the material phase structure is determined by XRD test and for magnetic properties using VSM. In the XRD test results, it can be seen that the sample is dominated by BaO6Fe2O3 phase and Al2O3 phase, with the dominant phase being BaO6Fe2O3. The evaluation results obtained the best value at a concentration of x=0.4% (gr/mol) with an optimum density of 4.13 gr/cm3 which has a porosity of 1.3% and for magnetic properties and material structure at x = 0.4% mol a remanence value of 2.02 emu/g is produced with coercivity of 0.531 kOe.

1. Introduction
Barium ferrites are well known hard magnetic materials, which are based on iron oxide. They are also called as ferrite magnets and could not be easily replaced by any other magnets [1,4]. Ferrite permanent magnet also known as ceramic magnet began to be developed in 1950 and began production in 1952 by Philips with the name Ferroxdure production as one of the results of the Stoner-Wohlfarth theory [2]. Barium ferrite possesses relatively high Curie temperature, coercive force and magnetic anisotropy field, as well as its excellent chemical stability and corrosion resistivity [3]. The Barium ferrite with hexagonal structure and molecular formula BaO6Fe2O3 (BaFe12O19) has a magnetoplumbiten structure, which are widely used in magnetic recording media, microwave devices and electromagnetic shielding fields [4]. As a result of their moderate coercivity, hexagonal barium ferrite are suitable for magnetic recording media application [5]. Moreover, due to their high anisotropy field, hexaferrite can be used at much higher frequencies than spinel ferrites or garnets [6,7]. Many methods of synthesis have been developed to obtain a low production cost of powder...
particles of Barium ferrite [8]. Powder metallurgy methods are used more frequently because they are relatively economical and easy to do [9]. There are several steps and methods to make permanent magnet better and superior, so this test is only limited to the effect of adding Al₂O₃ to find out and get coercivity, remanent and maximum energy product and pore concentrations seen from the change in density [10]. The purpose of this research is to determine the effect of Al₂O₃ addition on the crystal structure, physical properties (bulk density, porosity) and magnetic properties (remanence, coercivity and magnetic saturation) by using VSM and crystal structure by using XRD of permanent magnets Barium heksaferite (BaO₆Fe₂O₃) [11].

2. Experimental
For synthesis of BaO₆Fe₂O₃ (BaFe₁₂O₁₉), mixture of iron oxide Fe₂O₃ (99% purity) and barium carbonate BaCO₃ (99% purity) [11].

| Table 1. Composition for powder preparation  BaO₆Fe₂O₃ doped by Al₂O₃ |
|-----------------|-------|----------------|-----------------|
| Al₂O₃ (%mol)    | BaO (g) | Fe₂O₃ (g)    | Al₂O₃ (g)       |
| 0.1             | 2.062  | 12.94        | 0.002           |
| 0.2             | 2.062  | 12.93        | 0.005           |
| 0.4             | 2.062  | 12.92        | 0.0010          |

After preparation, powder in calcinations at temperature 1150 degree Celcius for 1 hour. Calcinations is useful for converting BaCO₃ compounds BaO by removing the CO₂. After calcinations, the powder is mixed with 3% seluna, then press with a compressive force of 8000 kgf. The powder is sintering at temperature 1150ºC for 1 hour. Physical properties analysis is obtained from measurements of density and porosity. While the material phase structure is determined by XRD test and for magnetic properties using VSM. The complete methodology can be seen in Figure 1.

3. Results and Discussions
From the result of the study found that the value of the bulk density and porosity after sintering can be seen in Figure 2. Figure 2 below shows that the increasing in addition of raw materials of Al₂O₃ cause the density values (bulk density) tend to increases due to diffusion process and addition of raw materials of Al₂O₃ may result in enhancement of grain growth so that the pores among the grains can be reduced more and more [9]. The greater shrinkage occurs when the raw materials Al₂O₃ increases causing the flux density increased as the bonding among the particles are getting stronger [8].
From Figure 2, the evaluation results obtained the best value at a concentration of x=0.4% (gr/mol) with an optimum density of 4.13 gr/cm$^3$ which has a porosity of 1.3%. The XRD pattern as shown in Figure 3 was used in the determination of the major and minor phase occurrence after being sintered at 1150°C.

From Figure 3, it can be seen that the major phase is Barium Hexaferrite ($\text{BaO}_6\text{Fe}_2\text{O}_3$) and the minor phase is hematite ($\text{Al}_2\text{O}_3$). From the result of the XRD analysis, from the 0.1% additive graph, it can be seen that the intensity for all 2θ values is still relatively small, which means that the polycrystalline structure has not yet been formed but is still dominated by amorphous structures [6]. A vibrating sample magnetometer (VSM) was used to obtain magnetic property. VSM measurements were done on an unoriented, random assembly of particles at room temperature and with a maximum applied field of 16 kOe [9]. For each measurement, a hysteresis loop was generated from which the intrinsic coercivity ($H_c$) and saturation magnetization ($\sigma_s$) were calculated [3]. Figure 4 is a plot hysteresis curve of sample $\text{BaFe}_{12-4x}\text{Al}_{4x}\text{O}_{19}$. 
Figure 4. Hysteresis curve of sample Ba-Ferrite with variation of concentration Al₂O₃

From Figure 4, it can be seen that the BHmax curve has a significant narrowing of the curve that indicates the decreasing of coercivity and increasing of magnetic remanence. The result of magnetic properties is shown in below Table 2.

| Concentration Al₂O₃ (% mol) | Coercivity (kOe) | Remanence (emu/gr) | Ms (emu/g) | BHmax (MGOe) |
|-----------------------------|------------------|--------------------|------------|--------------|
| 0.1                         | 3.079            | 25.54              | 57.44      | 20.123       |
| 0.2                         | 0.597            | 2.40               | 39.11      | 20.265       |
| 0.4                         | 0.531            | 2.02               | 44.52      | 20.288       |

From table 2, it can be seen that a maximum remanence value was obtained when the magnet additive at 0.1% mol is around 25.54 emu/gr with a coercivity value of 3.079 kOe and energy product value of 20.123 MGOe. From the result of the VSM analysis, it can be seen that the increasing Alumina (Al₂O₃) content added and assisted with the sintering process will increase the maximum energy product in the barium hexaferrite magnetic [6].

4. Conclusions
From result and discussion, effect of Al₂O₃ on the crystal structure and physical properties of permanent magnets Ba-Ferrite (BaO₆Fe₂O₁₉) is the greater of alumina added and assisted with the sintering process will reduce the pore content of the material [6]. This is possible Al₂O₃ material which has a relatively small grain size is able to insert between fragments of (BaO₆Fe₂O₁₉) material. Other that, additive of Al₂O₃ will increase the maximum energy product in the barium hexaferrite magnet [8]. From result VSM analysis, the highest energy product value was reached at 20.288 MGOe after being addition of Al₂O₃ with concentration 0.4% mol.

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