Ferrite-based material as a permanent magnet for components of electrical generators

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Abstract

A permanent magnet based on ferrite for use as components of electrical generators has been fabricated by two different methods: solid–solid mixing and cooprecipitation. The conventional solid–solid mixing method uses Fe$_2$O$_3$ fabricated by two different methods: solid–solid mixing and cooprecipitation. The ferrite-based material as a permanent magnet for components of electrical generators has been optimally used. Therefore technology transfer and/or technology development associated with the processing of iron sand into final useful products is needed.

1. Introduction

Permanent magnets were originally produced from metals and then by using metal alloys such as SmCo-magnets and AlNiCo-magnets. With further development, magnetic oxide and ceramic materials called hard ferrites have been fabricated [1]. This type of ceramic ferrite includes SrO·6Fe$_2$O$_3$, PbO·6Fe$_2$O$_3$ and BaO·6Fe$_2$O$_3$. Ferrite permanent magnets based on BaO and Fe$_2$O$_3$ are widely used as electromotor components, seals, filters and so on.

The requirement for permanent magnets in Indonesian industry is quite high. Currently they are still imported from Japan and China. Magnet manufacture does exist in Indonesia, for example in the area of Cilegon under the name of PT Sumimagne (since changed to Neo Max Indonesia). This company began to produce permanent magnets from the raw material in the form of scrap from PT Krakatau Steel. Indonesia is rich in natural resources and has relatively abundant natural minerals, in particular iron sand as Fe$_2$O$_3$ source which could be a raw material for the manufacture of magnets, but up to the present day it still has not been optimally used. Therefore technology transfer and/or technology development associated with the processing of iron sand into final useful products is needed.

2. Theoretical background

There are several factors affecting the magnetic properties of a material. They are the purity of the material, its composition, grain size refinement, etc. One of the most...
important parameters to be discussed here is grain size. It is known that the magnetic properties of a material with fine particle size (of the order of nanometers) could approach those of single magnetic domains [2]. In addition, nanoparticles might accelerate the sintering process, hence the contact between the particle surface area will be larger as well [1].

The conventional solid–solid mixing method using the ball mill technique is only capable of fabricating permanent magnet powder with particle size up to 10 µm. For example, the manufacture of a BaO·6Fe₂O₃ permanent magnet by means of the solid–solid mixing method and at sintering temperatures in the range 1250–1300 °C is able to produce magnetic particles with a grain size of approximately 1–2 µm [3]. Therefore, in order to prepare material with nanosized particles, several other different techniques such as wet chemistry (cooprecipitation), sol–gel, chemical vapor deposition (CVD), etc should be used.

3. Experimental

Materials for a ferrite-based permanent magnet used as a core component of electric generators have been prepared by using solid–solid mixing as well as cooprecipitation methods. In the solid–solid mixing process, starting materials BaCO₃ and Fe₂O₃ are used to fabricate BaO·6Fe₂O₃. The powder preparation process consists of the following stages: mixing two starting materials with distilled water, milling the mixture by ball mill for 24 h, drying at 100 °C for 12 h and calcining at 1100 °C to produce BaO·6Fe₂O₃ powder. The process is as follows:

\[ \text{BaCO}_3 + 6\text{Fe}_2\text{O}_3 \xrightarrow{1100\,^\circ\text{C}} \text{BaO} \cdot 6\text{Fe}_2\text{O}_3 + \text{CO}_2. \quad (1) \]

Characterization of the prepared powder was performed using x-ray diffraction (XRD) and a scanning electron microscope (SEM). Furthermore, in order to test the product in bulk form, polyvinyl alcohol (PVA) as adhesive material was added to the powder with a proportion of 3% by weight. The sample was pelletized by dry pressing with 2 ton pressure, sintered at a temperature of 1100 °C using an electric furnace and held for 2 h. Calcination method used BaCl₂ and FeCl₃ as starting materials with precursor distilled water, then a solution of ammonia with concentration of 1 : 1 was added until a pH value of 8 was obtained. During the addition of ammonia solution, the precipitate was filtered and washed with distilled water to remove chlorine. Once the sediment was free from chlorine, the washed filtrate was then tested by adding a solution of AgNO₃. It turns out that the filtrate does not show the existence of a white AgCl precipitate, as stated in the following reactions:

\[ \text{BaCl}_2 + 2\text{NH}_4\text{OH} \rightarrow 2\text{NH}_4\text{Cl} + \text{Ba(OH)}_2, \quad (2) \]

\[ \text{FeCl}_3 + 3\text{NH}_4\text{OH} \rightarrow 3\text{NH}_4\text{Cl} + \text{Fe(OH)}_3. \quad (3) \]

The precipitate formed by reactions (1) and (2) is a mixture of Ba(OH)₂ and Fe(OH)₃. This mixture is then dried at a temperature of 100 °C for 12 h and pulverized in a 400 mesh sieve. To produce the hexagonal phase structure of BaO·6Fe₂O₃ ferrite it is necessary to undergo a calcination process with following formation reaction:

\[ \text{Ba(OH)}_2 + 12\text{Fe(OH)}_3 \xrightarrow{T=900\,^\circ\text{C}} 19\text{H}_2\text{O} + \text{BaO} \cdot 6\text{Fe}_2\text{O}_3. \quad (4) \]

It is important to determine the composition of the mixture for the formation of BaO·6Fe₂O₃ hexagonal ferrite crystal: the ratio is BaO : Fe₂O₃ = 1 : 6. The microstructure of the BaO·6Fe₂O₃ powder was determined by XRD. The obtained samples still have a green color. They are then burned using the electrical furnace at a temperature of 1050 °C and detained for 2 h.

Sintered samples were characterized by density and porosity measurements as well as by XRD analysis. To study the magnetic properties of the samples they were magnetized in a magnetic field with strength in the range 10–50 kGauss. The hysteresis data were recorded.

4. Results and discussion

To measure the temperature of calcination (heat treatment) the powder obtained from the coacprecipitation process was analyzed by differential thermal analysis (DTA) and thermogravimetric analysis (TGA). The results are shown in figure 1.

There are two endothermic peaks at temperatures of 426 °C and 823 °C. The first one corresponds to the release of hydroxide groups and the formation of a compound oxide from BaO and Fe₂O₃. The second one shows that a reaction took place between the oxides Fe₂O₃ and BaO to form a new compound (BaO·6Fe₂O₃). On the basis of these data we chose the calcination temperature of the powder obtained from the coacprecipitation process to be 900 °C or higher.

For convenience of presentation of the experimental results each sample prepared by the solid–solid mixing method is called sample A, while that prepared by the coacprecipitation method is called sample B. Both samples A and B sintered at a temperature of 900 °C were analyzed by XRD (the results were not shown). The results of the XRD analysis of sample A sintered at a temperature of 1100 °C and
Figure 2. XRD patterns of (a) sample A sintered at 1100 °C for 2 hours and (b) sample B sintered at 1050 °C for 2 hours.

Figure 3. SEM images of (a) the sample A sintered at 1100 °C for 2 hours and (b) the sample B sintered at 1050 °C for 2 hours.

Figure 4. Density and porosity versus sintering temperature: (a) solid–solid mixing method and (b) co-precipitation method.

of sample B sintered at a temperature of 1050 °C for 2 h are shown in figures 2(a) and (b).

These XRD patterns showed that in the material of sample A sintered at 1100 °C there co-exist two phases, BaO·6Fe2O3 and BaFe12O19, while in the material of sample B sintered at 1050 °C there exists only a single phase, BaO·6Fe2O3.

SEM images of sample A sintered at 1100 °C and sample B sintered at 1050 °C are shown in figures 3(a) and (b).

These SEM images showed that the powder particles in sample A sintered at 1100 °C have a grain size in the range 0.5–1.5 µm, while those in sample B sintered at 1050 °C have a grain size of the order 30–60 nm.

Different samples A and B have been prepared, sintered at temperatures of 900, 950, 1000, 1050 and 1100 °C and held at the temperature (holding time) for 2 h. The results of the measurements of physical properties such as density and porosity are shown in figure 4. From figure 4(a) it is clear that with the increase of sintering temperature the density of the material in sample A tends to increase while its porosity tends to decrease until a sintering temperature of 1100 °C. At this temperature the density and the porosity are 3.43 g cm−3 and 9.16%, respectively. These results suggest that the optimum sintering temperature of the solid–solid mixing method had still not been achieved. On the other hand, the curves in figure 4(b) showed that with the increase of sintering temperature the density of the material in sample B sharply increases at temperatures in the range 900–1000 °C, while its porosity sharply decreases in this same temperature range. Meanwhile, at temperatures higher than 1000 or 1050 °C the values of the materials in sample B are kept almost constant—their variations are very small. The optimum sintering temperature of sample B is about 1050 °C and the corresponding values of the density and porosity are 4.34 g cm−3 and 3.36%, respectively.
Figure 5. Hysteresis curve measurement result from sample A prepared with solid–solid mixing and B by the cooprecipitation method sintered at temperatures of (a) 1100 °C and (b) 1050 °C.

Table 1. Magnetic properties of sample A (solid–solid mixing) and sample B (cooprecipitation method) with sintering temperature at 1100 °C and 1050 °C.

| Parameter               | Sample code |
|-------------------------|-------------|
| Saturation magnetization, Ms | A 47.72 emu g⁻¹ | B 63.4 emu g⁻¹ |
| Remanent magnetization, Mr | A 18.30 emu g⁻¹ | B 39.7 emu g⁻¹ |
| Mr/Ms                   | A 0.383     | B 0.626 |
| Remanence, Br           | A 0.0792 T  | B 0.2673 T |
| Magnetic saturation, Bs  | A 1.21 T    | B 1.42 T |
| Coercivity, Hc          | A 44.7 kA m⁻¹ | B 89.4 kA m⁻¹ |

According to [4] the density of barium ferrite has an approximate theoretical value of 5.3 g cm⁻³. Thus our experimental results have attained only up to 82% of the theoretical result. There might be different causes: the time duration for the formation of the crystal is not long enough, the material should be pelletized with the pressure higher than 3 tons, etc.

Hysteresis curves of sample A sintered at a temperature of 1100 °C and sample B sintered at a temperature of 1050 °C are shown in figure 5. [MH] curves of these samples have different widths. The sample with broader [MH] curve has greater coercive force. The narrow width of the [MH] curve of sample A is consistent with the above-mentioned co-existence of two different phases in the material of sample A: the magnetic crystalline phase BaO · 6Fe₂O₃ and the non-magnetic phase BaFe₁₂O₁₉ which tends to be soft magnetic. Because the material of sample B has only a single crystalline phase BaO · 6Fe₂O₃, its [MH] curve has a much broader width.

Different magnetic parameters of both samples A and B were measured using a vibrating sample magnetometer (VSM). The measurement results of saturation and remanent saturation Ms and Mr, remanence Br, magnetic saturation Bs and coercivity are presented in table 1.

It is worth comparing the values in table 1 with those in [1] for a magnet with density 5.3 g cm⁻³ sintered at temperatures in the range 1200–1300 °C: Br = 0.2–0.5 T and Hc = 200–500 kA m⁻¹.

5. Conclusion

Materials for a permanent magnet to be used as a component of electrical generators were prepared by two different methods: solid–solid mixing followed by a ball mill technique which produces powder with particle size in the micrometer range and the cooprecipitation method which prepares powder with a particle size in the range 30–60 nm. The nanostructure of the material clearly affects the physical properties of the prepared magnets. Moreover, in the material prepared by the solid–solid mixing method there co-exist two different phases—magnetic crystalline phase BaO · 6Fe₂O₃ and non-magnetic phase BaFe₁₂O₁₉—while the material prepared by the cooprecipitation method has only a single crystalline phase—BaO · 6Fe₂O₃. Therefore the hysteresis [MH] curve of the material prepared by the second method has a much wider width than that prepared by the first method. The samples obtained by both methods were characterized by XRD, SEM and DTA/TGA techniques. Their magnetic parameters were also measured. The results of the material characterizations and the measurements of the physical parameters were presented. Moreover, the optimum conditions for preparation of the crystal structure BaO · 6Fe₂O₃ were established: the material is prepared by the cooprecipitation method, the sintering temperature is 1050 °C and the material prepared at this optimum condition then has the following physical parameters: Br = 0.2673 T; Bs = 1.42 T; Hc = 89.4 kA m⁻¹; density 4.34 g cm⁻³ and porosity 3.36%.

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