The Effect of Composite Bonded Magnet NdFeB/BaFe$_{12}$O$_{19}$ Composition with an Addition of Bakelite to Physical and Magnetic Properties

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Abstract. Composite bonded magnet becomes one of interesting magnetic materials based on its mechanical and magnetic characteristics. Besides that, the application of composite in magnet materials fabrication reduces the cost which means that it is more effective and efficient. However, composite bonded magnet does not have a very good stability due to its chemical content. This study tested the properties of NdFeB based composite bonded magnet that was fabricated from the mixture of NdFeB and BaFe$_{12}$O$_{19}$ powders also bakelite polymer in compression method. Physical properties of the samples were analysed through bulk density with dimensional method, Vickers Hardness (HV) test, microstructure using optical microscope (OM), magnetic properties using Gaussmeter and hysterical curve using Vibrating Sample Magnetometer (VSM). From the overall of the characteristic tests, the best composition obtained for NdFeB composite bonded magnet is 95%wt NdFeB, 5%wt BaFe$_{12}$O$_{19}$ with 2.5%wt of bakelite. This composition results in NdFeB composite bonded magnet with bulk density of 4.148 g/cm$^3$, 981.75 Gauss for magnetic flux. While remanence value (Br), coercivity (Hcj) and product energy (BH max) respectively are 4.410 kG, 9.334 kOe, and 3.679 MGOe. Meanwhile, the best hardness value, 163.58 HV, is given at BaFe$_{12}$O$_{19}$ with 2.5%wt of bakelite. Particle size observer using OM with Image-j gave the size of around 0.938-1.450 μm. Overall, the addition of BaFe$_{12}$O$_{19}$ and bakelite reduces the density and magnetic flux values whereas the hardness value increases with the addition of the two materials.

Keywords: Composite Bonded Magnet, BaFe$_{12}$O$_{19}$, Bakelite, Density, Magnetic Flux, Microstructure, NdFeB

1. Introduction
Magnet material has an important role in electrical field. In the application, this material is used for several electric appliances. One of the important uses in electric technology is in the making of permanent material magnet with the ability to store the energy of magnetic flux. To answer that challenge, the usage of composite bonded magnetic becomes a solution as it has high magnetic properties with good thermal properties [1] Meanwhile, composite bonded magnet is able to convert electrical energy to become mechanical and vice versa efficiently [2].
Composite bonded magnet is produced by mixing magnetic materials in the form of powder. There are several types of methods to make NdFeB composite bonded magnet. The first fabrication was by hydrogenation disproportionation and desorption recombination (HDDR) that effectively increase magnetic coercivity [3]. In order to get a smaller size magnetic material, an effective method of fabrication is needed, however. One of them is by sintering and compaction process [4] [5] [6]. Nevertheless, the fabrication of NdFeB depends of the supporting materials. Therefore it is necessary to know the optimum composition for the manufacturing process.

One of the main aspects for the properties of bonded magnet material is the level of material coercivity, this property is influenced by several factors and the main one is material composition. The addition of several types of metal can influence that, V-M metal is for instance [7], Ti and C [8] also Terbium [9]. The three studies reported that the coercivity value of composite bonded magnetic fabricated increases with the addition of metals.

Also sintering process mentioned above also has the influence to bonded magnet properties. Sintering mechanism needs a high temperature in order to increase energy [10]. A gradual decrease in temperature is for compaction and aging process, so the material used can merge together [11]. However, to increase the mechanic properties, a resin that works as binder need to be added, and so magnet permanent properties can be produced. Muljadi et al., [12] uses epoxy resin and able to work at a rotation of 400 rpm.

The mixture of 2 magnetic materials with different size is reported to increase magnetic properties to 10.68% [13]. Therefore this study uses NdFeB and BaFeO powder to get the magnetic properties. Besides that, physical parameter is tested through the microstructure and mechanical properties.

2. Experimental
2.1 Materials
This research used Neodymium powder from Iron Boron (NdFeB) MOP-B+ type with a density of 5.6 - 6.0 h/cm³. Barium Ferrite (BaFe₁₂O₁₉) and bakelite powders,

2.2 Methodology
2.2.1 Mixing of Raw Materials
The first step is done by mixing raw material such as NdFeB MOP-B+ type with BaFe₁₂O₁₉ powder. The mixture is then added with 2.5%wt of bakelite powder and 5%wt of various composition of NdFeB MOP-B+ type and BaFe₁₂O₁₉ with ratio mixture of 95,90,85,60%wt and 5,10,15,20%wt. 2.5%wt of bakelite powder is mixed into a total of 15.375 gram and 15.75 gram.

| NdFeB Powder (% wt) | BaFe₁₂O₁₉ Power (% wt) | Bakelite Powder (% wt) |
|---------------------|------------------------|------------------------|
| 100                 | 0                      | 2.5                    |
| 95                  | 5                      | 2.5                    |
| 90                  | 10                     | 2.5                    |
| 85                  | 15                     | 2.5                    |
| 80                  | 20                     | 2.5                    |
Table 2. The mixture of raw material with a mass of 15.75 gram

| NdFeB Powder (% wt) | BaFe₁₂O₁₉ Power (% wt) | Bakelite Powder (% wt) |
|---------------------|------------------------|------------------------|
| 100                 | 0                      | 5                      |
| 95                  | 5                      | 5                      |
| 90                  | 10                     | 5                      |
| 85                  | 15                     | 5                      |
| 80                  | 20                     | 5                      |

2.2.2 Composite Fabrication

Composite fabrication is conducted with compression molding method in temperature of 160°C and 8 ton force/cm² pressure for 15 minutes. The first step of fabrication was done by mixing NdFeb MOP-B+ type, BaFe₁₂O₁₉ mixture with bakelite. Each material mass was measured in %wt using digital analytics with the predefined composition (Table 1 and Table 2). After the mixture was homogenized, sample is put in a molding pellet with a diameter of 1.837 cm and height of 0.502 cm. Then the material was prepared to be analysed for bulk density, hardness Vickers, microstructure and magnetic properties.

2.3 Characterization

Composite bonded magnet characterization was done to know the mechanic and magnetic properties, as well as its morphology. Density and hardness tests were done for mechanic properties, whereas Optical Microscope instrument was used to know the morphology. Magnetic properties were tested using several instruments explained below.

Magnetisation test was done to NdFeB composite bonded magnet sample using physic DrSteingroever GmbH Impulse Magnetizer K-Series with resistance (V) = 1800 Volt and current (I) produced was around 5.90 - 5.98 kA. To know the magnetic properties of the sample, Gaussmeter was used while the magnetic field was analysed using Vibrating Sample Magnetometer (VSM). The result of the test is a hysterical curve with permanent induction (Br) and coercive force (Hc) values.

The observation for microstructure was done using Optical Microscope (OM) to get the image of the surface and the particle size. Composite bonded magnet sample was put on evaporating dish. Then, the magnification was set at 40x and the image was taken. The image of microstructure was observed.

Mechanical properties were tested with Vickers Micro-hardness Tester to know the hardness level. In this test, vickers method was conducted according to ASTM C 849-81 standard. NdFeB composite bonded magnet was mixed with resin until the material becomes hard. Next, material surface was smoothened until it shines, and it was being etched. The composite was formed into a diamond shape therefore the diagonal length can be measured. Density was calculated with the measurement of diameter and thickness of sample pellet by using caliper and digital weigher.

3. Results and Discussion

3.1 Density

The density of composite bonded magnet based on NdFeB composition (95,90,85,80 %wt) : BaFe₁₂O₁₉ (5,10,15,20 %wt) with the addition of 2.5 and 5 %wt of bakelite is shown in Table 3 and Table 4 respectively.
Table 3. The density of composite bonded magnet with various composition of NdFeB and BaFe$_{12}$O$_{19}$ and 2.5%wt of bakelite addition

| NdFeB Composition (%wt) | Barium Ferrite Composition (%wt) | Density (gr/cm$^3$) |
|------------------------|---------------------------------|---------------------|
| 95                     | 5                               | 4.148               |
| 90                     | 10                              | 4.039               |
| 85                     | 15                              | 4.018               |
| 80                     | 20                              | 3.869               |

Based on Table 3, the biggest density is 4.148 gr/cm$^3$ while the smallest value is 2.869 gr/cm$^3$. This result shows that the biggest Barium Ferrite composition is, the smaller the density, with a decrease in stability.

Table 4. The Bulk density of composite bonded magnet with various composition of NdFeB and BaFe$_{12}$O$_{19}$ and 5%wt of bakelite addition

| Komposisi NdFeB (%wt) | Komposisi Barium Ferrite (%wt) | Bulk Density (gr/cm$^3$) |
|-----------------------|---------------------------------|--------------------------|
| 95                    | 5                               | 4.040                    |
| 90                    | 10                              | 4.007                    |
| 85                    | 15                              | 3.979                    |
| 80                    | 20                              | 3.860                    |

Figure 1. Bulk density with BaFe$_{12}$O$_{19}$ composition with 2.5%wt and Bakelite 5%wt.

Figure 1 shows that the density decreases with the addition of BaFe$_{12}$O$_{19}$. At the addition of 2.5% bakelite powder, the density of composite bonded magnet is maximum, obtained at 5%wt BaFe$_{12}$O$_{19}$ with value 4.148 gr/cm$^3$. But the density tends to decrease, 4.039 gr/cm$^3$, with the addition of BaFe$_{12}$O$_{19}$ to 10% wt, which means there was a decrease by 0.109 %wt. While in 5%wt bakelite powder, the density still decreases by 0.033%wt from 5%wt to 10%wt. With the addition of BaFe$_{12}$O$_{19}$ leads to the fall of density value due to the density of barium ferrite (4.79) is smaller than NdFeB (7.64). The decrease of density shows the reduction of compactness in sample as a result of bakelite polymer mixture in composite bonded magnet NdFeB and the density of bakelite (1.36) is smaller than NdFeB.
3.2 Hardness

The hardness test for NdFeB composite bonded magnet with the addition of BaFe$_{12}$O$_{19}$ with composition of 0, 5, 10, 15, and 20 %wt and the addition of 2.5 and 5 %wt of bakelite was done using Vickers method by Vickers Hardness AMH43. The result is shown in Figure 2 and presented in Table 5 and Table 6 below.

| NdFeB composition (%wt) | Barium Ferrite composition (%wt) | Average hardness value (HV) |
|-------------------------|---------------------------------|-----------------------------|
| 95                      | 5                               | 106.65                      |
| 90                      | 10                              | 163.58                      |
| 85                      | 15                              | 75.76                       |
| 80                      | 20                              | 64.23                       |

Table 5 above shows the highest hardness value is 106.65 HV and the lowest id 64.23 HV. There is a downward trend to the addition of barium ferrite. The significant decrease is given at 15%wt of barium ferrite addition where the value decreases by more than 50%.

| NdFeB composition (%wt) | Barium Ferrite composition (%wt) | Average hardness value (HV) |
|-------------------------|---------------------------------|-----------------------------|
| 95                      | 5                               | 81.59                       |
| 90                      | 10                              | 132.196                     |
| 85                      | 15                              | 42.01                       |
| 80                      | 20                              | 50.03                       |

Table 6 above shows the highest hardness value is 132.196 HV and the lowest is 42.01 HV. From Table 5 and 6, a relationship graph between hardness value and barium ferrite addition is plotted as follow:

![Figure 2. Hardness value and BaFe$_{12}$O$_{19}$ composition in sample with the addition of 2.5 and 5 %wt of Bakelite](image-url)
Figure 2 shows that hardness value in composite bonded magnet with the addition of 10%wt BaFe$_{12}$O$_{19}$is the highest among them. The highest hardness value is obtained at 90% NdFeB : 10% BaFe$_{12}$O$_{19}$ : 2.5%wt bakelite and 90% NdFeB : 10% BaFe$_{12}$O$_{19}$ : 5%wt bakelite compositions, with value of 163.58 HV and 132.20 HV respectively. It also shows that bakelite composition affected in hardness test. There is a decrease of hardness value along with the addition of bakelite to the sample. The decrease occurs due to the existence of polymer material with low hardness characteristic, 14 HV. Therefore, hardness value decreases with the increase of polymer composition in composite bonded magnet.

3.3. Microstructure Observation

Microstructure observation was done by studying morphology image of magnet service and determining particle size with Optical Microscope (OM). The observation with OM was done to 4 samples with various compositions of 5%wt and 10%wt Barium Ferrite with the addition of 2.5%wt and 5%wt bakelite. They are shown in Figure 3. Based on the result from OM, the average of particle size can be determined from the source material that can be seen in Figure 3 below.

![Figure 3](image1)

**Figure 3.** The morphology on the surface of NdFeB - BaFe$_{12}$O$_{19}$ composite bonded magnet with optical microscope using 40x magnification. (a) The addition of 5%wt barium ferrite and 2.5%wt bakelite (b) The addition of 5%wt barium ferrite and 5%wt bakelite (c) The addition of 10%wt barium ferrite and 5%wt bakelite (d) The addition of 10%wt barium ferrite and 5%wt bakelite
Based on the morphology of surface produced from OM, bakelite magnet can spread partly or equally in sample surfaces. As can be seen in the images, black colour is identified as barium ferrite, grey as NdFeB, white as Nd and the blurred one is adhesive to bakelite. There are differences in image (a) and (c) where they are darker with the addition of 2.5%wt bakelite when picture 3(b) and (d) is a little bit brighter with the addition of 5%wt bakelite. Particle size is determined with image-j shown in the table below. This microstructure observation shows that the distribution of barium ferrite is not well distributed which was caused by the agglomeration due to the compaction process.

Table 7. Particle size of NdFeB based composite bonded magnet with an addition of 2.5%wt bakelite.

| NdFeB Composition (%wt) | Barium Ferrite Composition (%wt) | Average particle size (µm) |
|-------------------------|----------------------------------|---------------------------|
| 95                      | 5                                | 1.109                     |
| 90                      | 10                               | 1.033                     |
| 85                      | 15                               | 1.376                     |
| 80                      | 20                               | 1.204                     |

Based on particle size table, the overall value shows an average of particle size at the range of 1 - 1.4 micrometer. The biggest size is obtained at NdFeB and barium ferrite with a composition of 85%wt and 15%wt respectively.

Table 8. Analysis with OM for average particle size of NdFeB composite bonded magnet at the addition of 5%wt NdFeB

| NdFeB Composition (%wt) | Barium Ferrite Composition (%wt) | Average particle size (µm) |
|-------------------------|----------------------------------|---------------------------|
| 95                      | 5                                | 1.137                     |
| 90                      | 10                               | 1.213                     |
| 85                      | 15                               | 0.938                     |
| 80                      | 20                               | 1.450                     |

Average particle size with an addition 5%wt bakelite is at 0.938 - 1.450 µm. Therefore from Table 7 and 8, average particle size can be known. The size of the particle is similar to the material with an addition of 2.5%wt bakelite, which shows that there is no increase in the size with the determined composition.

3.4 Magnetic Field Strength

Measurement with Gaussmeter was done to know the strength of magnetic field from sample of NdFeB magnetic hybrid. The results of the measurement with Gaussmeter (magnetic field strength) are shown in Table 9, Table 10 and Figure 4.

Table 9. The average of magnetic flux for NdFeB composite bonded magnet with composition variation of BaFe$_{12}$O$_{19}$ with an addition of 2.5%wt bakelite

| NdFeB Composition (%wt) | Barium Ferrite Composition (%wt) | Average Magnetic flux (Gauss) |
|-------------------------|----------------------------------|------------------------------|
| 5                       | 2.5                              | 981.75                       |
| 10                      | 2.5                              | 892.25                       |
| 15                      | 2.5                              | 843.70                       |
| 20                      | 2.5                              | 830.25                       |
Table 10. The average of magnetic flux for NdFeB composite bonded magnet with composition variation of BaFe$_{12}$O$_{19}$ with an addition of 5%wt bakelite

| NdFeB Composition (% wt) | Barium Ferrite Composition (% wt) | Average Magnetic flux (Gauss) |
|--------------------------|----------------------------------|-----------------------------|
| 5                        | 2.5                              | 845.85                      |
| 10                       | 2.5                              | 822.25                      |
| 15                       | 2.5                              | 815.30                      |
| 20                       | 2.5                              | 730.90                      |

Table 9 above shows the highest magnetic flux value of 981.75 Gauss and the lowest is 830.25 Gauss. Generally, the increase of NdFeB composition causes the gradual decrease of magnetic flux. The biggest decrease is found at 10%wt NdFeB addition which causes magnetic flux fell from 981.75 Gauss to 892.25 Gauss. While in Table 10 shows the highest magnetic flux at 845.85 Gauss and the lowest is at 730.90 Gauss. Based on the results shown in Table 9 and 10, sample with an addition of 5%wt barium ferrite composition and mixed with 2.5%wt bakelite has the highest magnetic flux at 981.75 Gauss. On the other hand, the lowest magnetic flux is found in sample with an addition of 20%wt barium ferrite and mixed with 5%wt bakelite. From Table 9 and Table 10, a graph to show the relationship between magnetic flux and barium ferrite addition was plotted as follow:

![Graph showing the relationship between magnetic flux and barium ferrite addition](image)

**Figure 4.** Magnetic field strength and the composition of barium ferrite with addition of 2.5%wt and 5%wt bakelite.

From figure 4 we can see that the increase of barium ferrite composition reduces magnetic flux in sample. This is because magnetic property in barium ferrite is lower than bonded NdFeB. Therefore when they are mixed together, there will be a reduction in its magnetic properties. There is a gradual increase of flux magnetic either in 2.5%wt or 5%wt of bakelite usage.

3.5 Vibrating Sample Magnetometer (VSM)

Vibrating sample magnetometer (VSM) is a type of equipment used to study magnetic properties of a material. VSM testing is done to obtain information about the scale of magnetic properties as a result of the changes in the outermost magnetic field. The scale of magnetic properties in a material can be known through the hysterical curve shown above. The curve shows the scale of remanence magnetization (M_r), saturated magnetization (M_s), remanence induction (B_r), coercivity (H_c) and product energy (BH$_{max}$). The result of VSM test is shown in Figure 5 below.
Figure 5. Hysteresis Curve of NdFeB Composite Bonded Magnet and Bakelite (a) hysteresis curve of 100% wt NdFeB with the addition of 2.5% wt of bakelite; (b) hysteresis curve of 95% wt of NdFeB with the addition of 5% wt of BaFe$_{12}$O$_{19}$ and 2.5% wt of Bakelite; (c) hysteresis curve of 95% wt of NdFeB with the addition of 5% wt of BaFe$_{12}$O$_{19}$ and 5% wt of Bakelite

At hysteresis curve above, NdFeB composite bonded magnet and bakelite adhesive are shown as hard magnet. Hard magnetic material (permanent magnet) is marked with coercivity value $H_c$ above 200 Oe. $H_c$ represents the scale of reverse magnetic field needed to kill the magnet in one material. While magnetic field is measured by remanence ($B_r$) scale from a material which is the remaining of outermost magnetic induction in a material after the magnetic field is killed. The two parameters can be observed directly from the hysteresis curve. The maximum product energy ($BH_{max}$) from the magnet is produced from the maximum value of the multiplication between $B$ and $H$ in second quadrant in the hysteresis curve. The higher the remanence value, the more developed its coercive force and hysteresis loop and therefore product energy is getting higher too.

Table 11. Vibrating Sample Magnetometer (VSM) test result

| Composition                     | $M_r$ (emu/g) | $M_s$ (emu/g) | $B_r$ (kG) | $H_{cj}$ (kOe) | $BH_{max}$ (MGOe) |
|---------------------------------|---------------|---------------|------------|----------------|-------------------|
| 100 % wt NdFeB + 2.5% Bakelite | 76.91         | 122           | 4.059      | 8.676          | 3.382             |
| (95 % NdFeB + 5% BaFe$_{12}$O$_{19}$ + 2.5% Bakelite) | 79.47         | 123           | 4.140      | 9.334          | 3.679             |
| (95 % NdFeB + 5% BaFe$_{12}$O$_{19}$ + 5% Bakelite) | 73.93         | 119           | 3.751      | 8.560          | 2.991             |

Table 11 shows the decrease in $B_r$ value in sample with addition of 5% wt BaFe$_{12}$O$_{19}$ with 2.5% wt bakelite against sample with addition of 5% wt BaFe$_{12}$O$_{19}$ and 5% wt bakelite in which the value dropped from 4.140 to 3.751 kG. Whereas $H_{cj}$ value fell from 9.334 kOe to 8.560 kOe and product energy decreased from 3.678 MGOe to 2.991 MGOe. The best $BH_{max}$ value is obtained from sample with an addition of 5 % BaFe$_{12}$O$_{19}$ and 2.5% bakelite.

4 Conclusions

In this study, composite bonded magnet is made using hydraulic press with an addition of bakelite in NdFeB and BaFe$_{12}$O$_{8}$ mixture. The average particle size obtained is below 1.5 micrometer with an uneven distribution in barium ferrite and bakelite. From the test result, it is known that the addition of NdFeB magnet powder causes the decrease in density and also with the increase of bakelite. However, hardness from bonded magnet material is increased with the percentage of bakelite added whereas in magnetic flux, the decrease goes along with the addition of barium ferrite and bakelite composition.
References

[1] M. T. Clavaguera and M. N. Clavaguera, "Nucleation and Growth of Nd2Fe14B crystals from rapidly quenched amorphous alloys: a kinetic study," Journal of Magnetic Material, pp. 66-68, 1999.

[2] L. Ferraris, P. Ferraris and E. Poskovic, "Theoretical and experimental approach to the adoption of bonded magnets in fractional machines for automotive applications," IEE. Trans. Ind. Electron, pp. 2309-2318, 2012.

[3] T. Takeshita and R. Nakayama, "11th Workshop on rare earth magnets and their applications," Pittsburgh, 1990.

[4] J. D. Livingston, "Magnetic domains in sintered Fe-Nd-B magnets," Journal Applied Physics, pp. 4137-4139, 1985.

[5] S. W. Tao, J. J. Tian, X. Lu, X. H. Qu, Y. Honkura and H. Mitarai, "Anisotropic bonded NdFeB magnets with radial oriented magnetization by 2-step warm compaction process," Journal Alloy. Compd., pp. 510-514, 2009.

[6] B. Ma, A. Z. Sun, Z. W. Lu, C. Cheng and C. Xu, "Research on anisotropic bonded Nd-Fe-B magnets by 2-step compaction process," J. Magn. Magn. Mater, pp. 802-805, 2016.

[7] H. Kanekiyo, M. Uehara and S. Hirosawa, "Magnetic properties and microstructure V-and-M-added, Fe3B-based, Nd-Fe-B nanocrystalline permanent magnets (M=Al, Si)," Mater. Sci. Eng. A., pp. 181-182, 1994.

[8] N. J. Harrison, H. A. Davies and I. Todd, "Nd2Fe14B-based nanocomposite magnets with transition metal and carbon additions," J. Appl. Phys., p. 08B504, 2006.

[9] P. Minxiang, Z. Pengyue, L. Xianjun, G. Honglian, W. Qiong, J. Zhiwei and L. Tingting, "Effect of Terbium addition on the coercivity of the sintered NdFeB magnets," Journal of Rare Earths, pp. 399-402, 2010.

[10] Q. Yu, R. S. Liu, K. T. Dong and Y. P. Zhang, "Key techniques for ultrahigh performance sintered Nd-Fe-B magnets preparation," Transworld Research Network, pp. 1-36, 2012.

[11] M. Kokab, F. Arabgol and M. Manteghian, "Permanent polymeric composite magnets," Iranian Polymer Journal , pp. 71-79, 2005.

[12] M. Muljadi, P. Sardjono and S. Suprapedi, "Preparation and characterization of 5 wt.% epoxy resin bonded magnet for micro generator application," in 2nd International Conference on Sustainable Energy Engineering and Application, ICSSA, 2014.

[13] B. Ma, A. Sun, X. Gao, J. Li and H. Lang, "Preparation of anisotropic bonded NdFeB/SmFeN hybrid magnets by mixing two different size powders," Journal of Magnetism and Magnetic Materials, 2017.