Physical and magnetic properties, microstructure of bonded magnet NdFeB prepared by using synthesis rubber

Suprapedi, P. Sardjono, Muljadi

Research Center for Physics, Indonesian Institute of Sciences

E-mail: suprapu@gmail.com

Abstract. The magnet permanent has been made by using NdFeB (NQP-B) powder with purity 99.90% and polymer rubber with purity 90%. This research was done to determine the effect of the polymer (rubber) composition as binder in the manufacture of bonded magnet NdFeB on physical properties, microstructure and magnetic properties. Bonded magnets are magnet material made from a mixture of magnetic powder as a filler and the polymer as a matrix material or as binder to bind the magnetic particles. The NdFeB (NQP-B) powder fractions of bonded magnets NdFeB investigated were as follows: a) 97 wt.% of NdFeB (NQP-B) and 3 wt.% of rubber, b) 95 wt.% of NdFeB (NQP-B) and 5 wt.% of rubber, c) 93 wt.% of NdFeB (NQP-B) and 7 wt.% of rubber, d) 91 wt.% of NdFeB (NQP-B) and 9 wt.% of rubber. Both raw materials were mixed by using mixer until homogen with total weight about 16 g for each sample, then added 0.3 ml of catalyst and mixed again and put in dies mould and compacted at pressure 30 MPa, then dried for 2 hours at room temperature. The dried samples was characterized such as: bulk density measurement and magnetic properties by using BH-curve permeagraph. The bulk density values of the sample bonded NdFeB magnets using the binder 3% wt. and 5% wt. rubber are respectively 4.70 g/cm$^3$ and 4.88 g/cm$^3$. The result from BH-curve shows that the highest value of remanence ($\text{Br} = 5.12$ kGauss) is at sample with 3 % wt. of rubber, but sample with 5 % wt. of rubber has lowest value of remanance ($\text{Br} = 4.40$ kGauss). Based on the observation of the SEM photograph shown that the rubber material has been successfully covered the whole surface of the grain and fill some of the voids that are in the grain boundary.

1. Introduction

Permanent magnet has been developed ; the first of magnetic iron was used then in developing that magnetic iron magnet has been made from some metallic alloy such as : SmCo, AlNiCo and rare earth base magnet (Nd$_2$Fe$_{14}$B) [1]. Also, some of magnetic materials are made from oxide base compounds as like Barium hexa ferrite BaO$_6$Fe$_2$O$_3$ and it is called hard ferrite [1]. It has limitations of magnetic properties, for example, Ba-hexaferrite has remanence (Br) about 3 – 4 kGauss and energy product [BH]$_{\text{max}}$ about 5 MGOe [2]. Compare of Ba-hexa ferrite to rare earth magnet (Nd$_2$Fe$_{14}$B ) that Nd$_2$Fe$_{14}$B magnet has higher magnetic performance, it has remanence (Br) about 5 – 13 kGauss and energy product [BH]$_{\text{max}}$ 5 - 30 MGOe [2]. Rare earth permanent magnet NdFeB is a new kind of magnetic material developed in the 1980's with excellent magnetic characteristics (high energy product and high coercive force etc.) and relatively low cost. It is getting to replace the traditional magnets of hard ferrite, AlNiCo and SmCo. There are two types of NdFeB magnet namely : (a) sintered magnet and (b) bonded magnet. Sintered NdFeB magnet is made by using sintering process at high
temperature in absence of oxygen [2,3]. Bonded NdFeB magnets are generally prepared by blending magnetic powder with binder followed by compacting or molding the material to final shape. The sintered Nd$_2$Fe$_{14}$B magnet has higher magnetic properties than bonded Nd$_2$Fe$_{14}$B magnet, and both have different application [4]. The magnetic properties of Nd$_2$Fe$_{14}$B are shown in Table 1.

| Properties               | Bonded NdFeB | Sintered NdFeB |
|--------------------------|--------------|---------------|
| Remanence Br (kG)        | 6.9          | 13.1          |
| Energy Product BHmax (MGOe) | 10          | 42            |
| Coercivity Hci (kOe)     | 9            | 14            |
| Curie Temperature Tc (°C) | 200         | 310           |

NdFeB magnets have disadvantages such as easy to oxidation or lower corrosion resistance at room temperature. To protection the corrosion or oxidation, the surface of NdFeB magnet should be coated. There are many kinds of coating materials such as: Ni-plated, chrome plated and epoxy or other polymer plated [5,6]. There are some of polymer materials for coating such as: thermostetting polymer epoxy. Bonded NdFeB permanent magnets are mainly composed of NdFeB magnetic powder and polymeric binders such as thermoplastic polyolefin; polyphenylene sulphide (PPS), polyvinyl chloride (PVC), latex or rubber, polypropylene (PP), polyethylene (PE), high density polyethylene (HDPE), polyamide or thermostetting epoxy resin, nitrile rubber or other elastomers [7]. The most important advantages of polymer bonded magnets are low weight, corrosion resistance, easy machining ability, forming and handling, high production rate. Bonded magnets provide an almost infinite variety of combinations of mechanical, physical, chemical, thermal and magnetic properties due to various kinds of polymeric matrices. Several authors have been conducting research manufacture bonded NdFeB magnets using various polymer materials bleak nylon powder, epoxy resin, PEG and so on [8,9,10]. While research manufacture bonded NdFeB magnets made using different binder material which is using polymer rubber. There has been no research manufacture of bonded magnets using rubber polymer, in addition to the price of rubber pasta is much cheaper compared to other polymers and require a low aging temperature is room temperature. The goal of this experiments are to know the influence of rubber composition to density and magnetic properties on preparation of bonded NdFeB magnet.

2. Experimental
Bonded magnets are magnet material made from a mixture of magnetic powder as a filler and the polymer as a matrix material or as binder to bind the magnetic particles. There are four kinds of samples bonded NdFeB magnets that have been made with a variety of polymer adhesive composition (rubber pasta). The raw material used to manufacture bonded NdFeB magnets are NdFeB powder (NQP-B) with a purity of 99.90% and gum paste with a purity of 90%. The NdFeB (NQP-B) powder fractions of bonded magnets NdFeB investigated were as follows: a) 97 wt.% of NdFeB (NQP-B) and 3 wt.% of rubber, b) 95 wt.% of NdFeB (NQP-B) and 5 wt.% of rubber, c) 93 wt.% of NdFeB (NQP-B) and 7 wt.% of rubber, d) 91 wt.% of NdFeB (NQP-B) and 9 wt.% of rubber. Both raw materials were mixed by using mixer until homogen with total weight about 16 g for each sample, then added 0.3 ml of catalyst and mixed again and put in dies mould and compacted at pressure 30 MPa, then dried for 2 hours in open room at 30°C. The characterizations were done such as: measurement of bulk density by using Archimedes method. The bulk density value of solid materials (in fragment form) can be calculated by using Archimedes method, mathematically, density is defined as mass divided by volume. A basic formula to calculate the density of materials can be used the following equation [11]:

\[
\rho_o = \frac{m_p f}{\Delta m}.
\]
Δm is the mass difference value objects hung in the air with the mass of the object in suspension in the liquid, ρf is density of liquid, and m is mass of objects. The observation micro structure was done by using Scanning Electron Microscope (SEM) and magnetic properties measurement by using Gaussmeter and permeagraph (BH-curve analyzer) to obtain value of magnetic flux density, remanence Br, coercivity Hic and energy product [BH]max.

3. Results and Discussion
The XRD profile of NdFeB (NQP-B) powder is shown at Figure 1. According on analyzing with GSASS is found that major phase is Nd₂Fe₁₄B [ICDD-96-100-8719] about 78.14 % and minor phase is Fe [ICDD-96-901-3473] about 21.86 %. XRD analysis of the raw magnetic powder is important to determine the content of the phase of the material (NQP-B powder), because there are many types of NQP-B powder were sold. With different content of the phases present in the raw magnetic powder will gave different magnetic properties, so the results of this experiment refers to a type of magnetic powder NQP-B.

![Figure 1. Diffraction patterns of NdFeB powders [NQP-B]](image)

The curve of bulk density versus composition of rubber is shown at Figure 2. The bulk density value has tendency decreasing when rubber composition is higher, because it follows a composite rule that density of rubber is lower then density of NdFeB powder. The highest density value is achieved about 4.88 g/cm³ at sample with 3 % wt. rubber and lowest density value is 4.20 g/cm³ at sample with 9 % wt. rubber. Theoretical density of bonded magnet NdFeB is about 5 – 6 g/cm³ [2]. The densification of the sample bonded NdFeB magnets from this experiment is still low at around 70-80%. It is caused by the pressure on forming process at 30 MPa is still relatively low.

![Figure 2. The curve of bulk density Nd₂Fe₁₄B for different rubber composition](image)

Before magnetic properties measured, all samples are magnetized with field strength about 8 kGauss by using impulse magnetizer. Then the magnetic flux density was measured by using Gaussmeter and the result is shown at Figure 3.
According to Figure 3, the highest value of magnetic flux density is achieved about 1805-1820 Gauss at samples with 3% and 5% wt. of rubber, but the magnetic flux density tends to decrease again until 1640 Gauss with increasing of rubber composition. Rubber polymer is classified as non-magnetic material, when the rubber composition of the sample bonded NdFeB magnets increases, the magnetic properties of bonded magnets will tend to decline. The BH – curve results for different of rubber composition is shown at Figure 4. The BH-curve shows that when rubber composition increase, the curve hysteresis will be smaller. Rubber polymer is non-magnetic material, when the rubber composition of the sample bonded NdFeB magnets increases, of course, the composition of the magnetic powder (NdFeB) will shrink. This implies that the magnetic properties of bonded magnets will tend to decline. The biggest of BH-curve is at sample with 3 and 5% wt. of rubber. According to BH-curve result, the magnetic properties such as remanence Br, coercivity Hc, and energy product [BH]max of each sample are presented at Table 2. The rubber composition influenced significantly value of Br, Hc and [BH]max.

The biggest of BH-curve is at sample bonded Nd$_2$Fe$_{14}$B magnet with 3 and 5% wt. of rubber. According to BH-curve result, the magnetic properties such as remanence Br, coercivity Hc, and energy product [BH]max of each sample are presented at Table 2. The rubber composition influenced significantly value of Br, Hc and [BH]max. Because rubber is non magnetic materials and Polymer that serves as a matrix wrap any Nd$_2$Fe$_{14}$B particle that magnetic properties of the particle will be isolated by polymer. Bonded Nd$_2$Fe$_{14}$B with composition of 3 and 5% wt. rubber have the best magnetic properties. They have Br value = 4.40-5.12kGauss, Hc value = 6.02-5.98 kOe and [BH]max = 10.12-8.77 MGOe.
Table 2. The value of Remanence (Br), Coercivity (Hc) and Energy Product (BHmax) of bonded magnet with variation of % of rubber.

| % wt.of Rubber | Br, Kgauss | Hc, kOe | BHmax, MGOe |
|----------------|------------|---------|-------------|
| 3              | 5.12       | 6.02    | 10.12       |
| 5              | 4.4        | 5.98    | 8.77        |
| 7              | 3.85       | 3.83    | 5.25        |
| 9              | 2.92       | 2.94    | 3.5         |

Based on the results of measurement of magnetic properties showed that the sample bonded NdFeB magnet with 3% rubber composition has most high value of Br, Hc and BH max respectively, are 5.12 kG, 6.02 kOe and 10.12 MGOe. Magnetic properties of bonded NdFeB magnet from literature [4] and experiments [2,8,9 and 10] that have been done several authors with other polymer binder resulting in a value range of Br = 5-6 KG, Hc = 5-9 Koe and BH Max about 10 MGOe. the results obtained from this study are not much different from the references or the results of other authors.

Figure 5 shows the SEM micro graph of the samples bonded magnet made by using 3 to 9 % wt. of rubber. From SEM images, it can be seen that the rubber binds all surface of grains. The image with grey colour is a grain of magnetic Nd$_2$Fe$_{14}$B and the images with white and black colour indicate as rubber polymer. The SEM images for sample with 7% and 9% of rubber show that the polymer rubber is seen not only bind the grains at the grain boundaries but also cover portion of a grains surface. The polymer rubber on sample with 5% and 3% of rubber bind the grain only on grain boundary that are responsible by decreasing density value and magnetic properties for sample with 7% and 9% wt. of rubber.

Figure 5. SEM images of samples bonded magnet NdFeB with variation of rubber binder
4. Conclusion

According to XRD result shows that NdFeB powder which used to make bonded magnet has \( \text{Nd}_2\text{Fe}_{14}\text{B} \) phase about 78.14 % and Fe phase about 21.86 %. The composition of rubber (3 to 9 % wt.) gives significant effect to bulk density and magnetic properties such as magnetic flux density, remanence \( B_r \), coercivity \( H_c \) and energy product \( [BH]_{\text{max}} \). These conditions can be used reference for making of bonded permanent magnet with variation of properties or customer made. Sample bonded NdFeB magnet with 3 % wt. rubber having the best magnetic properties. It has bulk density = 4.88 g/cm\(^3\), \( B_r \) value = 5.12 kGauss, \( H_c \) value = 6.02 kOe, \( [BH]_{\text{max}} \) = 10.12 MGOe and magnetic flux density = 1820 Gauss. The result of observation using the SEM of sample with 3% wt. rubber showed that almost all of the polymer rubber has been blanketing the surface of the particles and bind the grains with each other.

Acknowledgments

The author would like to thank Program Riset Mandiri LIPI as financial support, and thank you to Mr. Nanang Sudrajat ST PPET-LIPI Bandung for helping in Magnetizing characterization, and thank you for operators XRD, SEM at Research Center for Physics-LIPI.

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