The effect of dry and wet milling on microstructure and properties of bonded NdFeB magnets

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Abstract. The preparation of NdFeB has been conducted in a dry and wet milling condition using a planetary ball mill (PBM) for ½, 1, 2, 3, 4, and 5 hours. The dry and wet milling was performed in an argon gas and toluent, respectively with ball and powder ratio of 10:1. The milled powder was dried in the vacuum condition of 11 mBar at the temperature of 60°C for 10 hours. 5wt.%Celuna (WE-518) was used as binder, mixed with magnetic powder and anisotropically pressed at 25 kgf/cm². Curing process was conducted in a vacuum chamber of 11 mBar at temperature of 180°C for 1 hour. The particle diameter and the true density of NdFeB powder were measured by using PSA and Pycnometer, respectively. The morphology and phase composition were characterized using SEM and XRD, respectively. The obtained results show that the optimum milling time of NdFeB powder is 3 hours using wet milling method. The results also concluded that the powder true density of 7.79 g/cm³, the bulk density of 5.98 g/cm³ and the mean particle diameter of 9.59 µm can be achieved. The SEM image confirmed that the milling process has an effect in reducing the particles size. But the milling process lead to a damage in the crystal structure of the material, resulting in decreasing the magnetic flux density. The half hour dry milling is capable to produce a maximum magnetic flux density of 1816 Gauss.

1. Introduction

The demand for magnetic materials continues to rise in line with advances in the electronics technology, computerization and transportation [1]. It was begun after the discovery of the rare earth-based permanent magnets, such as NdFeB, RECo, and REFeB [2,3]. Currently, permanent magnet materials in a generator/electric motor have also been used to convert the electrical energy into the mechanical energy or vice versa [2,4]. The advantageous of a permanent magnet is the fact that the energy produced is quite high and environmentally friendly [5,6]. In general, the main qualities of permanent magnet are characterized by several parameters, such as magnetic remanence (Br), magnetic coercivity (Hc), the maximum energy product (BH₀), magnetic flux density, Curie temperature, etc [7,8].

The NdFeB permanent magnets have been widely used, especially in the electronic equipment, electrical generators, sensors transducer, automotive, petrochemical industries, medical equipment products and other applications. Currently, permanent magnet is also used in the automotive field as...
electric power steering system, sensor rpm/velocity and electric motor. The numbers continue to increase rapidly every year [6,7].

Currently, permanent magnet based on Nd$_2$Fe$_{14}$B phase is the best type of permanent magnet which has a maximum energy product (BH$_{\text{max}}$) that reaches up to 27 MGOe[9]. Therefore, small size Nd$_2$Fe$_{14}$B permanent magnet has high capability [10]. NdFeB magnets have also been used to replace samarium cobalt magnets in some applications, in particular for the application at the temperatures less than 80°C [5]. The disadvantage of this type of magnet is its low curie temperature, easily oxidized and difficult to be applied at high temperatures condition [11,12].

In order to support the development of NdFeB magnets, the research is focusing on the effect of dry and wet milling process on fabrication, microstructure and properties of NdFeB magnets. In this study, NdFeB powder (type MQP-B+ 10118-70) is selected as the main raw material, Planetary Ball Mill (PBM) is used for milling process, argon and toluent as milling media, and celuna (WE-518) as an adhesive medium. The compaction and curing process was conducted at 25 kgf/cm$^2$ in the magnetic field press and at 180°C in vacuum (11 mBar) for 1 hour. The powder mean diameter and true density, bulk density, morphology, microstructure, and magnetic flux density was characterized, measured and discussed.

2. Experimental Method

The raw material powders used in this study is NdFeB powder type MQP-B+10118-70 (technical raw materials). The specification of NdFeB powder MQP-B+ are specified in the following: residual induction (Br) = 8.95-9.15 kG, intrinsic coercivity (HcI) = 9.0-10.5 kOe, coercive force (Hc) = 6.8 kOe, energy product (BH$_{\text{max}}$) =15.8-16.8 MGOe, Curie temperature (T$_c$) =360°C, maximum process temperature = 200°C and maximum operating temperature =120-160°C. The wet and dry milling was done by using a Planetary Ball Mill (PBM) that has powder and ball ratio of 1:10 for variation of time: ½, 1, 2, 3, 4 and 5 hours. In the wet milling process, toluene was used as milling medium and in the dry milling process; argon gas was injected in the crucible PBM. The ball milled powder was dried in a vacuum oven of 11 mBar at 60°C for 10 hours.

![Flow diagram of preparation and characterization of bonded NdFeB permanent magnets.](image)

**Figure 1.** Flow diagram of preparation and characterization of bonded NdFeB permanent magnets.
For compaction, 3 grams of NdFeB powder was mixed with 5wt.% Celuna (WE-518) and pressed in a magnetic field press (anisotropy) with a pressure of 25 kgf/cm². The compact sample was then cured in a vacuum chamber of 11 mBar at temperature of 180°C for 1 hour. The specimen magnetization was conducted by using Impulse Magnetizer (voltage of 1300 volts). The specimen characterization tested were the mean particle diameter, true density, bulk density, morphology, microstructure, and magnetic flux density by using PSA, pycnometer, Archimedes principle, SEM, XRD and Gaussmeter, respectively. The flowchart of the fabrication process and the characterization of bonded NdFeB permanent magnets are shown in figure 1.

3. Results and Discussion
The cumulative diameter of NdFeB powders after dry and wet milling for 0.5; 3 and 5 hours were measured by using a Particle Size Analyzer (PSA) as shown in figure 2.

![Figure 2. Cumulative value versus diameter of NdFeB powders after dry milling for 0.5h, 3.0h, and 5.0h and wet milling for 0.5h, 3.0h and 5.0h.](image)

The diameter of NdFeB powder as a function of milling time prepared by dry and wet milling are shown in figure 3. It can be seen that the mean particle diameter decreased at longer milling time until it reaches the minimum conditions. However, when the milling time was extended, the grain size was not decreased due to the powder agglomeration. The wet milling is a relatively better technique compared with the dry milling. This is probably due to that the collisions between the ball and grains are more appropriately milled. The optimum milling time is achieved at 3 hours with a mean diameter of 13.12 µm for dry milling and 9.59 µm for wet milling.

![Figure 3. The diameter of NdFeB powders after dry and wet milling as function of milling time.](image)
Figure 4 shows the true density of NdFeB powder at various milling time. The highest true density was obtained after 3 hours milling time. This means that as the grain size decreased, the true density increased. The true density of wet and dry milled powders after 3 hours milling time is 7.79 g/cm$^3$ and 7.75 g/cm$^3$, respectively.

![Figure 4. True density of NdFeB powders as a function of milling time after dry and wet milling process.](image)

The SEM images of NdFeB powder in different condition of milling and time are shown in figure 5. All SEM photographs of NdFeB powder was taken with a magnification of 500X. Based on the results, it can be seen that the NdFeB powder before milling is relatively has larger diameter and randomly distributed. The SEM image also clearly shows the effect of milling in reducing the diameter of the particle. The smaller particles result in higher level of density and lower level of porosity. But, there are no significant changes in SEM images.

Figure 6 shows that the bulk density of bonded NdFeB magnets at various milling time. The results show that the bulk density of bonded NdFeB magnet increased with the increase in milling time for up to 3 hours and then decreased at extended milling time. The optimum bulk density of the specimen is 5.98 g/cm$^3$ for wet milling and 5.90 g/cm$^3$ for dry milling condition. The bulk density of NdFeB samples prepared by wet milling is relatively higher than the dry milling samples. At longer milling time (>3h), the bulk density of the sample is likely to decrease. This is due to the occurrence of grain or agglomeration which results in the presence of pores or voids in the sample.

![Figure 5. SEM images of bonded NdFeB magnets (a). without milling ; dry milling at : (b). ½ h, and (c). at 5 h ; wet milling at (d). ½ h and (e). 5 h.](image)

The relationship between magnetic flux density of bonded NdFeB magnets and the milling time is shown in figure 7. The optimum magnetic flux density of bonded NdFeB magnets is obtained at half-
The bulk density of dry milling process is relatively better than that of wet milling process. At half-hour milling time, the bulk density of the specimen is 1816 Gauss for dry milling and 1434 Gauss for wet milling. The magnetic flux density of bonded NdFeB magnet decreased with the increase in milling time. This suggests that the milling process can lower the magnetic properties of bonded NdFeB samples. This is probably due to the oxidation of the raw material during the milling or handling process.

Figure 6. Bulk density of bonded NdFeB magnets at varying milling time.

Figure 7. Magnetic flux density of bonded NdFeB magnet at varying milling time.

Figure 8 shows the XRD patterns of original powder, dry and wet milled powder (half and five hours). Apparently, Nd$_2$Fe$_{14}$B phase was detected in the original powder, but it disappeared in the dry milled and wet milled powder. The ball milled powder was turned into an amorphous phase. This means that the milling process has a detrimental effect on the structure of the material. The powder is likely oxidized to form the oxide layer during the milling process.

Figure 8. XRD pattern of (a) original powder, dry milled powder at : (b). ½ h and (c). 5 h; wet milled powder at : (d). ½ h and (e). 5 h.
4. Conclusion
Based on the above results, it can be concluded that the NdFeB powder that have been prepared using wet milling is relatively better compared to dry milling condition. The optimum milling time was achieved for 3 hours. These results show that a mean particle diameter of 9.59 µm and the true density of 7.79 g/cm$^3$ can be achieved. The optimum bulk density of bonded NdFeB magnets is 5.98 g/cm$^3$ that have been obtained using wet milling for three hours. The obtained results show that the milling process can reduce the size of the particles, but the milling process can also lead to damages in the crystal structure of Nd$_2$Fe$_{14}$B. Consequently, the magnetic flux density of bonded NdFeB magnet was decreased. The half-hour dry milling process was only able to reach a maximum magnetic flux density of 1816 Gauss.

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