Effect of Particle Size Distribution on the Preparation of Bonded NdFeB Permanent Magnet

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Abstract. In this paper, the effect of particle size distribution on the preparation of bonded NdFeB isotropic permanent magnets was investigated. Different particle size distribution of NdFeB powders was prepared by sieving method using standard test sieves of #100, #200, and #325 meshes. Varied distribution of particle size was mixed with 3 wt\% of epoxy binder and compacted by uniaxial hydraulic pressing machine to produce bonded isotropic green body. Bonded NdFeB green body was cured by vacuum drying machine under temperature 100 °C for 4 hours with vacuum pressure of 10 mbar. The characterization performed were bulk density measurement, particle size analysis, microhardness Vickers, SEM image and hysteresis curve. We find that the hardness of bonded NdFeB magnets decreased as the particle size decreases. Contrary to common belief that the finer powder with sharply particle size distribution not always give the highest product characteristics. The optimum characteristics of bonded NdFeB was obtained by the mixture of #100+#200 meshes powders with bulk density, hardness, magnetic remanence, coercivity, and energy product of 5.87 g/cm\textsuperscript{3}, 88 HV, 6.5 kG, 11.15 kOe, and 9.4 MGOe, respectively. Therefore, the middle range of particle size distribution is effective to obtain high quality bonded NdFeB permanent magnets.

Keywords: Particle size distribution, bonded NdFeB, sieving, energy product

1. Introduction
The bonded NdFeB permanent magnet is one of the important part of functional materials which have been applied in various electromechanical application [1–3]. Bonded NdFeB permanent magnet typically uses polymeric matrix with magnetic NdFeB powder as a filler. There are two types of techniques to fabricate bonded NdFeB, such as compression moulding [4,5] and injection moulding [6,7].

The fabrication of bonded NdFeB was intended to make a high dense final product so that the larger energy product could be achieved. The starting particle size of NdFeB powder as a filler was kind of important factor to achieve good arrangement of isotropic bonded green body. In sintered type NdFeB, the fine particle with narrow distribution (\textasciitilde 3 μm) could increase magnetic coercivity effectively, as studied by Namkung et al. [8]. the study of Li et al., showed that the coarser particle distribution by
screening process yields a better magnetic energy product due to starting powder anisotropic effect on hot deformation process [9]. The element addition of NdFeB powder was also used to enhanced magnetic energy product by approaching exchange coupling interaction between hard-soft magnetic phases. Some kind elements such as Fe, Zr, Mn, Fe-Mn, and MnBi was introduced to NdFeB magnetic phases to obtained higher magnetic remanence and coercivity [10–14].

On the other hand, compaction and aging process were also important to make higher density and energy product of bonded NdFeB magnets. Various attempts have been made by some researchers to make denser green body. Herchenroeder et al., use combustion driven compaction to achieved higher density bonded NdFeB up to 10% compared to the typical commercial product [15]. The uses of low vacuum curing process was also effective in producing better bonded NdFeB density and higher magnetic properties [16]. However, there is lack discussion of the particle size distribution in the making of bonded NdFeB.

In this paper, we have investigated the effect of particle size distribution on the preparation of bonded NdFeB isotropic permanent magnets by compression moulding followed by low vacuum curing process. Physical and mechanical properties of bonded NdFeB were also observed. It is found that the middle range of particle size distribution is effective to obtain high quality bonded NdFeB permanent magnets.

2. Experiment
The NdFeB powder from Magnequench Inc. type MQP-B+ was used in the experiment. Normal MQP-B+ powder was composed by mixture of Nd-Fe-Co-B alloy with theoretical density of 7.63 g/cm$^3$. The powder was isotropic type magnetic powder so no external magnetic field needed to form permanent magnet green body. The varied standard test sieves of #100, #200, and #325 meshes were utilized to separate particle size distribution of the powders. For optimizing particle distribution, the mixture of passed powder of #100 and #200 mesh with composition ratio of 1:1 was also made for bonded magnet.

Bonded magnet green bodies were made by uniaxially compacted the NdFeB powder after mixed with 3 wt.% of epoxy resin binder under pressure of 749 MPa. Based on optimizing parameters for curing process in our previous study, the green bodies were aged in the vacuum oven ($\approx$10 mbar) in temperature of 100 $^\circ$C for 4 hours [16].

The final bonded magnet samples were characterized by XRD (Rigaku Smartlab) for analyzing magnetic crystalline phase, PSA (Cilas) for particle size distribution measurement, Archimedes’s method for bonded magnet density measurement, VSM (Dexing Magnet Ltd.) for analyzing magnetic hysteresis curve, SEM (Hitachi) for analyzing surface morphology and particle configuration, and Vickers hardness tester (LECO) for analyzing mechanical properties of bonded magnet samples.

3. Results and discussion
The XRD result as displayed in Figure 1 showed that the used starting powder of MQP-B+ is composed by Nd2Fe14B (2:14:1) magnetic phase compared to the data base number COD #96-100-8719. The composition of this 2:14:1 phase in the structure of bonded permanent magnet contributed to the high magnetic properties such as, magnetic flux density, remanence, and coercivity. Therefore, good crystallite structure is needed to produce high quality bonded permanent magnets [4].

The starting powder was measured its particle size distribution after sieved by multi-stage standard test sieved number 100, 200, and 325 meshes. The result of PSA analysis of the sieved powders is showed in Figure 2. It is showed in Figure 2(a), the starting powder has broad particle size distribution with 10% (Q1) and 90% (Q3) of cumulative values of 11.1 and 101.5 $\mu$m, respectively. On the other hand, median value (50% distribution or Q2) is around 42.4 $\mu$m, so that the starting powder has one

peak Gaussian particle distribution. The sieved powder particle distribution was also measured by PSA and depicted in Figure 2(b). It is showed that the increased of sieved number (smaller opening size) results a smaller particle size distribution. However, there is an overlap distribution size from one sieve to another. This is affected by particle shape which is not perfectly rounded, so the larger size could be passed through rectangular lattice of the sieves.
Each of the sieved powder distribution was compacted by compression molding method with 3 wt% of epoxy resin addition as a binder. After ageing process as mentioned in experimental detail section, the bonded magnet sample was measured its density by Archimedes method and the results were displayed in Figure 3. It is showed that the sample density is decreased as the particle size distribution decreased. This result was counter our intuitive that the smaller particle size should produce a higher density than the larger size. This is happened because of the contribution of narrow particle size and powder configuration in the bonded magnet products. However, the middle particle size distribution which composed by #100 and #200 meshes particle size with mix ratio 1:1 of mass produces highest density of 5.87 g/cm$^3$. Compared to the starting powder of MQP-B+ particle size distribution, the sample density was relatively lower to the sieved powder samples with average bonded magnet density of 5.7 g/cm$^3$. By analytic calculation of composite density of bonded magnets, the theoretical density of mixed Nd$_2$Fe$_{14}$B powder and 3 wt% of epoxy resin was around 6.71 g/cm$^3$. So the highest density of mixed #100 and #200 meshes powder could improve the bonded magnet density by 1.5% from 86% to 87.5% compared to the original particle distribution of MQP-B+ powder. The higher sample density is correlated to the higher magnetic properties of bonded NdFeB products.

The magnetic properties of bonded NdFeB magnet samples was measured by VSM as displayed in Figure 4 and the hysteresis curve was analyzed to achieved the intrinsic magnetic properties such as magnetic remanence, coercivity, and energy product (BHmax). It is showed from Figure 4(a) that the hard magnetic properties were dominant for all samples depicted by large hysteresis loops. From the Figure 4(b) as the enlargement of hysteresis loop, it is found that the remanence and coercivity are

![Figure 1. X-ray diffraction profile of NdFeB powder type MQP-B+](image)

![Figure 2. (A) Original particle size distribution histogram of MQP-B+ measured by Particle Size Analyzer and (B) particle size distribution of 10%, 50%, and 90% from different sieved powder of #100, #100 + #200, #200, and #325 meshes.](image)
decreased as the particle size distribution decreased. This phenomenon is agreed with sample density measurement. The detail value of magnetic properties is showed in Table 1. It is showed that the highest magnetic properties was produced by the mixed powder of #100 and #200 meshes with highest magnetic saturation, remanence, coercivity, and energy product of 8.40 kG, 6.49 kG, 11.16 kOe, and 9.40 MGOe, respectively. So the middle range of particle size distribution is effective in the making of bonded NdFeB permanent magnets.

Table 1. Magnetic properties of bonded NdFeB magnet with different particle size distribution.

| Description | $4\pi M_s$ (kG) | $4\pi M_r$ (kG) | $H_{rc}$ (kOe) | $BH_{max}$ (MGOe) |
|-------------|----------------|----------------|---------------|------------------|
| Original Powder | 7.76 | 5.58 | 10.28 | 6.88 |
| #100 mesh | 8.19 | 6.08 | 11.06 | 8.13 |
| #100+300 mesh | 8.40 | 6.49 | 11.16 | 9.40 |
| #200 mesh | 7.85 | 6.03 | 11.05 | 8.02 |
| #325 mesh | 7.60 | 5.66 | 10.77 | 7.09 |

Figure 3. Sample density of bonded NdFeB made from different particle size distribution by sieving process

To have a better analysis of powder configuration in bonded NdFeB system, the morphology of bonded magnet surfaces have been analyzed using SEM as showed in Figure 5. The different particle size distribution samples are used for comparison. It is showed that the smaller particle with narrow distribution produced a less density configuration in the epoxy matrix. The Figure 5(c,d) showed that the big empty filler of NdFeB powder was produce as shown as the dark spot in the SEM images. This emptiness reduced the density and magnetic properties of the bonded NdFeB samples. Compared to the larger size distribution in Figure 5(a,b), the filler configuration is relatively tight through the epoxy matrix. However, the narrower size from large particle only have made a wide range of filler emptiness as showed in Figure 5 (a). The SEM image also showed that the NdFeB powder has irregular shape. That is why the overlap intervals of sieved powder are existed.

Figure 4. Hysteresis measurement of bonded NdFeB with different particle size distribution by Vibrating Sample Magnetometer with (A) full hysteresis loop and (B) magnified second quadrant for enhanced view of remanences and coercivities.
Figure 5. SEM analysis of bonded NdFeB with different particle size distribution of (A) #100 mesh, (B) #100 + #200 mesh, (C) #200 mesh, and (D) #325 mesh. The right images are from the lower magnification of SEM to show the particle and epoxy binder arrangement of bonded NdFeB.

The mechanical analysis of bonded NdFeB samples are important to achieve good performance of magnet products in real application. So the Vickers hardness analysis had been performed for all kind samples as showed in Figure 6 under load of 300 gram-force for 13 seconds. It is showed that the smaller particle size distribution produce lower value of hardness. Compared to the original powder, the hardness of the sample reaches the value of around 117 HV. The hardness values for #100, #100+#200, #200, and #325 meshes type powder distribution are 102 HV, 88.7 HV, 84 HV, and 80.4 HV, respectively. So the highest value of density and magnetic properties not always produce higher mechanical properties of bonded NdFeB due to the lower hardness value of the epoxy resin binder [17].

Figure 6. Hardness measurement by microhardness vickers tester of bonded NdFeB with different particle size distribution.

4. Conclusion
In this study, the bonded NdFeB magnets were produced from different particle size distribution of NdFeB powders which was prepared by sieving method using standard test sieves of #100, #200, and #325 meshes. Varied distribution of particle size was mixed with 3 wt% of epoxy binder and compacted by uniaxial hydraulic pressing machine to produce bonded isotropic green body. It is showed that the sample density is decreased as the particle size distribution decreased. The magnetic properties are decreased as the particle size distribution decreased, except for the bonded magnet produce from the mixture of #100 and #200 meshes powder. The optimum characteristics of bonded NdFeB was obtained by the mixture of #100+#200 meshes powders with bulk density, hardness, magnetic remanence, coercivity, and energy product of 5.87 g/cm³, 88.7 HV, 6.5 kG, 11.15 kOe, and 9.4 MGOe, respectively. So the middle range of particle size distribution is effective in the making of bonded NdFeB permanent magnets.
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References
[1] Sugimoto S 2011 Current status and recent topics of rare-earth permanent magnets J. Phys. D. Appl. Phys. 44
[2] Lewis L H and Jiménez-Villacorta F 2013 Perspectives on Permanent Magnetic Materials for Energy Conversion and Power Generation Metall. Mater. Trans. A 44 2–20
[3] McCallum R W, Lewis L, Skomski R, Kramer M J and Anderson I E 2014 Practical Aspects of Modern and Future Permanent Magnets Annu. Rev. Mater. Res. 44 451–77
[4] Deng X, Liu Z, Yu H, Xiao Z and Zhang G 2015 Isotropic and anisotropic nanocrystalline NdFeB bulk magnets prepared by binder-free high-velocity compaction technique J. Magn. Magn. Mater. 390 26–30
[5] Ma B, Sun A, Lu Z, Cheng C and Xu C 2016 Research on anisotropic bonded Nd-Fe-B magnets by 2-step compaction process J. Magn. Magn. Mater. 401 802–5
[6] Nlebedim I C, Ucar H, Hatter C B, McCallum R W, McCall S K, Kramer M J and Paranthaman M P 2017 Studies on in situ magnetic alignment of bonded anisotropic Nd-Fe-B alloy powders J. Magn. Magn. Mater. 422 168–73
[7] Li L, Tirado A, Nlebedim I C, Rios O, Post B, Kunc V, Lowden R R, Lara-Curzio E, Fredette R, Ormerod J, Lograsso T A and Paranthaman M P 2016 High Performance Bonded NdFeB Magnets Sci. Rep. 6 1–7
[8] Namkung S, Kim D H and Jang T S 2011 Effect of particle size distribution on the microstructure and magnetic properties of sintered NdFeB magnets Rev. Adv. Mater. Sci. 28 185–9
[9] Li Y, Kim Y B, Wang L, Suhr D S, Kim T K and Kim C O 2001 Influence of the powder particle size on the anisotropic properties of NdFeB magnets produced by single-stage hot deformation J. Magn. Magn. Mater. 223 279–83
[10] Hou F, Li X, Zhang G, Hua Y, Li M, Lou L, Huang G, Li W and Zhang X 2018 Fabrication of bulk anisotropic Nd2Fe14B/α-Fe nanocomposite magnets with two-step high-pressure thermal compression J. Alloys Compd. 764 718–23
[11] Han J, Liu S, Wang C, Chen H, Du H and Yang Y 2009 Effects of the conventional HDDR process and the additions of Co and Zr on anisotropy of HDDR Pr-Fe-B-type magnetic materials J. Magn. Magn. Mater. 321 1331–4
[12] Guozhi X, Yuping W, Xiaoyan L, Zehua W, Pinghua L, Benxi G and Youwei D 2006 Ferromagnetic/antiferromagnetic exchange coupling in melt-spun NdFeB nanocomposites J. Non. Cryst. Solids 352 2137–42
[13] Kurniawan C, Purba A S, Setiadi E A, Simbolon S, Warman A and Sebayang P 2018 Effect of Fe-Mn addition on microstructure and magnetic properties of NdFeB magnetic powders Effect of Fe-Mn addition on microstructure and magnetic properties of NdFeB magnetic powders IOP Conf. Ser. J. Phys. Conf. Ser. 985 012044
[14] Nguyen T X, Nguyen K Van and Nguyen V Van 2018 Enhancement of exchange coupling interaction of NdFeB/MnBi hybrid magnets Phys. B Condens. Matter 532 130–4
[15] Herchenroeder J, Miller D, Sheth N K, Foo M C and Nagarathnam K 2011 High performance bonded neo magnets using high density compaction J. Appl. Phys. 109 1–4
[16] Kurniawan C, Hutahaean R M and Muljadi 2015 The Effect of Low Vacuum Curing to Physical and Magnetic Properties of Bonded Magnet Pr-Fe-B Adv. Mater. Res. 1123 84–7
[17] Zhang X H, Xiong W H, Li Y F and Song N 2009 Effect of process on the magnetic and mechanical properties of Nd-Fe-B bonded magnets Mater. Des. 30 1386–90