Synthesis and Characterization of Nd$_2$Fe$_{14}$B Powder by using Mechanical Milling of Flakes NdFeB

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Abstract. Flakes NdFeB is one of type NdFeB alloy made and it was produced by strip casting method. Flakes NdFeB can be used as a raw material to produce sintered permanent magnet and it has about 0.2 – 0.3 mm in thickness. The flakes of NdFeB was milled by using High Energy Milling (HEM) machine and used a liquid toluene as milling media. The milling time was varied such as : 15, 30, 60 and 90 minutes. After milling, the samples were dried by using vacuum dryer at temperature 80°C under vacuum (10 mmmbar) for 2 hours. The fine powder samples were analyzed crystall structure by using XRD, measured magnetic properties by using VSM and also measured particle size distribution (PSA). The characterization result show that the milling time gives an effect on the crystal structure and magnetic properties, where samples with milling time 60 dan 90 minutes has Fe phase as dominant phase and the Nd$_2$Fe$_{14}$B phase does not existing also the coercivity value tends to decrease until lower than 900 Oe. Samples with milling time about 15 and 30 minutes have Fe phase and Nd$_2$Fe$_{14}$B phase also they have coercivity value about 2000 – 2250 Oe. The means of particle size is achieved about 43.59 µm at sample with milling time 15 minutes.

1. Introduction
The Nd–Fe–B alloy with formula Nd$_2$Fe$_{14}$B is the most superior magnetic materials with tetragonal crystal structure [1,2]. The Nd$_2$Fe$_{14}$B is one type of permanent magnet based rare earth metals, it has high value of maximum energy product about 10 - 50 MGOe, magnetic remanence about 5 – 11 kGauss and curie temperature about 310°C. [1,3]. The permanent magnet Nd$_2$Fe$_{14}$B has highest magnetic properties compared to other type of permanent magnet such as : SmCo, AlNiCo and Ferrite. The disadvantages of permanent magnet Nd$_2$Fe$_{14}$B are easy to corrosion at room temperature and expensive in raw materials [3]. They are widely applied in various fields such as, medical equipments, information technology, hybrid or electrical vehicle (HEV or EV), energy-saving appliance, wind power generator and so on [4]. The synthesis of Nd$_2$Fe$_{14}$B alloy can be done by some of method such as : smelting using induction furnace, strip casting, melt spinning, mechanical alloying and sol gel process [5,6,7]. Flakes NdFeB is one of type NdFeB alloy and it was produced by strip casting method. Flakes NdFeB can be used as a raw material to produce sintered permanent magnet [8]. The NdFeB alloy in form flakes is more stable in environmental compared in the form powder, where if the NdFeB powder is more finer , it is very easy to oxidize and flammable, The milling of flakes NdFeB is important parameter in the manufacture of permanent magnet NdFeB. There are several ways of refining of NdFeB alloy materials : through a milling process using a jet mill, through the
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2. Experiments

Synthesis of alloy \( \text{Nd}_2\text{Fe}_{14}\text{B} \) powder was done using flakes NdFeB from China by using mechanical milling. Applied MA is through the process of High Energy Milling (HEM) with rotation speed 3000 rpm. The flakes NdFeB was weighing about 20 g, then inserted in a vial and toluene is added as much as 15 ml and added steel ball about 120 g. Furthermore, vial tube is closed and begins the milling process for: 15, 30, 60 and 90 minutes. The powder preparation from upload materials to vial tube until closing the vial tube have been done in Glove Box at vacuum condition (P vac = 10 mbar). The all samples after milling process were removed and continued with drying process using vacuum dryer at 100°C and 10 mbar for 2 hours, then fine powder samples were analyzed crystal structure by using XRD-Rigaku, measured magnetic properties by using Vibrating Sample Magnetometer (VSM) and also measured particle size distribution (PSA).

3. Results and Discussion

The samples powder obtained from milling process were measured particle size distribution and the results were shown at Figure 1.

Figure 1. Particle size distribution of Samples after milling (a) 15 minutes, (b) 30 minutes, (c) 60 minutes and (d) 90 minutes
According the results of measurement of particle size (Figure 1) show that the average particle size tends to decrease with increasing of milling time. Sample with variation milling time 15, 30, 60 and 90 minute has average particle size about 43.59 µm, 30.82 µm, 24.20 µm and 17.92 µm respectively. Based on research that uses a rotary ball mill can produce powder with the same size range but requires a longer time (12 hours) [11], but other studies using jet mill can produce smaller sizes in the same time[12]. The results analysis XRD of samples after milling are seen at Figure 2.

Figure 2. XRD patterns of Samples after milling process.
The XRD patterns of sample after milling 15 and 30 minutes as seen at Figure 2 are similar, it is found two phases, namely Nd$_2$Fe$_{14}$B phase and Fe phase by matching between experimental patterns with data reference of Nd$_2$Fe$_{14}$B (JCPDs 96-100-8719) and data reference of Fe (JCPDs 96-900-6602). But the XRD patterns of samples after milling 60 and 90 minutes are different with sample after milling 15 and 30 minutes, where only Fe phase is found but the Nd$_2$Fe$_{14}$B phase is not appeared, it is possible that the intensity of the Nd$_2$Fe$_{14}$B phase will decrease. Based on the results of XRD analysis, shows that the milling time can affect the NdFeB flake crystal structure, if the milling time is too long that is more than 30 minutes then the Nd$_2$Fe$_{14}$B phase decomposition will occur. This is because the longer the milling time, the finer particle size is obtained, so the more reactive the contact with oxygen, so that the crystal structure of Nd$_2$Fe$_{14}$B becomes damaged, the NdFeB based material is known to be very sensitive to oxygen.

The results of the analysis of magnetic properties measured using VSM from samples that have been milled at various times of milling and samples before being milled (NdFeB flakes) are shown in the form of a B-H curve in quadrant II region as shown in Figure 3.

![Figure 3. The B-H curve of samples after milling and before milling.](image)

Based on the results of B-H curve as shown in Figure 3 shows that sample flakes NdFeB before milling has a remanence value about 2150 Gauss and coercivity value about 1790 Oe. The samples after milling 15 and 30 minutes have higher remanence and coercivity value than flakes NdFeB, but the remanence and coercivity value of samples after milling 60 and 90 minutes tend decrease. It is due to changes in the Nd$_2$Fe$_{14}$B crystal structure after the milling process in 60 and 90 minutes. The highest remanence and coercivity values were achieved by 2800 Gauss and 2250 Oe in the sample powder after 15 minutes milling process. The magnetic properties (remanence and coercivity) of NdFeB produced from the NdFeB flakes material in this study, are still lower than the magnetic properties of NdFeB produced by melt spun NdFeB, which uses melt spun resulting in remanence of = 4500 Gauss and coercivity of = 8200 Oe[13].

4. Conclusion
The Nd$_2$Fe$_{14}$B powder has been made by using flakes NdFeB raw material through the milling process using HEM within 15 minutes with toluene milling media, in this condition Nd$_2$Fe$_{14}$B magnetic powder obtained with an average particle size of 43.59 um, remanence of Br = 20150 Gauss and coercivity of Hc = 2250 Oe. The Nd$_2$Fe$_{14}$B powder preparation from NdFeB flakes with a milling time longer than 30 minutes can damage the Nd$_2$Fe$_{14}$B crystal structure and reduce magnetic properties.
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