Reduced Remanence to Coercivity Increase Ratio of \( \text{Pr}_{11.76} \text{Dy}_x \text{Fe}_{82} \text{B}_6 \) (\( x = 0; 1; 2; 3 \)) Permanent Magnets

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Abstract. Dysprosium substituted \( \text{Pr}-\text{Fe}-\text{B} \) permanent magnet samples with \( \text{Pr}_{11.76} \text{Dy}_x \text{Fe}_{82} \text{B}_6 \) (\( x = 0, 1, 2 \) and \( 3 \)) composition were made up by compacting the crystalline fine powders prepared through mechanical milling and successive high power ultrasonic irradiation. The \( (\text{Pr},\text{Dy})-\text{Fe}-\text{B} \) alloys with designated compositions were obtained from arc melted the charges that consisted of respectively \( \text{Dy}-\text{Fe}, \text{Fe}_3\text{B} \) alloys and pure \( \text{Fe} \) and \( \text{Pr} \) metals. Results of the xrd analysis are permanent magnetic samples confirmed that the mass of the compound fraction formed is 90.07%. A change in lattice parameter of \( \text{Pr}_{2}\text{Fe}_{14}\text{B} \) phase occurred when \( \text{Pr} \) was partially substituted by \( \text{Dy} \). Furthermore, the effect of \( \text{Dy} \) partial substitution on magnetic properties can increase the coercivity value, but with a decrease in saturation magnetic. \( \text{Dy} = 3 \) substitution produces coercivity and remanence values of 160 kA / m and 0.275 T respectively in the permanent magnet category.

Keywords: permanent magnets, \( \text{NdFeB} \), coercivity, sintered magnets, magnetic properties

1. Introduction
Praseodymium Iron Boron is a magnetic alloy that promises for permanent magnet application and supporting electronic devices, this is because this material has the same crystal structure with \( \text{NdFeB} \), also the same coercivity and remanence on room temperature \([1]\). \( \text{NdFeB} \) is a superior permanent magnet to replace \( \text{SmCo} \) in electric vehicle applications \([2]\) and until recently no one compound that replaced it. \( \text{NdFeB} \) has a low currie temperature while the application of an electric motor requires high room temperature coercivity. Recently, a newly developed process called the grain boundary diffusion process (GBDP), provides a suitable solution for the problems mentioned above by placing \( \text{Dy} \) between \( \text{NdFeB} \) grains \([3]\). Although the saturation magnetization (\( \text{Ms} \)) and the maximum energy product (\( (\text{BH})_{\text{max}} \)) of \( \text{Nd-Fe-B} \) magnets decreases with increasing \( \text{Dy} \) content, \( \text{Dy} \) is usually used for the improvement of the coercivity in \( \text{Nd-Fe-B} \) magnets. \([4]\) In particular in the application of permanent magnet, high coercivity field (\( \text{Hc} \)) is a very basic characteristic because \( \text{Hc} \) is one of the main parameters for maximum energy product (\( (\text{Bh})_{\text{max}} \)) besides magnetic remanence. \([5]\) A lot of research that attempts to increase the coercivity field include adding \( \text{Dy} \) to \( \text{NdFeB} \) compound \([6]\). In this experiment dysprosium was substituted to \( \text{PrFeB} \) to increase the coercivity field. Samples were analyzed using X-Ray powder diffraction (XRD), surface morphology using optical microscopy,
and magnetic loop hysteresis curves using Permagraph Magnet-Phisyk which were carried out at room temperature. So the purpose of this experiment is to determine the effect of phase changes and microstructure on changes in the intrinsic properties of alloyed magnetic materials Pr$_{11.76-x}$Dy$_x$Fe$_{82}$B$_6$.

2. Experimental Method
Dysprosium is substituted for Paraesidimyum Iron Boron (Pr$_{11.76-x}$Dy$_x$Fe$_{82}$B$_6$) with several variations of composition $x = 0, 1, 2, \text{ and } 3$. The samples Pr$_{11.76-x}$Dy$_x$Fe$_{82}$B$_6$ are made by Pr, Dy, Fe and B commercial ingots and melted using arc melting in the Argon environment, then the ingots are crushed and milled using planetary Ballmill for 6 hours with a revolution speed of 500 RPM and an evolution speed of 1000 RPM to get the permanent magnetic powder. After being powdered, it was compressed at a pressure of 5 tons and synthesized in a vacuum furnace 1100 °C for 2 hours, stoichiometry calculations for the substitution variation of the day are shown in Table 1.

| composition $x$ | Samples name       | Mass of raw materials (gram) |
|-----------------|---------------------|-------------------------------|
| $x$             | Pr$_{11.76}$Fe$_{82}$B$_6$ | Pr | Dy | Fe | B |
| 0               | Pr$_{11.76}$Fe$_{82}$B$_6$ | 3.9446 | 0 | 10.9009 | 0.1544 |
| 1               | Pr$_{10.76}$Dy$_1$Fe$_{82}$B$_6$ | 3.5968 | 0.3855 | 10.8637 | 0.1538 |
| 2               | Pr$_{9.76}$Dy$_2$Fe$_{82}$B$_6$ | 3.2515 | 0.7684 | 10.8267 | 0.1533 |
| 3               | Pr$_{8.76}$Dy$_3$Fe$_{82}$B$_6$ | 2.9084 | 1.1487 | 10.7900 | 0.1528 |

Phase analysis of the samples was measured using an XRD device which was used by the Brucker brand with an X-ray Cu anode anode ($\lambda = 1.5406$ Å) with a step size of 0.01. Quantitative analysis was carried out using a general structure analysis system (GSAS) software. Surface morphology using the JEOL JED 350 SEM brand, and magnetic properties testing using a Permagraph Magnet-Physik 255 with a range of -0.6 to 0.6 tesla.

3. Results and Discussions
The X-ray diffraction pattern (XRD) of the Pr$_{11.76-x}$Dy$_x$Fe$_{82}$B$_6$ alloy sample for $x = 0 - 3$ is shown in Figure 1. The results of the qualitative analysis using the Match program found that all samples consisted of the neodymium iron boron phase. The peaks of the XRD pattern of all samples are very dominant corresponding to the ref data of JCPDS 96-100-8719 forming the tetragonal structure Pr$_2$Fe$_{14}$B. But all samples have three phases, Pr$_2$Fe$_{14}$B, Fe, and Pr.

![Figure 1. Sample X-ray diffraction patterns of Pr$_2$Dy$_x$Fe$_{14}$B ($x = 0$–$3$).](image-url)
The result of phase identification shows that the arc melting process has succeeded in forming the dominant phase Pr$_{11.76-\text{x}}$Dy$_\text{x}$Fe$_{82}$B$_6$ in the composition x = 0 - 3. It can be seen in Figure 1 that the greater the composition of Dy, the number of minor phase contents decreases, so further analysis is needed, determine the phase change, the amount of mass fraction formed, and the influence of Dy substitution into the Pr atom.

In Figure 2 shows the results of the improvement of X-ray diffraction patterns from the sample Pr$_{11.76-\text{x}}$Dy$_\text{x}$Fe$_{82}$B$_6$ with variations in composition (x = 0 - 3). Figure 2 (a-d) is the result of the refinement of the XRD pattern for x = 0-3 which has formed the Bragg diffraction peak with the dominant phase following the structure of Pr$_{11.76-\text{x}}$Dy$_\text{x}$Fe$_{82}$B$_6$. Qualitative and quantitative analysis refers to Open Database Crystallography with card numbers (JCPDS: 00-043-0002), (JCPDS: 00-033-0664), and (JCPDS: 00-037-1493) for phase Pr$_{11.76}$Fe$_{82}$B$_6$ respectively, Fe, and Pr.

A complete summary of the results of the refinement of the X-ray diffraction pattern from the Pr$_{11.76-\text{x}}$Dy$_\text{x}$Fe$_{82}$B$_6$ sample with variations in composition (x = 0 - 3) for all samples is shown in Table 2.

| Sample (x) | Phase | Lattice parameter (Å) | V (Å$^3$) | $\rho$ (g/cm$^3$) | Fraction wt% | Rwp (%) | $\chi^2$ |
|-----------|-------|-----------------------|--------|-----------------|--------------|--------|-----|
| 0         | Pr$_{11.76}$Dy$_\text{x}$Fe$_{82}$B$_6$ | 8.822(2) 8.822(2) 12.261(3) | 954.4(6) | 7.524 | 81.11 | 19.26 | 2.28 |
|           | Fe    | 2.8716(4) 2.8716(4) 2.8716(4) | 23.68(1) | 7.832 | 18.61 |        |       |
|           | Pr    | 3.730(5) 3.730(5) 11.211(3) | 135.0(5) | 10.399 | 0.28  |        |       |
Figure 2 and Table 2 show that the results of refinement of X-ray diffraction patterns have very good fitting quality according to the criteria of fit (Rwp) and goodness of fit ($\chi^2$). Rwp is the ratio of the weight of the difference between the pattern of observation and calculation of XRD (ideal value of Rwp <10%). Whereas $\chi^2$ (chi-squared) is the ratio of the XRD pattern observations that are comparable to expectations.

In Figure 3 is a sample observation using a 200x magnification optical microscope and shows that the distribution of the grain boundary of the composition $x = 0$ - 0.3 has very good and uniform particle homogeneity across the sample surface but there is another element in the compound boundary with the estimated Fe and Pr regardless of the results of the XRD analysis. The diameter of the grain decreases relative to the addition of Dy metal.

![Figure 3](image-url)

**Figure 3.** The morphology of the Pr$_{11.76-x}$Dy$_x$Fe$_{82}$B$_6$ particles observed using an optical microscope a) $x = 0$; b) $x = 1$; c) $x = 2$; and d) $x = 3$.

In addition, the presence of Dy can influence the magnetic properties of this system. In Figure 4 shows the hysteresis curve measured by magnetic properties after sintering 1100°C celcius for 2 hours using permagraph Magnet-Phisyk. All samples have a high magnetic saturation value, and the value of the remanent magnet is not less than half the value of magnetic saturation. Samples with the composition $x = 4$ have remanent and coercivity values in the category of permanent magnets [7], to
increase the characteristic value in the category of permanent magnet requires proper sintering and annealing process for grain growth and grain boundary [8,9].

**Figure 4.** Hysteresis curve of the Pr$_{11.76}$Dy$_x$Fe$_{14}$B.

The addition of Dy substitution to compound composition causes an increase of coercivity but decreases of magnetic saturation [10,11] as shown in Figure 5.

**Figure 5.** Ratio between saturation and coercivity.

Figure 5 shows the ratio between saturation magnetic and coercivity with the addition of Dy, the significant coercivity value increases while the saturation magnetic significantly decreases, and Dy $x = 4$ has reached the saturation and coercivity values of the permanent magnet category characterized by a high remanent value, also Mr = 0.275 Tesla. It can be said that the coercivity value is inversely proportional to the saturation magnet or the remanent magnet.
4. Conclusion
Successfully made by dy-sintered permanent magnet PrDyFeB by mixing the weight percent of elements of Pr, FeDy and FeB alloys to form Pr11.76-xDyxFe82B6 compound with variations x = 0-3. The results of the XRD analysis showed that there were still three phases of PrDyFeB, Fe and Pr with a weight percent fraction of about 90% PrDyFeB. All experimental results have shown high saturation magnetic values and substitution of x = 3 has been able to show the properties of permanent magnets with a remanent value of 0.275 T and a coercivity value of 160 kA / m, still requiring an annealing process to increase these values.

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