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Influence of Milling Time on Magnetic Properties and Microstructure of Sintered Nd-Fe-B Based Permanent Magnets

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Abstract. In this paper, the effect of the grain size on sintered Nd-Fe-B based permanent magnets was investigated. In order, the magnets were produced by different milling times at 200 rpm and then vacuum sintered at 1373 K for 60 minutes followed by cooling outside the furnace. The magnets either produced by lower and higher milling times (30 and 75 minutes) exhibited lower remanence and coercivity, due the inhomogeneous distribution of the grain sizes. The magnet produced by intermediary milling time (45 minutes) exhibited the highest properties among all samples, with remanence of 1.06 T, coercivity of 891.3 $KA.m^{-1}$, maximum energy product of 211 $KJ.m^{3}$ and a squareness factor equal 0.92.

Introduction

Sintered Nd-Fe-B based permanent magnets are well known by its excellent magnetic properties and its applications in many technological fields [1–3]. It has been found that the microstructure of the sintered Nd-Fe-B magnet plays an important role on the magnetic properties. Since the powder metallurgy route it is the main route to produce permanent magnets, the HD process and the milling step are vital to produce the most proper microstructure in the sintered magnet. The magnetic properties are strongly related to the grain size and its distribution [4,5]. It was found that insufficient milling times are deleterious to the magnetic properties due heterogeneities of particle sizes (even at high speeds). However, long milling times are also detrimental due the destruction of the surface of the grains and abnormal grain growth during the sintering [6,7].

Experimental

A $(Nd,Pr)_{13.49}Fe_{77.72}B_6Co_{1.1}Al_{1.2}Nb_{0.28}Cu_{0.2}$ strip-cast alloy was used to produce the permanent magnets. In order, 15 g of the bulk material was placed in a stainless steel hydrogenation vessel which was evacuated to backing pump. Hydrogen was then introduced to a pressure of 2 Bar which resulted in decrepitation of the bulk material. The decrepitated hydride material was transferred to a planetary ball mill (Fritsch Pulverisette 5) and milled at 200 rpm for several times using cyclohexane as milling medium. The milling times were varied from 30 to 75 minutes with steps of 15 minutes. The ball-to-powder weight ratio was kept constant for all experiments (10:1). The resultant powders were dried for about 30 minutes and then transferred to cylindrical rubber tube under nitrogen atmosphere. The powders were then pulsed at a 6 T magnetic field, isostatically pressed at 200 MPa, vacuum sintered at 1373 K for 60 minutes followed by cooling outside the furnace.

Magnetic characterization of the HD sintered permanent magnets was carried out using a permeameter. Remanence (B_r), intrinsic coercivity ($\mu_{0i}H_c$), maximum energy product (BH_{max}) and squareness factor have been obtained from the second quadrant of the hysteresis curve obtained from a permeameter. Microstructural observations were carried out using a scanning electron microscope (SEM). Grain size measurements were carried out using an image analyzer (Image J). The samples for grain analysis were etched with Marble solution in order to reveal the grain boundaries.

Results and Discussion

The demagnetization curves of the permanent magnets produced are show in Fig.1. The values of remanence and coercivity start to increase, reach a maximum and then diminish, with the increase of milling time. The magnetic properties are presented in Figs.2a and 2b. The maximum value of remanence was 1.06 T (10.6 KOe) for the magnet produced by 45 minutes of milling time. The maximum value of coercivity was 907.2 $K.A.m^{-1}$(11.4 KOe) for the magnet produced by 60 minutes of milling time. Both the samples exhibited the highest values of maximum energy product, which was 211 KJ/m^3 and 199 KJ/m^3, respectively. The magnetic properties are listed in Table 1.

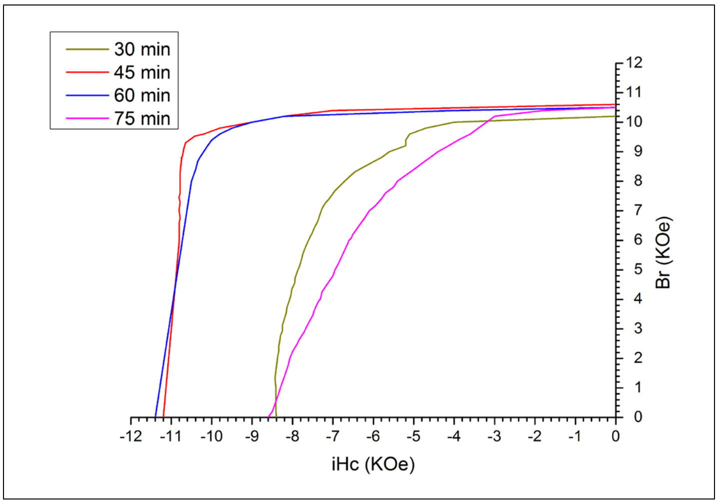

Fig. 1. Demagnetization curves of the samples produced by 30, 45, 60 and 75 minutes, respectively.

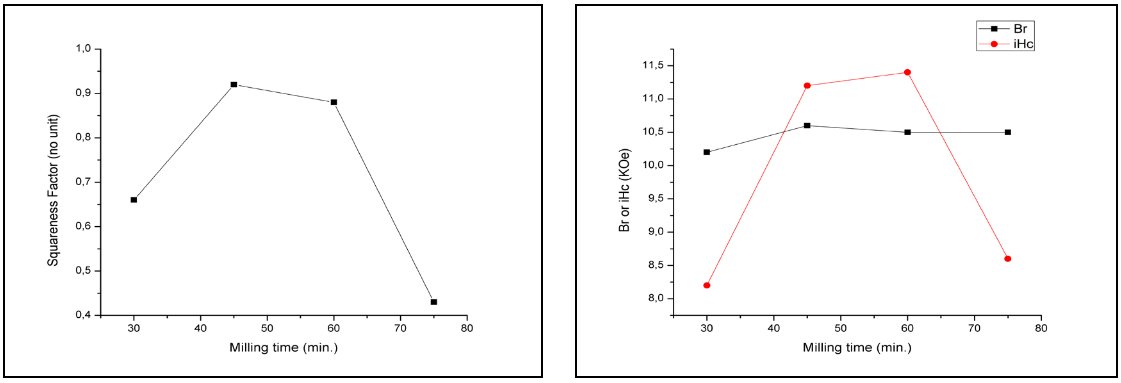

Fig. 2. (a) variation of remanence and coercivity and (b) squareness factor in function of milling time.

This result shows the fact that exist an optimum milling time which magnetic properties are higher. Many works reported the effects of insufficient or exaggerated milling times on the microstructure and magnetic properties of sintered magnets. The deleterious effects on the magnetic properties are due the heterogeneities of grain sizes (lower milling times) and abnormal grain growth (higher milling times) [6–8].

Table 1. Magnetic properties of the sintered permanent magnets produced by 30, 45, 60 and 75 minutes of milling time, respectively.

Milling time (min.)	B_r (T)	$\mu_{0i}H_c$ (KA/m)	$_bH_c$ (KA/m)	BH_{max} (KJ/m^3)	Squareness factor
30	1.02	652.5	572.9	143	0.66
45	1.06	891.3	779.8	211	0.92
60	1.05	907.2	771.9	199	0.88
75	1.05	684.4	509.3	135	0.43

The microstructures of the sintered magnets are shown in Fig.3a-d. It has been verified that low milling times led to heterogeneities of particle size, as can been seen in Fig.3a. It has also observed that high milling times led to the same heterogeneities, as can been seen in Fig.3d. Intermediates milling times (45 and 60 minutes) led to more homogeneous grain size distribution, as can been seen in Fig.3b-c. This effect is clearly seen on the magnetic properties, since coercivity and squareness factor are strongly dependent on grain size and format [6]. The higher magnetic properties were presented by the magnets produced by intermediate milling times, which resulted in more homogeneous grain size distribution with lower standard deviations, shown in Table 2.

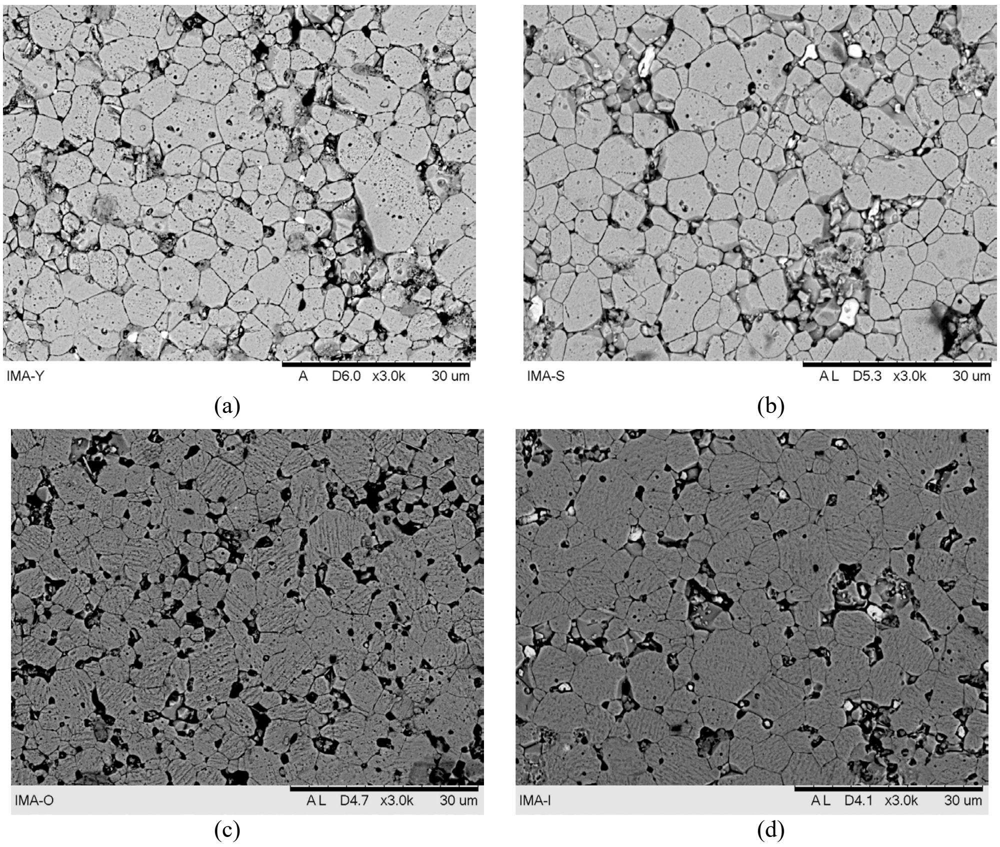

Fig. 3. General view of the microstructure of the magnets produced from decrepitated material milled for (a) 30 minutes; (b) 45 minutes; (c) 60 minutes and (d) 75 minutes.

Table 2. Mean grain sizes in function of milling time.

Milling time (min.)	Mean grain size (μm)	Standard deviation (μm)
30	5.61	2.88
45	5.31	1.98
60	5.73	1.92
75	6.12	2.64

Conclusion

The influence of several milling times of the strip-cast alloy employed in the production of sintered Nd-Fe-B based permanent magnets was investigated. It is clear that, employing the appropriate parameters of processing, in this case the milling time, it is possible to improve the magnetic properties of sintered magnets. The highest overall magnetic properties were obtained for the magnet produced with the alloy hydrogen decrepitated and milled at 200 rpm for 45 minutes. Intermediate milling times improved the grain size distribution, resulting in the gain of magnetic properties. Also, the squareness factor was dramatically improved as the grain size distribution was improved, clearly showing the correlation between microstructure and magnetic properties.

Acknowledgments

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