Materials Science Forum
ISSN: 1662-9752, Vol. 899, pp 567-571
doi:10.4028/www.scientific.net/MSF.899.567

Submitted: 2016-12-29
Accepted: 2016-12-29
Online: 2017-07-03

Dry and Wet Milling Comparison of Nd-Fe-B Magnets Based on Strip Cast Alloys

Maccari F.[1,a], Well R.V.[1,b], Eller G.[1,c], Hoffmann M.S.T.[1,d,]; Lopes L.U.[2,e], Takiishi H.[2,f] and Wendhausen P.A.P.[1g]

[1] Mechanical Engineering Department, Federal University of Santa Catarina – UFSC Florianópolis, Brazil.

[2] Nuclear Energy Research Institute - IPEN, São Paulo, Brazil.

[a]fernando.maccari@yahoo.com.br, [b]raphavanwell@gmail.com, [c]gabieller@hotmail.com, [d]marinasthoffmann@gmail.com, [e]leonardo.ulian@gmail.com, [f]paulo.wendhausen@ufsc.br

Keywords: NdFeB magnets, Strip Cast, Magnetic Texture, High Energy Milling

Abstract. In this work, the influence of milling medium was investigated in order to achieve high energy-product Nd-Fe-B magnets, mostly by the remanence improvement related to the obtention of monocrystalline particles during milling. Nd-Fe-B alloy made by strip-casting process were used as starting material, which exhibits refined grain structure and demands special attention during milling in relation to coarse-grained, conventionally cast alloys. It was found that by using liquid medium during ball milling process, the mean particle size decreased, as well as the size distribution, which improved the particle alignment and hence the remanence in the sintered magnet. Texture was quantified by magnetic characterization based on reference isotropic magnets made in same conditions, and microstructure development was evaluated by optical microscopy. Moreover, after post-sintering treatment, the coercivity increased 20% compared to as sintered state without changing the remanence, providing an anisotropic magnet with high coercivity.

Introduction

High-performance, Nd-Fe-B based permanent magnets have been thoroughly studied and widely used in many applications, including electric motors and windmill generators. In order to maximize magnetic properties, the adequate microstructure needs to be tailored by the use of adequate starting materials and the control of powder metallurgy processing route. For starting materials, alloys produced by strip-casting technique show advantages on eliminating free-iron precipitation during cooling [1-3], thus eliminating additional processes, like time consuming heat treatments aiming alloy homogenization. However, the small grain size achieved by this rapid-cooling process can be a concern for the obtention of monocrystalline particles after the milling step, which is critical for texture development and high coercivity of the magnet [4-5]. Jet-milling process is widely used in industry and R&D and yields excellent particle size control and texturization, albeit costly and unable to operate for small powder volumes [6-10]. An alternative is the use of ball milling, but its characteristic wider particle size distribution must be controlled to achieve effective separation of small grains from strip cast alloys, hence allowing an effective particle alignment. In this study, the effectiveness of high-energy ball milling in different mediums was investigated aiming to achieve high degree of alignment and increase magnetic properties values in sintered Nd-Fe-B magnets.

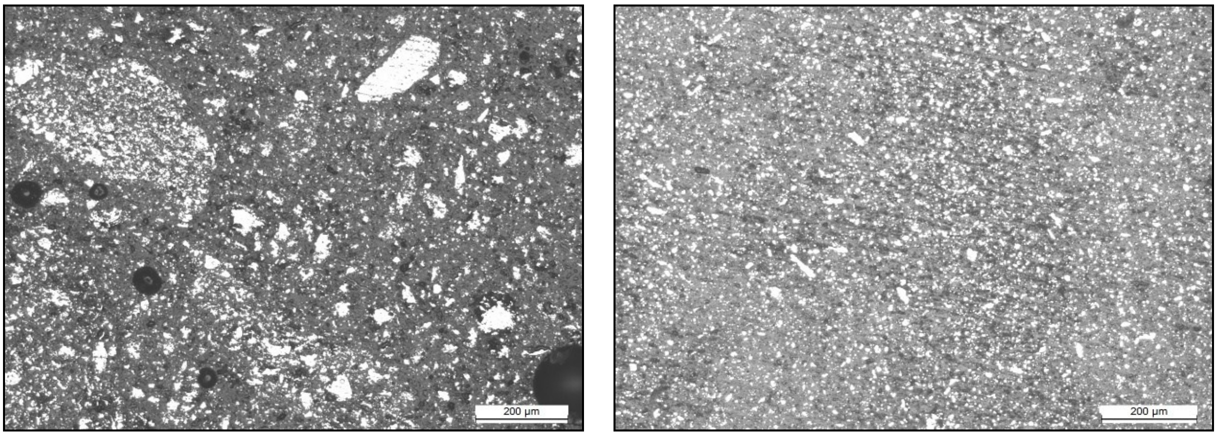
Experimental

In order to, investigate the proper ball milling conditions for fabricating Nd-Fe-B magnets, a comparison of the milling process und dry and wet mediums was performed. In this study, a $Nd_{30}Fe_{67}Co_1Al_1B_1$ strip-cast alloy was employed. After hydrogen decreptation (HD) process, the material was milled in high-energy planetary ball mill, using a BPR of 7.5 and rotation of 300 RPM for 1 hour. For sample A, the material was dry milled in argon atmosphere and for sample B, hexane was used as a liquid medium during the milling process. Powder milled under hexane underwent a drying process by a vacuum pump. The fine powders were aligned in a magnetic pulsed field of 4.5 T and then isostatically pressed under 150 MPa in order to fabricate anisotropic green compact. Isotropic magnets for reference purposes were also manufactured by skipping the aligning step. The specimens were sintered under a temperature of 1050 °C during 2 hours and pressure below 10^{-5} mbar. A post sintering heat treatment was employed in wet milled powders at 550°C for 1 hour.

Densities of the sintered specimens were measured using Archimedes displacement principle in water, and magnetic properties were evaluated by means of a hysteresigraph, after a magnetization step by a pulsed field with intensity of 4.5 T. The powder appearance and the microstructure of sintered magnets were evaluated using optical microscopy.

Results and Discussion

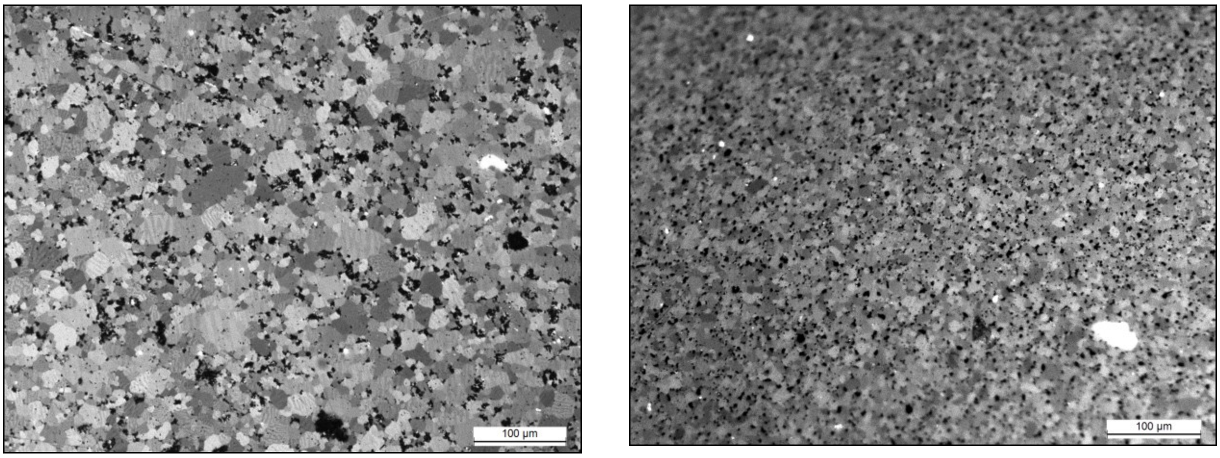
Optical microscopy pictures of alloy after milling under dry and wet conditions are shown in Fig. 1. From the pictures, it can be noticed a wider particle size distribution given by dry milling process (Fig 1-a) when compared to wet milling (Fig 1-b). Moreover, mean particle size is also larger in dry milling process, as well as the existence of very large, not milled particle of around 200 µm. The use of liquid medium during milling was proved more efficient due to the creation of a suspension of Nd-Fe-B particles with the solvent. This fact prevents the agglomeration of particles and clinging to grinding vessel, improving milling efficiency.



(a) (b)

Fig. 1: Micrographies of Nd-Fe-B powders produced by (a) dry milling and (b) wet milling.

After aligning, pressing and sintering the powders milled in both conditions, the microstructures were evaluated and are shown in Fig. 2. The differences between particles' size and distribution in dry-milled powder lead to large and heterogeneous grain size (Fig. 2-a) and more homogenous and smaller grains in wet milled powder, a consequence of de conditions of each powder.



(a) (b)

Fig. 2: Polarized light micrographies of Nd-Fe-B sintered magnets produced by (a) dry milling and (b) wet milling.

Narrow size distribution and lower mean particle size are key to obtain high coercivity, related to the nucleation of reverse domains that is facilitated for larger grains. In order to evaluate the impact of the different microstructures in the magnetic properties, demagnetization curves of isotropic and anisotropic samples made from powders milled in the two conditions were measured and are shown in Figs. 3-a and 3-b. Table 1 summarizes the main magnetic properties. Isotropic magnets allow a more independent evaluation of the coercivity, whereas anisotropic magnets show the overall result of the combination of magnetic hardness and texture development.

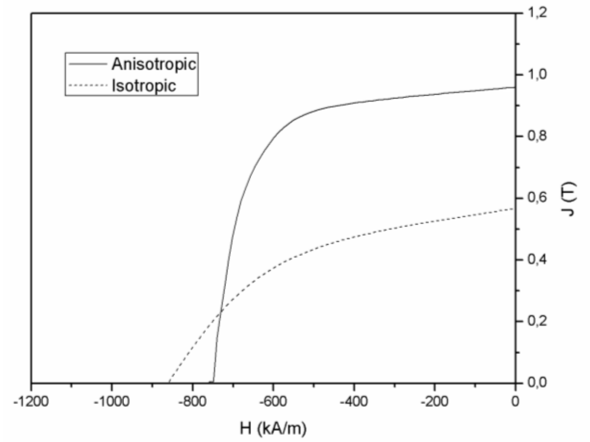


Fig. 3-a: Dry Milling - Demagnetization curve of isotropic and anisotropic samples.

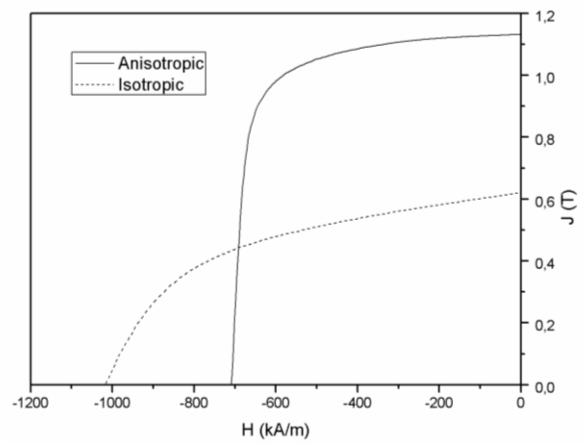


Fig. 3-b: Wet Milling - Demagnetization curve of isotropic and anisotropic samples.

Table 1: Magnetic properties of sintered magnets.

Sample	Iso/Aniso	Br (T)	H_{cJ} (kA/m)	$(BH)_{máx}$ (kJ/m³)	Density (g/cm³)
A (dry milled)	Isotropic	0.57	861	55	7.47
A (dry milled)	Anisotropic	0.96	760	167	7.46
B (wet milled)	Isotropic	0.62	1,017	66	7.47
B (wet milled)	Anisotropic	1.13	709	233	7.47

It can be seen, by comparing both isotropic magnets in Figs. 3-a and 3-b that the coercivity is higher for the sample mate with powder produced by wet milling, achieving values over 1000 kA/m. This result indicates the importance of the microstructure for coercivity (Fig. 2-b) with a smaller mean grain size and narrower distribution.

When comparing the magnetic characterization of anisotropic magnets, it is clear that the Sample B (Fig. 3-a) show a remarkable remanence increase, over 17% higher than sample A.

Considering that similar densities were achieved (Table 1), the difference is related to the higher texture present in sample B. The difference can be explained by the more efficient single-crystal particles generated by wet-milling, which in turn is easier rotated to the aligning field direction previous to the pressing stage. Strip-cast alloys show very narrow grains in a columnar structure, which require an efficient milling process to a proper single-crystal particle formation. If large particles are present, as can be seen in Fig. 1-a, those particles are definitely polycrystalline and would not rotate in the presence of the aligning field offering poor texture in the sintered magnet.

The application of a post-sintering treatment to the sample with best properties was also evaluated, and the magnetic evaluation is shown in Fig. 4, compared to the demagnetization curve in the as-sintered condition. The 1-hour, 550 °C heat treatment lead to the improvement of coercivity from 709 kA/m to 920 kA/m, an almost 30% improvement. This process is related to the grain boundary phase redistribution, performing a better grain decoupling and reducing the nucleation of reverse domains, which increases coercivity.

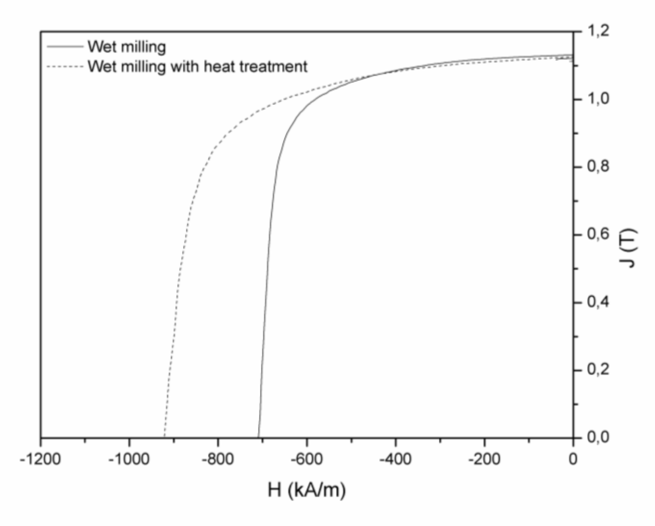


Fig. 4: Magnetic measurements of the effect of post-heat treatment application.

Conclusions

By using hexane as a wet medium during high-energy ball milling, a smaller particle size and narrower distribution was obtained, in relation to dry-milling, mostly due to particle agglomeration and clinging that reduced milling efficiency. The powder characteristics were noticed in the microstructures of sintered magnets made with them, in terms of grain size and distribution. Magnets made with wet-milled powder showed higher coercivity and higher remanence, leading to higher BH-max, due to better texturization provided by more efficient single-particle formation during wet-milling, and smaller and homogeneous grain size, which led to higher coercivity.

References

[1] L.Q. Yu, M. Yan, J.M. Wu, W. Luo, X.G. Cui, and H.G. Ying: Phys. B Condens. Matter Vol. 393 (1–2) (2007), p. 1.

[2] W. Pei, C. He, F. Lian, G. Zhou, and H. Yang: J. Magn. Magn. Mater. Vol. 239 (2002), p. 475.

[3] H. Chiriac and M. Marinescu: J. Non. Cryst. Solids Vol. 287 (1–3) (2001), p. 140.

[4] E.A. Périgo, E.P. Soares, R.M. Leal Neto, C.C. Motta, and R.N. de Faria Jr.: Mater. Res. Vol. 10 (3) (2007), p. 311.

[5] K. Uestuener, M. Katter, and W. Rodewald: IEEE Int. Magn. Conf. Vol. 42 (10), p. 228, 2006.

[6] K. Hirota, H. Nakamura, T. Minowa, and M. Honshima: IEEE Trans. Magn. Vol. 42 (10) (2006), p. 2909.

[7] T.H. Kim, S.R. Lee, S. Namkumg, and T.S. Jang: J. Alloys Compd. Vol. 537 (2012), p. 261.

[8] H. Nakamura, K. Hirota, T. Ohashi, and T. Minowa: J. Phys. D. Appl. Phys. Vol. 44 (6) (2011), p. 064003.

[9] F. Vial, F. Joly, E. Nevalainen, M. Sagawa, K. Hiraga, and K. T. Park: J. Magn. Magn. Mater. Vol. 242–245, no. Part II (2002), p. 1329.

[10] Z.H. Cheng, M.X. Mao, C.L. Yang, Y. De Zhang, F.S. Li, B.G. Shen, J.X. Zhang, and J.J. Sun: J. Appl. Phys. Vol. 76 (1994) (1994), p. 2981.