

The production of rare-earth-sintered magnets by a low-cost powder technique

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Abstract

This paper describes a simple system which may be constructed in the laboratory to produce rare-earth iron–boron permanent magnets. Unlike conventional preparation of sintered magnets in laboratory, the technique described does not require a glove box. Sintered Nd-based magnets have been successfully produced using this new technique and the hydrogen decrepitation (HD) process. The technique proved to be very useful for studying magnets on a laboratory scale.

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1. Introduction

Hydrogen decrepitation (HD) followed by jet or roller milling is well established as an effective route of powder preparation [1–3] and it is now widely used in production of Nd–Fe–B-sintered magnets both commercially and on a laboratory scale [4]. The powder handling stages of the processing have proved to be the most difficult to control due to the rapid oxidation which can take place with fine rare-earth-containing powders. The production of HD-sintered magnets on a laboratory scale requires a good inert atmosphere glove box with an oxygen content of less than 30 ppm [3]. To maintain this oxygen level the glove box must be operated using a continuous oxygen-free nitrogen supply flowing through the main chamber. Transfer of materials into and out of the chamber is via ports which can be evacuated to backing-pump pressure and backfilled with nitrogen to retain the low oxygen concentration inside the main chamber. The glove box is essential during the transference of the highly reactive fine milled hydride powder to the rubber tube used for isostatic pressing. A glove box, which can be routinely operated at oxygen levels below 30 ppm, has a considerable capital, operational and maintenance cost. In the present work a technique for

preparation of magnets in the laboratory without a glove box, has been developed. A system has been constructed to allow the fine hydride powder to be dried and transferred from the roller milling pot to the isostatic rubber tube without being exposed to air. Hydrogen decrepitation-sin-

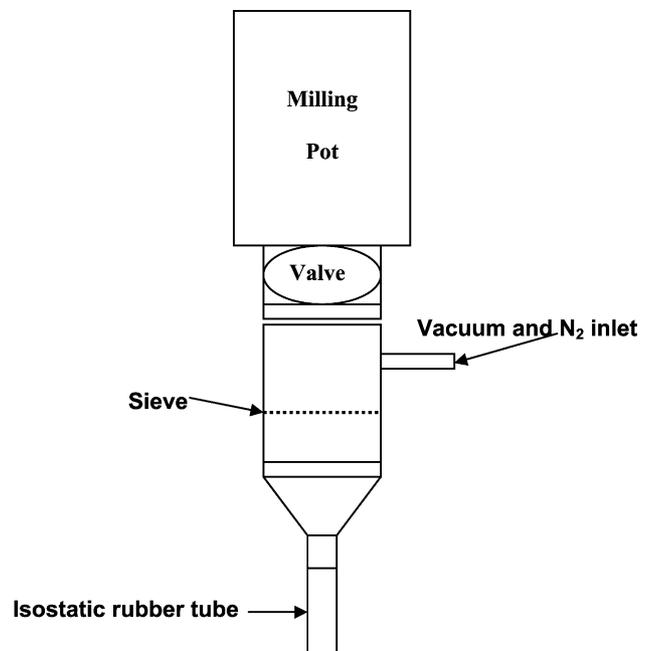


Fig. 1. Schematic drawing of the powder transfer apparatus.

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Table 1
Magnetic properties of Nd_{14.5}Dy_{1.5}Fe₇₆B₇Nb₁ HD sintered magnets

| Milling time [h] | B_r (± 10) [T] | $\mu_{oi}H_c$ (± 10) [T] | $\mu_{ob}H_c$ (± 10) [T] | μ_0H_k (± 10) [T] | SF = H_k/iH_c [ratio] | $(BH)_{max}$ (± 10) [kJ/m ³] | Density (± 0.02) [g/cm ³] |
|------------------|------------------------|--------------------------------|--------------------------------|-----------------------------|-------------------------|--|---|
| 6 | 1.12 | 1.40 | 1.11 | 1.24 | 0.89 | 249 | 7.40 |
| 12 | 1.14 | 1.56 | 1.11 | 1.42 | 0.91 | 252 | 7.53 |
| 18 | 1.18 | 1.50 | 1.15 | 1.35 | 0.90 | 271 | 7.52 |
| 24 | 1.20 | 1.40 | 1.18 | 1.27 | 0.91 | 285 | 7.58 |
| 36 | 1.19 | 1.40 | 1.16 | 1.24 | 0.89 | 271 | 7.56 |
| 48 | 1.19 | 1.38 | 1.14 | 1.19 | 0.86 | 268 | 7.55 |
| 60 | 1.06 | 0.56 | 0.54 | 0.30 | 0.53 | 111 | 7.45 |

tered permanent magnets have been produced via this low-cost technique employing various milling times.

2. Experimental

A well-known commercial Nd-alloy with the composition close to that of ‘Neomax’ with 1.5 at.% of dysprosium and 1 at.% of niobium (Nd_{14.5}Dy_{1.5}Fe₇₆B₇Nb₁) was used to test the new system. The alloy was processed in the as-received condition (conventional cast ingot state). In order to produce the magnets via the HD process [5,6], the following procedure was adopted. Small pieces of the bulk ingot were placed in a stainless steel hydrogenation vessel, which was then evacuated to backing-pump pressure. Hydrogen was introduced to a pressure of 1 bar, which resulted in decrepitation of the bulk material. This material was then transferred (in air) to a ball milling pot which was filled with cyclohexane and roller milled for several hours (6–60 h). The milling pot was then coupled to the transfer system. Fig. 1 shows schematically the system which permitted backing-pump vacuum for cyclohexane removal and gas admission for powder transfer. The milled hydride powder was then vacuum dried for 1 h and transferred under N₂ to a small cylindrical rubber tube. Powder transfer was assisted with some tapping on the walls of the system and on the isostatic tube. This system was also provided with a 100 mesh sieve

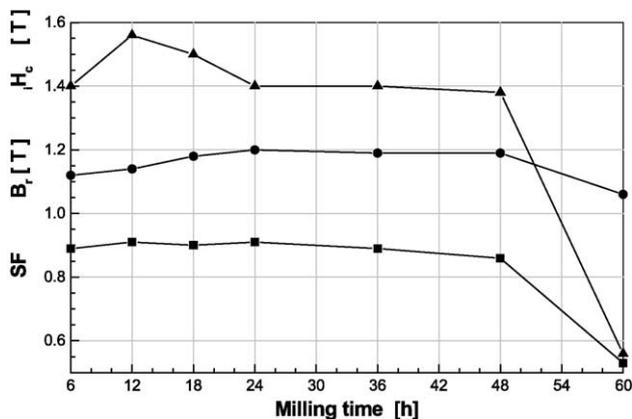


Fig. 2. Variation of B_r , H_c and SF with milling time for the sintered magnets.

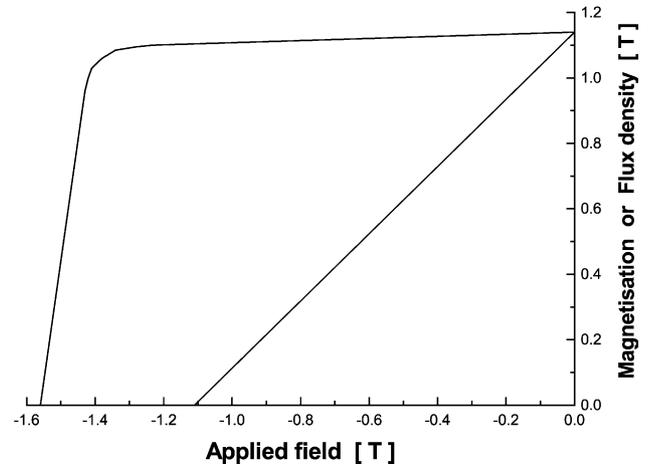


Fig. 3. Demagnetization curve of a magnet produced from the ingot milled for 12 h.

which retained the ball bearings and any coarse particles. The isostatic rubber tube was externally covered by a thin transparent flexible bag for an extra protection during and after transfer. The fine powder was aligned by pulsing three times to a 6 T magnetic field, pressed isostatically at a pressure of 200 MPa and then vacuum sintered for 1 h at 1050 °C, followed by cooling on the furnace. Magnetic characterization of the HD-sintered magnets was carried out using a permeameter. Measurements were performed after saturation in a pulsed field of 6 T. Magnet densities were measured using a liquid displacement system.

3. Results and discussion

The densities and magnetic properties of the Nd_{14.5}Dy_{1.5}Fe₇₆B₇Nb₁-sintered magnets prepared using the powder transfer technique are given in Table 1. In general, good densification has been achieved. HD permanent magnets produced using a milling time between 12 and 24 h showed good magnetic properties. Fig. 2 shows the variation of intrinsic coercivity, remanence and squareness factor with

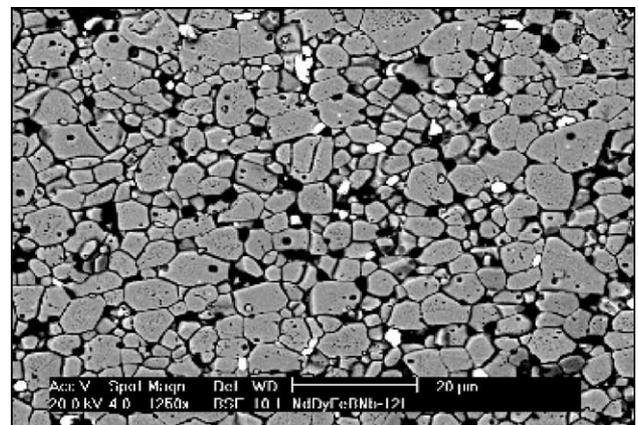


Fig. 4. Microstructure of a magnet produced from the ingot milled for 12 h.

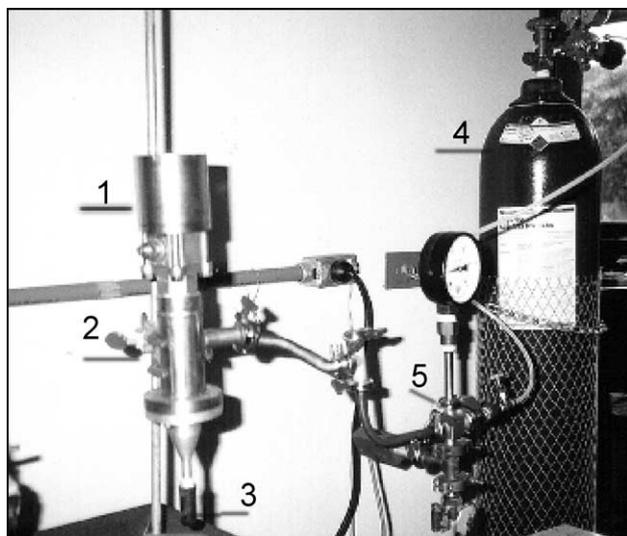


Fig. 5. Powder transfer facility. (1) Milling pot; (2) powder transfer device; (3) isostatic rubber tube; (4) N₂ gas cylinder; (5) vacuum system.

milling time for the HD-sintered permanent magnets. There is a maximum intrinsic coercivity of 1.56 T for the HD alloy milled for 12 h, a plateau between 24 and 48 h and a sharp decrease for longer milling times. This is also the behavior found in magnets prepared using the standard laboratory technique and can be explained as follows. The preparation of sintered magnets from rare-earth-based powders becomes difficult when highly reactive powders with very fine particle size are to be processed because some residual oxygen is always present even in a good glove box. A dramatic decrease in remanence and coercivity of sintered magnets has been reported when the particle size decreases below a critical value and this behavior was attributed to oxidation of the fine powder [7]. Furthermore, it has been reported that overmilling reduces the intrinsic coercivity by excessively damaging the particle surface [8]. It has also been shown that [9] for a particularly long milling time, grain growth is also a contributory factor to the decrease in the magnetic properties. Fig. 3 shows the demagnetization curves for the magnet that exhibited the best coercivity, and Fig. 4 shows the microstructure of this HD-sintered magnet which had a mean grain size of $5.0 \pm 0.2 \mu\text{m}$.

Unlike to the initial behavior of the coercivity, the remanence increases gradually with increase in milling time up to 24 h, remains stable up to 48 h and then also decreases, but not as dramatically as the coercivity. It has been shown for Pr–Fe–B-sintered magnets that the increase in remanence, with longer milling time of the HD starting material, is due to an increase in the degree of alignment (ϕ grains) of the sintered permanent magnets [10]. In this work, it has also been shown that high degrees of orientation can

be obtained with longer milling times, but better processing conditions or techniques need to be used to avoid degradation of the intrinsic coercivity. The squareness factor showed a similar behavior to the remanence. Magnetic properties of Nd_{14.5}Dy_{1.5}Fe₇₆B₇Nb₁ commercial permanent magnets [11] exhibited slightly superior intrinsic coercivity and inferior remanence ($\mu_0 H_c = 1.88 \text{ T}$, $B_r = 1.10 \text{ T}$) compared to those of the present hydrogen decrepitation-sintered magnets. Fig. 5 shows a picture of the assembled system employed in the present work. Although the present arrangement would be unsuitable for large-scale application, the technique proved to be very useful for studying magnets on a laboratory scale.

4. Conclusions

The powder transfer technique developed in the present work proved to be very satisfactory in producing sintered magnets on a laboratory scale. Hydrogen decrepitation magnets prepared using this new technique exhibited magnetic behavior similar to that seen in previously reported work that employed standard procedures. The magnetic properties of the permanent magnets produced via this low-cost system were equivalent to those of commercial magnets with identical composition.

Acknowledgements

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