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Pressure Effect on Magnetic Properties of Isotropic Nd-Fe-B Resin Bonded Magnets for Automotive Sector Applications

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Authors' contributions

This work was carried out in collaboration between all authors. Author SK under the guidance of author DN designed the study, performed the experiments, elaborated all graphics and wrote the first draft of the manuscript. Author MG helped in performing the SQUID measurements and reviewed the first draft of the manuscript. Author MP helped in performing the SQUID measurements. Author VP performed the XRD measurements and author DN assisted the study design, supervised the analyses and reviewed the manuscript. All authors read and approved the final manuscript.

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ABSTRACT

Aims: The purpose of this work is to study the effect of compression pressure on isotropic epoxy resin bonded Nd-Fe-B magnets, with the goal of achieving high densities at room temperature.

Methodology: We present data on the effect of the applied pressure on the structure and magnetic properties of a series of polymer bonded magnets based on MQ-powders with a distribution of grain sizes and coated with various thickness of epoxy resins.

Results: Densities of the order 83% of the theoretical estimated density value were achieved with an energy product of 10.33 MGOe. By increasing the molding pressure the density and the energy product of the bonded magnets was increased.

Conclusion: The combination of high density with good energy product is desirable for numerous applications especially within the automotive sector.

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1. INTRODUCTION

Nd-Fe-B based permanent magnets, one of the crucial materials supporting modern technologies are widely used in various fields including disc drives for information-storage devices, hybrid and electric vehicles, electric bicycles, and transducers [1-4]. These magnets are produced as sintered (SMs) and bonded magnets (BMs). Sintered magnets with a density close to the maximum theoretical value have high energy products $(BH)_{max}$ but the sintering process is complex and expensive [5]. Amongst the advantages of polymer bonded magnets (PBMs) are the possibility to form near net-shape magnets with accurate dimensional tolerances, low weight and corrosion resistance, high production rates, and excellent mechanical properties [6-8].

Thus, BMs based on the Nd–Fe–B alloys represent a rapidly growing sector of the permanent magnet industry [7]. The bonded magnet fabrication process involves mixing magnet powders with a binder such as thermoplastic polyolefin, polyphenylene sulphide (PPS), polyvinyl chloride (PVC), polypropylene (PP), polyethylene (PE), high density polyethylene (HDPE), polyamide, or thermosetting epoxy resin, followed by compression or injection molding [8-11]. These magnets provide a range of combinations of mechanical, physical, chemical, thermal and magnetic properties arising from different polymers used [10].

For isotropic bonded magnets the densities obtained with the compression molded technique are in the range of 75-78% of the theoretical full density, whereas for injection molding it can reach up to 65%. The dilution of the magnetic component within the polymer binder means the bonded magnets exhibit a lower remanence Mr, and $(BH)_{max}$ compared to fully dense sintered magnets [7]. Commercially available bonded Nd-Fe-B magnets typically have $(BH)_{max}$ values of 10–12 MGOe. The $(BH)_{max}$ of BM's depends on the magnetic properties of the magnet powders and the loading fraction in the binder which depends in turn on the molding process selected [12,13].

In our previous work we have demonstrated the fabrication of bonded magnets with densities in excess of 80% using methyl methacrylate (MMA) and benzoyl peroxide (BPO) and curing at temperatures below 100°C [14]. The objective of this work is to study the effect of compression pressure during the preparation of isotropic epoxy resin bonded Nd-Fe-B magnets, while aiming at achieve densities above 83% at room temperature. Our approach is a refinement of the compression molding process, in order to pack more magnetic powder and achieve higher densities, using a trimodal grain size distribution of the magnetic powders [15] and a resin with low curing temperature (25°C). We have used MQ-type powders, kindly provided by Molycorp, which can be molded up to 300°C.

2. MATERIALS AND METHODS

Epoxy resin (Epofix, Struers, Germany), triethylenetetramine (Struers, Germany), and zinc stearate (Acros Organics, Italy) were used as received. MQP and MQFP magnetic powders were provided by Molycorp.

A two-step method for producing the isotropic Nd-Fe-B bonded magnets is shown in (Fig. 1). First bisphenol-A-(epichlorhydrin) epoxy resin (EPO) including a triethylenetriamine (TETA) curing agent was dissolved in solution with the appropriate amounts of ethanol. Then the magnetic powder was added into the resin solution under rigorous stirring. The magnetic powder, composed of grain sizes with: 67% above 150 μm, 23% between 13 and 55 μm, and 10% below 10 μm. The powder fraction with large grains are added first and then those with smaller grain sizes. Lastly, zinc stearate (0.2%) was added as a lubricant. Three kinds of epoxy resins 2.5, 5, and 7.5 wt% that fully encapsulated Nd-Fe-B powders were produced. Subsequently these epoxy, magnetic powder and lubricant mixtures were subjected to a uniaxial compression up to 1.5 GPa. The curing of the compressed powders, with cylindrical shape, was carried out at room temperature for 12h. The different applied pressures are presented in (Table 1).

The densities were determined by measuring the mass and calculating the volume from the sample dimensions as well as with an electronic analytical balance (accuracy of 0.1 mg) using the Archimedes method. Based on the density values as well as on the magnetic properties of the produced bonded magnets, we present the measurements related to the samples with the optimum properties.

The compression was applied using an Instron press with pressures ranging from 0.25 to 1.5 GPa. The crystal structures were investigated using a Siemens D500 diffractometer in Bragg–Brentano geometry with Cu-Kα radiation, while the morphology of the samples was examined using a Phenom Pro SEM. The magnetic measurements were carried out at room temperature using a Quantum Design magnetic property measurement system (SQUID) with 5 Tesla maximum field.

3. RESULTS AND DISCUSSION

In (Table 1) the sample codes, production parameters, density, saturation magnetization, remanence and energy product of the produced bonded magnets are presented. The $(EPO)_{2.5/1.5}$ sample displays the highest density, 83%, as well as the highest energy product, 10.33 MGOe.

3.1 Structural Characterization of Plastic-Bonded Magnets

Fig. 2(a) and (b) shows the diffraction patterns of MQP powder and $(EPO)_{2.5/1.5}$ bonded magnet, respectively. The X-ray analysis shows that the $Nd_2Fe_{14}B$ phase is the main phase for both samples while minor (<2%) traces of Fe were observed in the $(EPO)_{2.5/1.5}$ sample.

The MQP-MQFP powder microstructure is presented in (Fig. 3(a-c)), and that of the epoxy bonded magnets in (Fig. 3(d-f)). MQP powder grains with sizes above 150 μm (Fig. 3a) have a plate-like, irregular polygon shape. For smaller grains, the shape becomes more spherical (Fig. 3(b,c)). The grain shape and size affect the packing density of the powder. Plate-like grains result in a higher packing density under the same compression force [10]. In figures 3(d), 3(e) and 3(f) the magnetic grains in the epoxy matrix for the $(EPO)_{2.5/0.5}$, $(EPO)_{2.5/0.75}$ and $(EPO)_{2.5/1.5}$ samples display cracks. These may be a result of the stress applied to the magnetic powder during compression.

3.2 Magnetic Behavior

In Fig. 4(a) and (b) the magnetization versus magnetic field data for the MQP powder and for the $(EPO)_{2.5/1.5}$ are presented. From (Fig. 4b) it is clear that there is no decrease of the coercivity. Densities of the order of 83% ($\rho_{EPO2.5/1.5}$ = 6.32 g/cm^3) of the theoretical estimated density value (ρ_{th} = 7.62 g/cm^3) were obtained. The $(BH)_{max}$ was estimated for the $(EPO)_{2.5/1.5}$ at ~10.33 MGOe while that of the MQP powder was ~14.2 MGOe (Molycorp MAGNEQUENCH, File: 00642438.DAT/lot: B43729 14-12-150M). This reduction in the $(BH)_{max}$ value can be attributed to the dilution of magnetic powder in the epoxy matrix as well as a slight degradation in the squareness of the demagnetization curve.

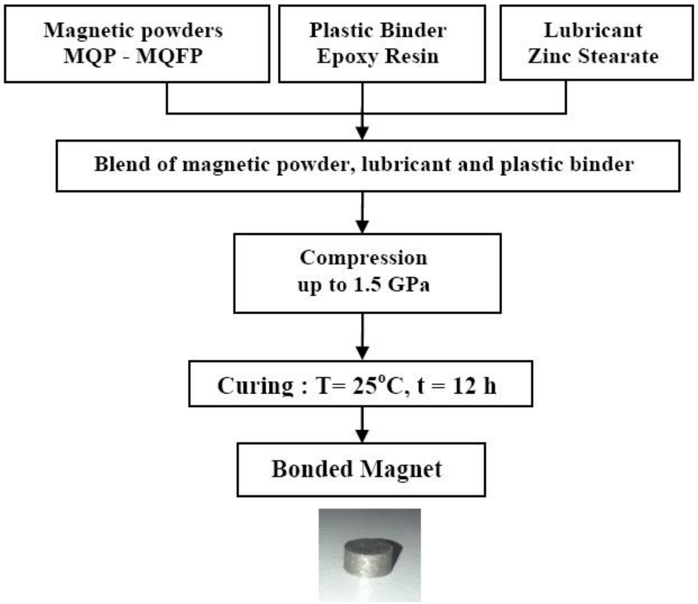


Fig. 1. Experimental approach used for making resin bonded magnets

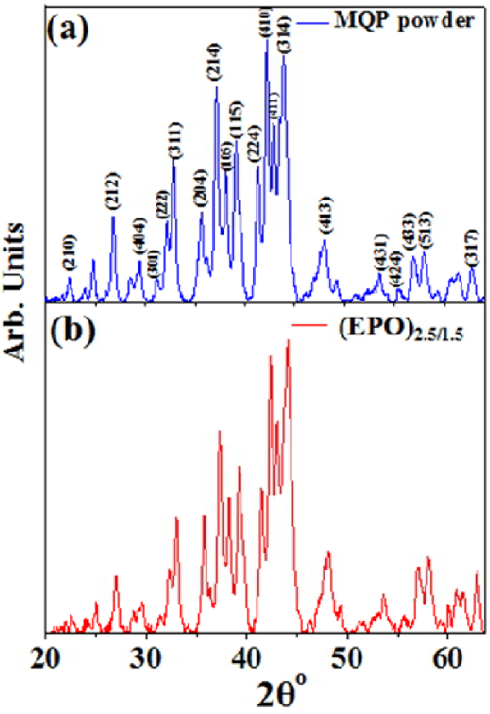


Fig. 2. X-ray Diffraction pattern of (a) MQP magnetic powder and (b) $(EPO)_{2.5/1.5}$ bonded magnet

The dependence of the density and the remanence Mr for the 2.5 wt % epoxy resin bonded magnet on the molding pressure is shown in (Fig. 5) with the correlation between the molding pressure and the energy product $(BH)_{max}$ illustrated in (Fig. 6).

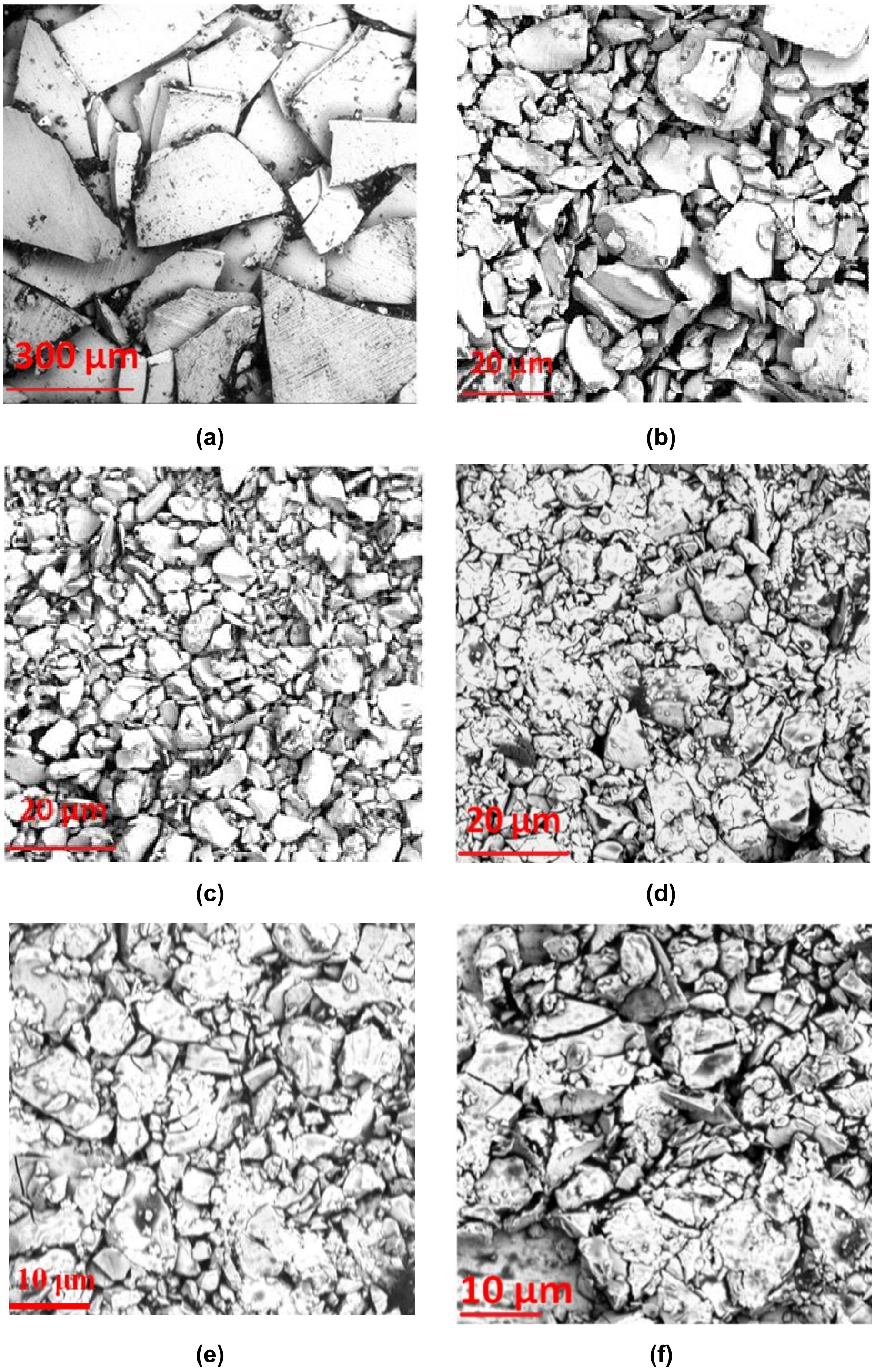
The relationship between molding pressure and density in the produced bonded magnets was evaluated. At 0.5 GPa of molding pressure, the $(EPO)_{2.5/0.5}$ had the lowest density 81.5%. In contrast, at the highest applied molding pressure 1.5 GPa, the $(EPO)_{2.5/1.5}$ showed the highest value of density 83%. Generally, increased applied pressure gives increased density. Furthermore, the remanence Mr remains almost constant by increasing the applied pressure. Therefore it is possible to obtain high remanence bonded magnets with low molding pressures at room temperature. Concerning the relationship of the applied pressure with the energy product, the $(EPO)_{2.5/0.5}$ had the lowest $(BH)_{max}$ 9.74 MGOe, whereas the $(EPO)_{2.5/1.5}$ present the higher value 10.33 MGOe. Generally, by increasing the applied pressure the $(BH)_{max}$ is increased (Fig. 6).

Beyond the concentration of 2.5% wt., two different concentrations of epoxy resin 5 and 7.5 % wt. were used. According to the density values, the bonded magnets with 5% wt epoxy resin, present densities in the range of 82% for

Table 1. Bonded magnet sample codes, production parameters, density, saturation magnetization, remanence and energy product values

a/a	Sample code $E_{x/y}$*	Production parameters			Density (%)	Ms (Emu/g)	M_r (Emu/g)	$(BH)_{max}$ (MGOe)
		System pressure (GPa)	T (°C)	T (h)				
1.	$EPO_{2.5/0.50}$	0.50	25	12	81.5	138.3	83.9	9.74
2.	$EPO_{2.5/0.60}$	0.60	25	12	81.7	139.6	84.0	9.98
3.	$EPO_{2.5/0.75}$	0.75	25	12	82.0	139.6	83.9	9.99
4.	$EPO_{2.5/1.50}$	1.50	25	12	83.0	140.2	83.8	10.33

** the code of the materials contain the corresponding symbols :*
E : Epoxy resin BMs, x: defined epoxy resin concentration (%wt.), y: defined applied pressure



(a) **(b)**

(c) **(d)**

(e) **(f)**

Fig. 3. SEM images of (a) MQP powder with grain size >150 µm, (b) MQFP powder with grain size range of 13 - 55 µm, (c) MQFP powder with grain size < 10 µm, (d) $(EPO)_{2.5/0.5}$, (e) $(EPO)_{2.5/0.75}$ and (f) $(EPO)_{2.5/1.5}$ bonded magnets

low applied pressures (0.5 and 0.6 GPa), whereas for higher applied pressures the densities are decreased. Indicatively, the remanence of the bonded magnet produced at

0.5 GPa was 82.8 Emu/g, whereas the $(BH)_{max}$ was 10 MGOe. Moreover, for 7.5% wt. epoxy resin, the values of density ranges between 64 and 68% at applied pressures 0.25 GPa and 1.5 GPa, respectively. These low values might be attributed to the large amount of epoxy resin that encapsulate the magnetic grains and thus obstruct the adhesion of the magnetic powder. The corresponding values of remanence and $(BH)_{max}$ for the bonded magnet produced at 1.5 GPa were 80.62 Emu/g and 6.21 MGOe, respectively.

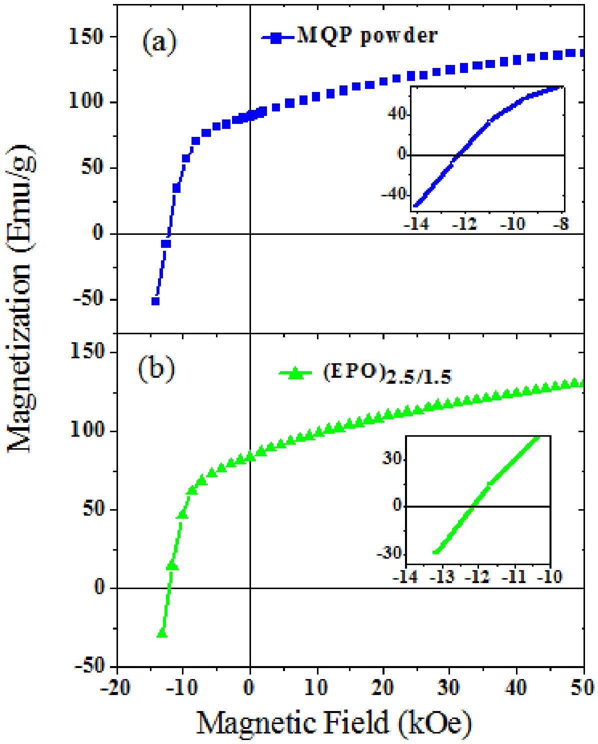


Fig. 4. (a) Magnetization data for the original MQP powder and (b) data obtained with our process for $(EPO)_{2.5/1.5}$. The insets show an enlargement of the loops

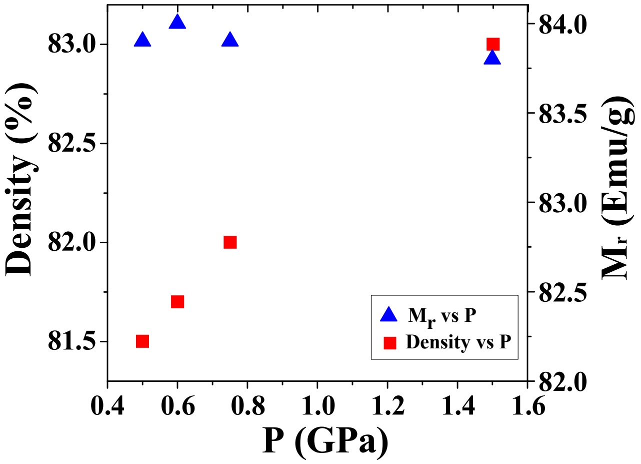


Fig. 5. Density (%) versus molding pressure and remanence M_r versus molding pressure of the 2.5 wt % epoxy resin bonded magnet

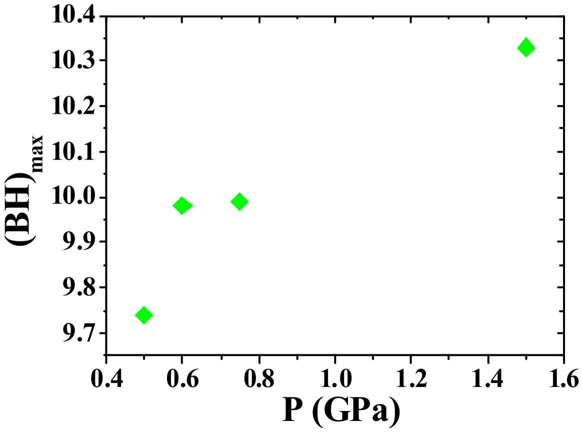


Fig. 6. Energy product $(BH)_{max}$ versus molding pressure of the 2.5 wt % epoxy resin bonded magnet

4. CONCLUSION

We have reported the fabrication of epoxy resin bonded magnets with $(BH)_{max}$ values comparable with the state of the art. Using commercial MQP powders we have achieved densities in excess of 80% using epofix resin and curing at room temperature. This approach is considered as very promising for the production of PBMs with energy products greater than those commercially available. The combination of high density with good energy product is very promising for many applications especially within the automotive sector.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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