J. Min. Metall. Sect. B-Metall. 46 (1) B (2010) 25 - 32

Journal of Mining and Metallurgy

MECHANICAL AND MAGNETIC PROPERTIES OF COMPOSITE MATERIALS WITH POLYMER MATRIX

A. Grujić[*], N. Talijan[#*], D. Stojanović[**], J. Stajić-Trošić[*], Z. Burzić[***], Lj. Balanović[****] and R. Aleksić[**]

[*]Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Serbia,
[**] Faculty of Technology and Metallurgy, University of Belgrade, Serbia
[***] Military Technical Institute, Belgrade, Serbia
[****]Technical Faculty in Bor, University of Belgrade, Serbia

(Received 25 February 2010; accepted 15 April 2010)

Abstract

Many of modern technologies require materials with unusual combinations of properties that cannot be met by the conventional metal alloys, ceramics, and polymeric materials. Material property combinations and ranges have been extended by the development of composite materials. Development of Nd-Fe-B/polymer composite magnetic materials has significantly increased interest in research and development of bonded magnets, since particles of Nd-Fe-B alloys are proved to be very suitable for their production. This study investigates the mechanical and magnetic properties of compression molded Nd-Fe-B magnets with different content of magnetic powder in epoxy matrix. Mechanical properties were investigated at ambient temperature according to ASTM standard D 3039-00. The obtained results show that tensile strength and elongation decrease with an increase of Nd-Fe-B particles content in epoxy matrix. The modulus of elasticity increases, which means that in exploitation material with higher magnetic powder content, subjected to the same level of stress, undergoes 2 to 3.5 times smaller deformation. Scanning Electron Microscopy (SEM) was used to examine the morphology of sample surfaces and fracture surfaces caused by the tensile strength tests. The results of SQUID magnetic measurements show an increase of magnetic properties of the investigated composites with increasing content of Nd-Fe-B particles.

Keywords: *Composite materials; Permanent magnets; Nd-Fe-B; Epoxy resin; Mechanical properties; Magnetic properties.*

[#] *Corresponding author: ntalijan@tmf.bg.ac.rs*

DOI:10.2298/JMMB1001025G

1. Introduction

A composite is considered to be any multiphase material that exhibits a significant portion of properties of both constituent phases such that a better combination of properties is realized.

The dispersed phase for particle-reinforced composites can have quite a variety of geometries, but for an effective reinforcement particles should be equiaxed i.e. particle dimensions are approximately the same in all directions, should be small enough and evenly distributed throughout the matrix.

Composite materials with reinforcing particles based on permanent magnetic Nd-Fe-B materials are of current research interest [1,2], due to numerous possible applications, such as hard disc components, electric appliances and automobile industry [3,4].

Bonded magnetic materials based on Nd-Fe-B magnetic powders can be manufactured in several ways, and one of the most innovative is the process of compression molding.

Compression bonding is a die-pressing process that involves mixing of magnetic powders with an epoxy binder and processing that mixture into a finished product [3,5]. As a method of production, compression bonding provides magnets that exhibit high energy products, e.g. more than 80 kJ/m^3. However, they are limited to more basic shapes due to the nature of the die-pressing process.

The Nd-Fe-B/epoxy composite materials offer an excellent combination of superior magnetic and mechanical properties. The most important advantages of polymer bonded magnets are low weight, near-net shape manufacture, good mechanical strength and corrosion resistance [6,7] as well as isotropic magnetization. Oxidation and corrosion of polymer-bonded Nd–Fe–B magnets during their service life is a significant problem because it limits their possible applications in automobiles, computers, and medical devices designed for use in hostile environments [8].

The nanocrystalline magnetic materials based on rapidly solidified Nd–Fe–B alloys have been recently researched despite the poor B-H loop squareness and poor H_{ci} (400 kA/m) due to the fact that they contain up to three times smaller amount of expensive rare earth element (Nd) comparing to conventional rapid-quenched and sintered Nd–Fe–B magnets [9].

High values of remanence are achieved when the iron content is increased and $Nd_2Fe_{14}B/\alpha$-Fe and/or $Nd_2Fe_{14}B/Fe_3B$ type nanocomposite microstructure are developed [1,10]. Nanocrystalline permanent magnets have been extensively studied because of their potential for production of bonded magnets [11,12,13].

The scope of this paper was to demonstrate the influence of various amounts of magnetic Nd-Fe-B filler on mechanical and magnetic properties of final magnetic composite materials with epoxy matrix. The amount of the magnetic filler is directly responsible for magnetic behavior of obtained bonded magnets.

In other hand, the fraction of epoxy polymer plays the most important role in mechanical properties of the final Nd-Fe-B/epoxy composite materials.

2. Experimental

The rapid quenched Nd-Fe-B powder with low neodymium content was used to produce polymer bonded magnetic composites. The chemical composition of the starting Nd-Fe-B alloy was Nd-12 mass%, Pr-0.2 mass%, B-4.2 mass%, Al-0.3 mass%, Fe-balance. The initial magnetic properties measured on a vibrating sample magnetometer after optimal annealing are: Br = 1.09 T, H_{cb} = 215 kA/m, H_{cj} = 223 kA/m and $(BH)_{max}$ = 85 kJ/m^3. Used magnetic powder was fairly characterized in previous investigations [14,15,16].

As a polymer matrix for magnetic composite materials preparation epoxy resin was used. The applied thermosetting epoxy system is combination of liquid mixture of Bisphenol A and Bisphenol F resins and cross linking agent (hardener) which cures fully at room temperature. The cured epoxy resin has tensile strength ~ 58 MPa, elongation ~ 2.8%, compression strength ~ 96 MPa, flexural strength ~ 78 MPa and density ~ 1.2 g/cm^3.

In order to understand the effect of a different content of the Nd-Fe-B magnetic powder – filler on magnetic and mechanical properties of the Nd-Fe-B/epoxy bonded magnetic materials samples with varied filler content, from 10 to 95 wt% were prepared.

The investigated composite materials were compression molded under a pressure of 4MPa at room temperature using a lab scale compression molding press. The molded samples were allowed to cure under the molding pressure about one day.

Observations of morphology of sample surfaces and fracture surfaces of the synthesized composite materials were made on the JEOL JSM-5800 Scanning Electron Microscope, with the accelerating voltage of 20 kV.

The Nd-Fe-B/epoxy composites and pure epoxy polymer samples were machined into tensile specimens, which were then tested at ambient temperature. The tensile specimen was dumbbell shaped as required by ASTM D 3039-00 [17], with a cross-section of 40 x 5 mm in the gauge length. At least five specimens were tested in tests for each composite and pure epoxy specimen. The dumbbell-shaped tensile specimens of pure epoxy resin were fabricated by casting the resin into the rubber moulds and following the recommended curing cycle from the material manufacturer. A universal material testing machine (Schenck TREBEL RM 100) was used for mechanical tests. The moduli of elasticity were derived from the linear portion of the stress–strain curves obtained by tensile tests.

Magnetic properties of the samples were measured at ambient temperature on the Superconducting Quantum Interference Device (SQUID) magnetometer with magnetic field strength in range −5 to 5 T.

3. Results and discussion

Since the nanocomposite structure of a nanocrystalline Nd-Fe-B alloy with reduced Nd content has a very important role for bonded magnetic materials production, characterization of an optimally annealed rapid quenched $Nd_{4.5}Fe_{77}B_{18.5}$ alloy was done in order to gain insight into micro- and nanostructure, as well as phase composition and magnetic characteristics. The SEM microphotograph of the used Nd-Fe-B alloy presented in Fig 1.a shows angular plate-like particles with mean size of approximately 86 μm. The SQUID hysteresis loop (Fig 1.b) is typical for high remanence Nd-Fe-B alloys [6,18]. Remanence ratio is higher than the theoretical limit proposed by Stoner-Wolfhart theory [19], which suggests the $Nd_2Fe_{14}B/Fe_3B$ and/or α-Fe nanocomposite structure of the magnetic alloy. The bright field TEM micrograph presented on (Fig. 1c) showing the microstructure of the analyzed alloy in the optimal magnetic state illustrates the average crystal grain size below 30 nm and confirms suggested nanocrystalline structure. Further HREM analysis (Fig. 1d) has confirmed that in the microstructure of the $Nd_{4.5}Fe_{77}B_{18.5}$ alloy there are crystal grains with the sizes about 10 nm and less.

Good adhesion between polymer matrix and Nd–Fe–B powder is essential for the composites, especially at temperatures above the glass transition temperature of the polymer. In addition, for permanent bonded magnets, particle size of the magnetic powder plays an important role for determination of powder to binder ratio, degree of particle alignment and magnetic and mechanical properties [20]. Generally speaking, under the optimal compression

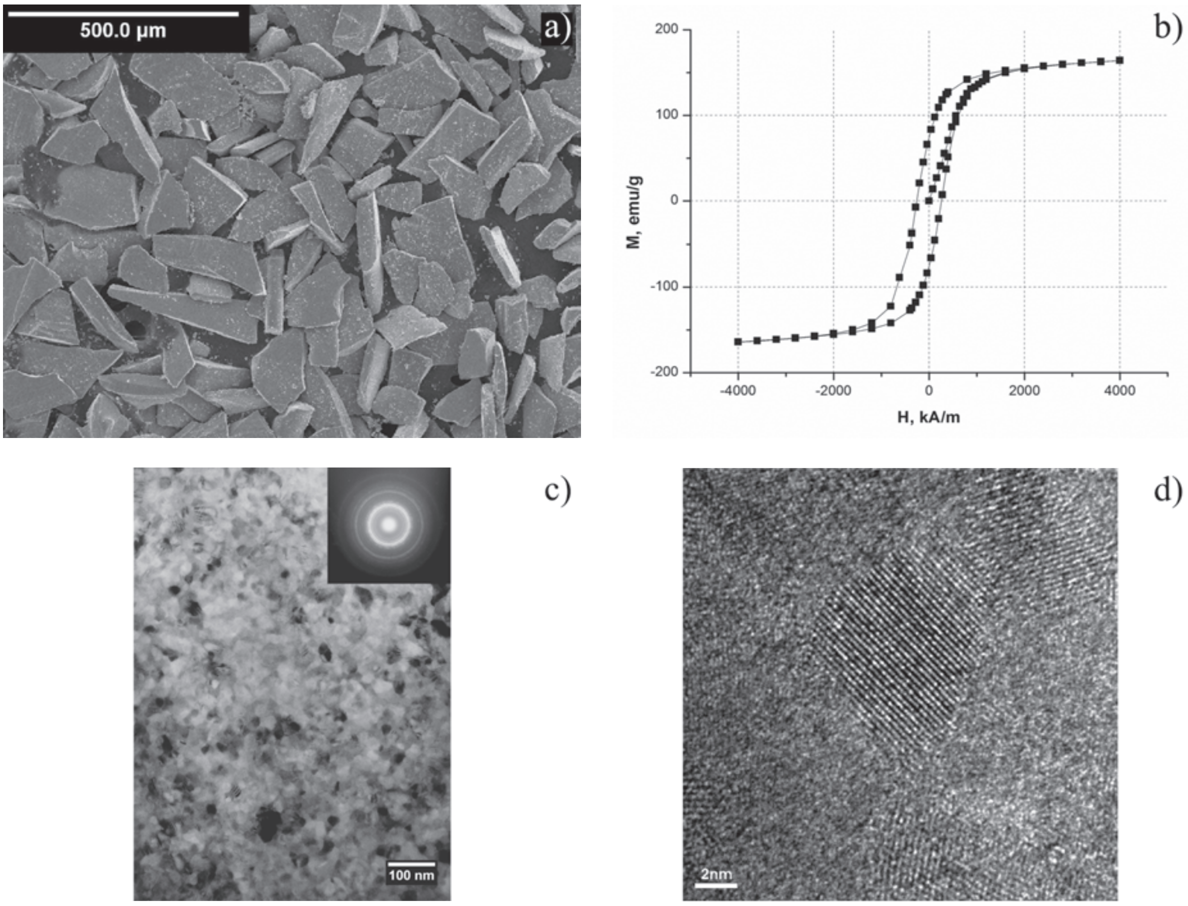

Fig 1. a) SEM micrograph, b) SQUID hysteresis loop, c) TEM micrograph and d)HREM nanograph of the used Nd-Fe-B alloy

conditions the plate–like particles would result in higher packing density [2,21].

SEM micrographs of the sample surface and the fracture surface morphology of the Nd-Fe-B/epoxy composite with different amount of the magnetic filler are presented in Fig.2. The Nd–Fe–B particles are shown as light grey and the epoxy matrix as dark. The darkest gray parts represent the holes ensued by pulling out of the Nd-Fe-B particles during the mechanical breaking (tension). Although the Nd–Fe–B particles are of variable size and shape, they seem to be attached rather well to the matrix.

In order to determine the in-plane tensile properties of the polymer matrix composite materials reinforced by the Nd-Fe-B magnetic particles, standard test tensile method was used [13]. The ultimate tensile stress σ_m of the investigated magnetic composite materials was calculated using Eq. 1:

$$\sigma_m = \frac{F_{\max}}{b \cdot d} \qquad .....(1)$$

Where: σ_m – ultimate tensile stress, MPa; F_{max} – maximal load before failure, N; b – sample width, mm; and d – sample thickness, mm.

The values of ultimate tensile stress,

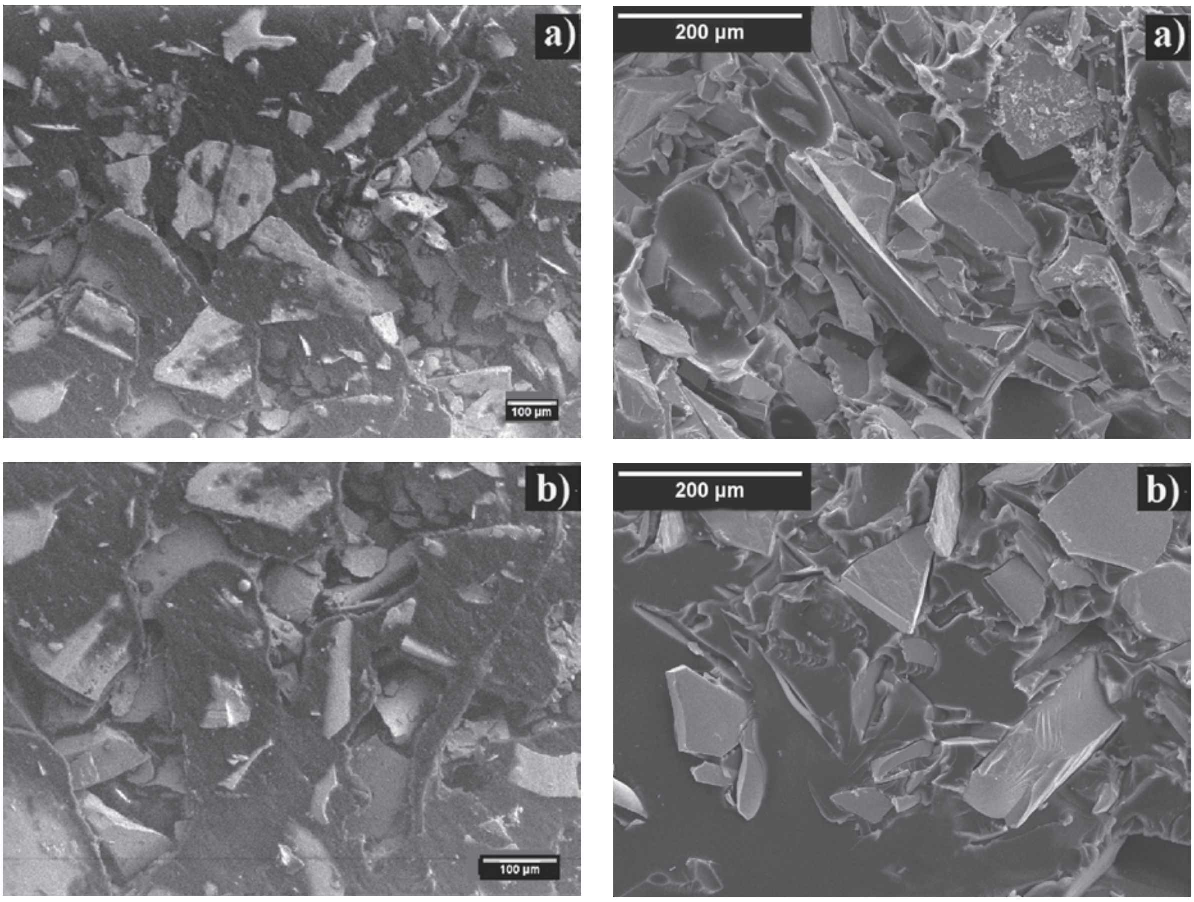

Fig 2. SEM micrographs of the sample and fracture surface of the Nd-Fe-B/epoxy composite material with:a) 95wt% and b) 50 wt% of Nd-Fe-B filler

elongation and modulus of elasticity have been taken from obtained stress-strain diagrams for all investigated composites and presented in function of the Nd-Fe-B content in the epoxy matrix respectively (Fig. 3, 4 and 5).

The elastic modulus E of the investigated magnetic composite materials was calculated using Eq. 2 [17]:

$$E = \frac{\Delta\sigma}{\Delta\varepsilon} = \frac{\Delta F}{\Delta\varepsilon} \cdot \frac{1}{b \cdot d} \quad .....(2)$$

Where the ratio $\Delta\sigma/\Delta\varepsilon$ is determined by linear regression method from linear portion of stress-strain curves i.e. in the Hookean region.

With decreasing amount of the thermosetting epoxy polymer in the composites from 95 to 10 wt%, i.e. with increasing content of the Nd-Fe-B filler, the values of tensile stress and elongation are decreasing (Fig. 3 and 4), while the composite materials become more brittle and as a consequence the ductility of composites is lower.

Modulus of elasticity is increasing with higher amount of magnetic filler which is crucial in analysis of possible use of the investigated magnetic composite materials as functional materials (Fig. 5.).

This means that the material with higher amount of Nd-Fe-B filler, subjected to the equal level of stress (ballast), tolerates 2 to 3.5 times lower deformation. The modulus of elasticity is very important parameter for analysis of the composite magnetic materials behavior in conditions when the discontinuous load is applied. There are no significant changes of the elastic modulus for the composites with Nd-Fe-B filler content

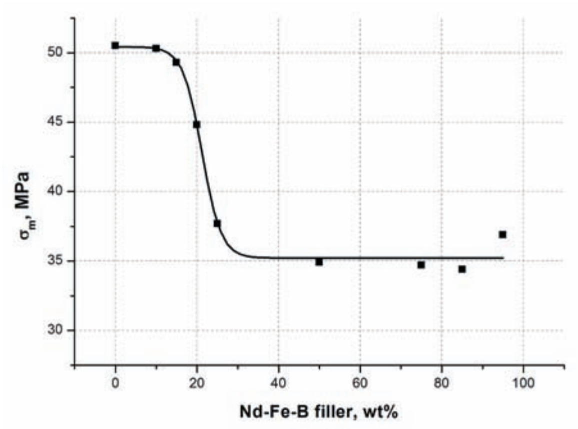

Fig. 3 – Ultimate tensile stress in function of Nd-Fe-B content for investigated composites

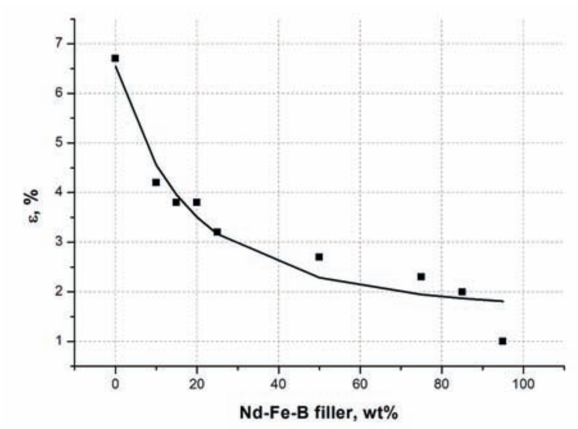

Fig. 4 – Ultimate tensile strain in function of Nd-Fe-B content for investigated composites

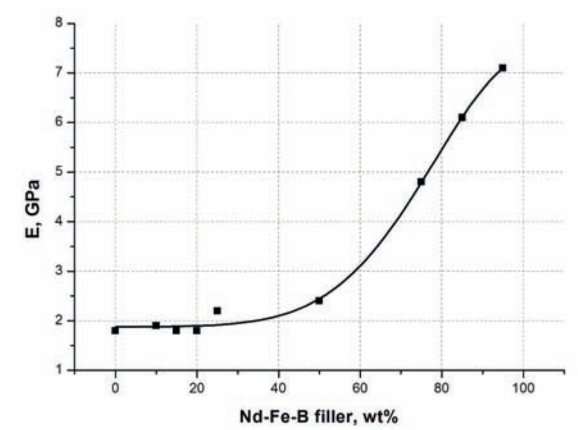

Fig. 5 – Modulus of elasticity in function of Nd-Fe-B content for investigated composites

up to 20 wt% in epoxy matrix which could be explained as low sensitivity of method. For instance, dynamic-mechanical analysis (DMA), one of the most sensitive technique, could be applied for investigation in mentioned narrow region (from 0 to 3.8 vol. % Nd-Fe-B) and explore additional information about temperature transitions in polymer composites [22,23]. The values of elastic modulus upswing with increasing amount of Nd-Fe-B powder from 20 to 95 wt% achieve 7.1 GPa. This value of elastic modulus is 3 times higher than the composites with up to 20 wt% of magnetic filler in epoxy matrix.

The SQUID magnetic measurements of the investigated composite materials reveal a typical shape of hysteresis loops for Nd-Fe-B materials with reduced Nd content [24] and indicate various values of magnetic parameters. The quantitative values of coercivity (H_{cb}), remanence (B_r) and maximal energy product $(BH)_{max}$ were taken from obtained B-H SQUID hysteresis loops and presented in Table 1. This type of rare-earth materials is often called high remanence magnetic materials because of their high values of remanence, which has a direct influence on high values of maximal energy product.

Table 1. – The results of SQUID magnetic measurements for investigated composites with 15, 50, 75 and 95 wt % of Nd-Fe-B filler in epoxy matrix

Nd-Fe-B	H_{cb}	B_r	$(BH)_{max}$
[wt %]	[kA/m]	[T]	[kJ/m^3]
95	175.1	0.56	27.1
75	135.3	0.31	10.3
50	103.5	0.19	5.6
15	71.6	0.13	2.4

The presented results show the substantial difference between the four obtained composite materials with different content of the Nd-Fe-B particles in epoxy matrix. It is obvious that composite materials with higher amount of magnetic powder have improved magnetic properties.

4. Conclusions

Composite magnetic materials (bonded magnets) with varied content of Nd-Fe-B particles in epoxy matrix from 10 to 95 wt% were produced and studied.

The Nd-Fe-B powder as functional magnetic component is responsible for magnetic properties of final composite materials, while quantity of epoxy matrix is more important parameter for mechanical properties.

Uniform particle distribution and good adhesion between polymer matrix and magnetic filler were obtained, which are crucial for superior mechanical properties.

Experimental results of the tensile tests show considerable improvement in the modulus of elasticity caused by the higher amount of the Nd-Fe-B magnetic filler.

Also, with increasing amount of Nd-Fe-B particles in epoxy matrix the values of tensile stress are decreasing.

As expected, magnetic properties are significantly enhanced with higher amount of Nd-Fe-B magnetic powder.

In addition, this study provides a method for tailoring of properties of epoxy bonded magnetic composites in general, especially for automobile and information technology applications where relatively brittle metallic permanent magnets are not useable.

Acknowledgement

This work has been supported by the Ministry of Science and Technological Development of the Republic of Serbia under Project: OI 142035 B.

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