# Structure and Properties of Polyethylene-Based Magnetic Composites

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Abstract- Magnetic polymer composite materials have very good rheological (that are characteristic for polymers used as matrix) and magnetic properties superior to those of polymers (due to their magnetic filler) making them useful in many power applications (electromagnetic shielding, permanent magnets etc.). In this paper the results of an experimental study regarding the manufacture and characterization of some composites which have low density polyethylene (LDPE) as matrix and neodymium (Nd) and neodymium-iron-boron (Nd-Fe-B) as filler are presented. The manufacture process (with mass content between 0 and 15 %) and their structure (obtained by electronic and optical microscopy) are presented. It is shown that the samples are inhomogeneous and isotropic and the filler particles form clusters of variable dimensions and distances between them. The rheological behavior of composites was analyzed and more rapid melting was observed for composites compared to neat polyethylene

Then, the first magnetization cycles obtained on neodymium (A) and neodymium-iron-boron (B) samples for three mass concentrations (5, 10 and 15 %) are presented. It is shown for both samples type (A and B) that the hysteresis area cycles and the hysteresis losses increase with filler content being greater for samples B than A. It is shown, also, that the magnetic permeability values increase with filler content, but decrease very quickly with the magnetic field strength.

## I. INTRODUCTION

Polymer composites with conductive or magnetic filler have electrical, magnetic and thermal properties superior to those of polymers, easy process ability and low cost [1]. For their manufacture polyethylene, polyvinylchloride, polycarbonate, resins, epoxy acrilonitril-butadien-stiren (ABS), polystyrene (PS), nylon 6,6 etc. as matrix and aluminum [2] carbon and graphite [3,4], copper [5], steel [6], aluminum nitrides [7], nickel or silver particles [9], polyacrylonitrile (PAN) [8], [9], barium titanate [10] as fillers are used.

The polymer composites with conductive/magnetic fillers are used for encapsulating, thin film coating, packing of electronic circuits [4], electromagnetic and radio- frequency interference (EMI/RFI) shielding for electronic devices and electrostatic dissipation (ESD) [9], [11].

The properties of polymer composites depend, on one hand, on the matrix and fillers characteristics and, on the other hand, on their technological process and environment action [12-14]. In a series of papers the influence of

nature, concentrations and filler dimensions on the properties of composite materials with conductive [15-18] and magnetic [14], [19-20] fillers are presented. It is shown that there is a critical concentration for which the percolation phenomenon occurs, leading to important variations of the electrical and magnetic properties allowing more diverse applications [14], [21-23].

Due to the many applications of polymer composite materials with magnetic filler (hard disc components, electric appliances, automobile industry, sensing elements, electronic, small motors in video recorders, camcorders, printers, communication and micro-electromechanical system (MEMS) applications, actuators, magnetic buffers etc.), in the last decades multidisciplinary research were performed in order to obtain bonded magnetic materials (based on Nd-Fe-B magnetic powders and epoxy resins), rubber magnets (using the same magnetic powders, but with thermoplastic polymers). These researches are directed into four directions: (a) increase of magnetic energy density, (b) improving corrosion resistance, (c) optimization production process of process parameters and (d) reduction of the subtle rare earth content (Nd), targeting decreasing the price of the magnetic material [24].

Neodymium-iron-boron (Nd-Fe-B) composite materials are an excellent combination of superior magnetic and mechanical properties. The most important advantages of polymer bonded magnets are their simple technology, possibility of forming their final properties, lowering manufacturing costs, low weight, near-net shape manufacture, good mechanical strength and isotropic magnetization.

On the other hand, the oxidation and corrosion of polymer-bonded Nd–Fe–B magnets limit their possible applications in automobiles, computers, and medical devices designed for use in hostile environments [24].

The amount of the Nd-Fe-B powder in the bonded magnet is directly responsible for magnetic and mechanic [25-26] behaviour of bonded magnets. A higher content of Nd-Fe-B powder usually results in a higher remanent magnetization  $(M_r)$  and maximum energy density  $(BH)_{max}$  and therefore, it is desirable from the magnetic perspectives.

On the other hand, a higher content of magnetic filler may change the rheology of polymer melt during the process, in the detriment of the mechanical strength of bonded magnets. The rheological behaviour of polymer composites is very important for the melt processing step final characteristics and depends on the material microstructure, the state of filler dispersion, the aspect ratio of the filler, the interaction between the filler and the polymer chains and filler-filler interactions [6, 27-32].

The use of coupling agents is highly recognized as benefic in terms of enhanced wettability of the filler surface by non-polar polymers and enhanced interfacial adhesion resulting in better mechanical properties [33-34]. Copolymers containing maleic anhydride, such as maleic anhydride-grafted polyethylene, are the most commonly reported coupling agents: the anhydride groups of the copolymers can react with the surface of the filler and the other end of the copolymer entangles with the polymer matrix because of their similar polarities [35-36].

Different metallic particles (iron, aluminum, high alloy steel, casting cooper alloy with tin etc.) were also used besides Nd-Fe-B in bonded magnets. These materials have improved mechanical properties but lower magnetic properties than composites which contain only powder of Nd-Fe-B [37]. Consequently, a balance between magnetic properties and corresponding dynamic mechanical behavior is an important issue for bonded magnet applications [24].

In many papers the magnetic and mechanical characteristics of composites with thermoplastic polymers matrix and different magnetic filler (rubber magnets) are presented. Low and high density polyethylene [38-41], copolymers with polystyrene end blocks and a rubbery poly(ethylene-butylene) mid block [42], natural rubber [43-44], silicone polymers [45-46], polyethylene glycol and styrene butadiene rubber [47], pentaerythritol tetrapolyethylene glycol ether with four thiol-modified terminals [48] etc. were used as matrix.

As fillers, Nd-Fe-B [38-39, 43, 47], ferrites [40-41, 43, 49], nickel [41] and nickel alloys [40, 42, 49], iron [42], magnetite [44, 48] and Fe-Si-B [45], Ni fibres [41, 46] etc. were used.

It has been found that the remanent magnetization of polymer composites with magnetic fillers increases with filler degree at first, but it decreases at high Nd-Fe-B levels because of interactions among magnet particles [39].

The relative magnetic permeability  $\mu_r$  increases with the filler content  $c_{\nu}$ , an estimation of this increase might be obtained with the linear equation:

$$\mu_r(c_v) = 1 + Cc_v,\tag{1}$$

where  $c_v$  is the volume concentration of the filler and *C* is a coefficient which depends on the magnetic properties, the shape and the volume fraction of the filler [40-41].

For spherical particles it may be considered C = 3 [41], equation (1) becoming

$$\mu_r(c_v) = 1 + 3c_v.$$
(2)

If the filler content has greater values ( $c_v > 0.25$ ), the variations of the magnetic permeability significantly deviates from the linear ones, a parabolic variation being presented in some papers [42]:

$$\mu_r(c_v) = 1 + C'c_v^2, \tag{3}$$

where C' is a material constant.

As is presented in [41], a hybrid magnetic polymer composite is defined as a composite containing two or more fillers with different magnetic properties, sizes, size distributions and shapes. The relative magnetic permeability of a hybrid magnetic polymer composite consisting of similar particles (shape, size, size distributions) but having different magnetic properties is given by the equation:

$$\mu_r(c_{v1}, c_{v2}) = 1 + C'c_{v1}^2 + C''c_{v2}^2, \qquad (4)$$

where  $c_{v_1}, c_{v_2}$  are the the volume concentrations of the filler 1 and 2, respectively [41].

On the other hand, the mechanical properties (the tensile strength etc.) of the Nd-Fe-B composites are lower comparative to the unfilled ones [38, 43, 46]. An improvement of their properties is obtained if nanometric fillers are used [44-46, 48].

An experimental study of electrical conductivity – measured both in DC and AC – of some polyethylene polymer composites with neodymium particles as filler is presented in [14]. A theoretical and experimental study of dielectric response function and electrical permittivity of these materials is presented in [23] and a study concerning the dielectric losses on the same composites type is presented in [50].

In this paper the results of an experimental study regarding the manufacture and characterization of some composites of low density polyethylene (LDPE) as matrix and Neodymium and Neodymium-Iron-Boron as fillers are presented. The structural characteristics (by electronic and optical microscopy) but also the magnetic ones are analyzed and conclusions regarding the influence of the nature and fillers content on the magnetic properties are drawn.

#### **II. EXPERIMENTS**

Experiments were performed on flat samples of composites prepared from low density polyethylene (LDPE) with a melt flow index (190 °C, 2.16 kg) of 0.3 g/10 min, a density of 0.920 g/cm<sup>3</sup> at 23 °C and an electrical conductivity of  $5 \cdot 10^{-17}$  S/m.

As fillers, neodymium particles were used (for samples A) and Nd-Fe-B (for samples B). The particles of Neodymium have the length of 75-100  $\mu$ m and the width of 30-50  $\mu$ m (Fig. 1), the density of 7 kg/dm<sup>3</sup> and the electrical conductivity of 1.56  $\cdot$ 10<sup>6</sup> S/m.

The particles of Neodymium-Iron-Boron have the length of 60- 570  $\mu$ m and the width of 30-300  $\mu$ m (Fig. 2) and the density of 7.5 kg/dm<sup>3</sup>. Maleic anhydride-grafted polyethylene (MA-PE), with a density of 0.925 g/cm<sup>3</sup> and a melting point of 105 °C, was used as compatibility agent.

A 50 cm<sup>3</sup> mixing chamber of a Brabender LabStation was used for mixing and homogenizing Neodymium and Nd-Fe-B powders with the polymer matrix and the compatibility agent (5 wt % MA-PE).

Metal powders (concentrations of 5, 10 and 15 wt %) were slowly added (~ 2 minutes) to the mixture of LDPE and MA-PE and mixed at 160  $^{0}$ C, for 8 min (the speed of the rotors being 100 rpm) [14].



Fig.1. Neodymium particles (Optical microscopy, 200 X).



Fig.2. NdFeB particles (Optical microscopy, 100 X).

 TABLE I.

 FILLER CONCENTRATIONS FOR SAMPLES A AND B

Sample	Filler mass	Filler volumic			
	concentration $c_m$ (%)	concentration $c_v$ (%)			
A0, B0	0	0			
A1, B1	5	0.688			
A2, B2	10	1.377			
A3, B3	15	2.064			

By hot pressing at 170 °C for 5 min., with a force of 50 kN, square plates of  $100 \times 100 \times 0.5$  mm<sup>3</sup> have been realized. After pressing, the samples were cooled to room temperature under a pressure of 5 bars.

The structure of samples and the dispersion of the fillers in polyethylene matrix were analyzed using the optical microscopy (with a NIKOV TI-e microscope) and Scanning Electron Microscopy SEM (with a workstation Karl Zeiss SMT-model Auriga and detector type Everhart-Thornley) [14].

The major hysteresis cycles were determined with a vibrating sample magnetometer VSM 7304 (produced by Lake Shore) for magnetization fields between 0 and 1000 kA/m). The very good absolute accuracy (better than 2% of reading  $\pm 0.2\%$  of full scale) and reproducibility (better



Fig. 3. Rheological behavior of A1 – A3 during melt processing in the Brabender LabStation; Torque vs. time curves.

(than  $\pm 1\%$ , or  $\pm 0.15\%$  of full scale, whichever is greater) allow the measurement of magnetic characteristics for weak magnetic samples (magnetic moment between 5 x  $10^{-6}$  emu and  $10^{3}$  emu).

For each type of material (A0, A1, A2, A3 and B0, B1, B2, B3, see Table I), 3 samples were manufactured. All measurements were performed 3 times on each sample and the average values were calculated.

## III. RESULTS AND DISCUSSIONS

For all samples, their structure and magnetic characteristics (hysteresis cycles, remanent magnetization and coercive field) were determined. Their dependencies on the nature and filler content were analyzed.

#### A. Melt rheology

The torque vs time curves were recorded for giving some insight into the rheological behavior of composites. Fig. 3 shows the plots for A0 - A3 samples. More rapid melting was observed for composites compared to neat polyethylene. This was expected because of the increased abrasion of the metallic filler which favor increased friction in the matrix and local increase of temperature. The influence of the coupling agent and of the local increase of the temperature is observed from the lower melt viscosity of composites compared to the matrix (A0).

No significant differences were observed between the melt viscosities of the composites, regardless of the filler content which emphasize the low influence of small amount of filler on the melt rheology of composites. For correctly explain the viscoelastic behavior of polyethylene-based magnetic composites, it is necessary to take into consideration the particle distribution, shape factor, particle-particle interactions as well as particle-polymer matrix interactions [55].

### B. Microscopic investigation

SEM analysis reveals a small (almost negligible) porosity in both surface and volume of the samples (P, Fig. 4).

Low density polyethylene (LDPE) shows a lamellar structure and neodymium particles form clusters (the so called metal "islands" [50]) (Figures 5 - 8) having variable dimensions (Figures 7 and 10, Table II). These clusters have a cvasiuniform distribution in the samples and the distance *d* between them decreases with the increase of the filler content (Figures 9 - 12, Table II). For example, in the case of samples A1 the distance *d* takes values between 99.5  $\mu$ m and 275  $\mu$ m and for A3 samples, the quantity *d* varies between 67.5  $\mu$ m and 211  $\mu$ m.

On the other hand, for the both types of samples (type A and B), the dimensions of the clusters increase with the filler content (Table II).

The presence of agglomerates greatly influences the maximum possible loading of the filler and the final rheological properties of the material. These agglomerates absorb the polymer in their inter-particles voids and pores, thereby increasing the viscosity in the melt processing step. Such increase of the viscosity in composites compared to the matrix was not observed in torque vs. time graph shown in Figure 3. This is probably due to the proper choice of processing parameters.



Fig. 4. Neodymium particles clusters and pores in LDPE matrix (SEM, 1000 X).

Sample	Clusters dimension (µm)	Distance between clus- ters (µm)
A0, B0	0	0
A1	95.92	117.81
A2	100.95	94.04
A3	103.6	67.5
B1	178.6	510.55
B2	200.0	415.0
В3	228.55	284.24

 TABLE II.

 DIMENSIONS AND DISTANCES BETWEEN CLUSTERS



Fig. 5. Neodymium particles clusters (metal "island") in A2 samples (Optical microscopy, 200 X).



Fig. 6. Neodymium particles clusters (metal "island") in A3 samples (Optical microscopy, 200 X).



Fig. 7. NdFeB particles clusters (metal "island") in B1 samples (SEM, 1000 X).



Fig. 8. NdFeB particles clusters (metal "island") in B3 samples (SEM, and 500 X).



Fig. 9. Dimensions between neodymium particles clusters in A3 samples (Optical Microscopy, 200X).



Fig. 10. Distances between neodymium particles clusters in A3 samples (Optical Microscopy, 200X).



Fig. 11. Dimensions between Nd-Fe-B particles clusters in B3 samples (SEM, 100 X).



Fig. 12. Distances between Nd-Fe-B particles clusters in B3 samples (SEM, 100 X).

## C. Magnetic properties

In Figures 13-15 the magnetization cycles of the samples A3, B1 and B3 for values of the magnetic field between 0 and 955 kA/m are presented. It was found that their shapes depend essentialy on the filler content. Thus, in the case of filler of Nd particles (samples A), the cycles are very thin and unsaturated (Fig. 13), while in the case of Nd-Fe-B (samples B) are larger and present magnetic saturation phenomenon (Figs. 14 and 15). It results that the samples A have the characteristics of soft magnetic materials, while the samples B of hard magnetic materials.

The shape of the hysteresis cycle for sample A3 (Fig. 13) is similar to the known magnetization characteristic of thin films. An explanation could be the small distance between clusters (see Table II), like the distance between two layers into a multi-layers microstructure. One must notice that the samples A1 and A2 have a weak magnetic moment, close to the magnetometer limits (5 micro-emu) and the measured hysteresis cycles are distorted. Although its magnetic moment of the neodymium atom  $m_{Nd}$  ( $m_{Nd} = 3.15 m_{B_1} m_B = 9.27 \cdot 10^{-24} \text{ A} \cdot \text{m}^2$  is the Bohr



Magnetic field (kA/m)

Fig. 13. Magnetization cycle for samples A3.





Fig. 14. Magnetization cycle for samples B1.

Fig. 15. Magnetization cycle for samples B3.

magneton) is greater than of Fe  $m_{Fe}$  ( $m_{Fe} = 2.24 m_B$ ), the samples A (Fig. 13) have, for the same value of the filler content, lower values of the magnetization than samples B (Figs. 14 and 15). This may be explained by the

interactions between the spin magnetic moments of the electrons with the unpaired spins from layers 3d (that corresponds to transition metal ions: Mn, Fe, Co, etc.) and 4f (that corresponds to rare earth ions: Nd, Sm, Gd, Tb, etc.), respectively to the exchange integral J [51-52]. In the case of elements from Fe group,  $J_{FeFe} > 0$  and the orientation of the atom magnetic moments is the same, the particles having a ferromagnetic behavior [51]. In the case of unpaired electron coupling from layers 3d (Fe ions) and 4f (neodymium ions) the exchange integral is also positive (respectively  $J_{FeNd} > 0$ ). The atom magnetic moments have also the same orientation (ferromagnetic behaviour) [53]. Therefore, the samples B (that contain Nd-Fe-B) have a ferromagnetic behavior and the magnetization is proportional with the concentration and module of filler magnetic moments.

Experiments performed on polymer nano composites with neodymium filler shown that the magnetic state in strongly influenced by the temperature: for temperatures greater than 19 K the bodies are in a paramagnetic state, if the temperature has values between 19 and 7.5 K the bodies pass in anti-ferromagnetic state and if the temperature is below 7.5 K these pass in a ferromagnetic state with a relative complicated magnetic structure [54]. On the other hand, as the interactions between the unpaired electrons of neodymium atoms are poor, is very difficult to determine the precise state of the neodymium particles [53]. Therefore, the samples A (with neodymium filler) have at the room temperature (where the tests were performed) a more paramagnetic behavior and the magnetization takes lower values than samples B (with Nd-Fe-B filler).

For samples with neodymium filler, the values of the coercive field  $(H_c)$ , remanence  $(M_r)$  and maximum magnetization  $(M_{max})$  are lower than for the Nd-Fe-B samples (Table III). This is due, as was shown above, to interactions between the unpaired electron moments of type Fe-Fe and Fe-Nd for samples B [42] and only Nd-Nd for samples A. On the other hand, the values of the ratio  $k = M_r/M_{max}$  are lower (k < 0.2) for samples A and greater (k > 0.73) for samples B. It comes out that Nd-Fe-B composite materials may be used to permanent magnets manufacture, while those based of Nd for magnetic circuits, electromagnetic shields [14] etc.

Increasing the filler content, the remanent and maximum magnetization increase also (Table III). The coercive field values decrease slightly (due to reduction of the distances between the magnetic particles and interactions intensification between them [42]).

TABLE III.

VALUES OF COERCIVE FIELD  $H_c$ , REMANENT MAGNETIZATION  $M_r$ ,

MAXIMUM MAGNETIZATION  $\,M_{\rm max}$  , MAXIMUM MAGNETIC

SUSCEPTIVITY	$\chi_{m,\max}$	AND RATIO	k	=	$M_r$	/	M max
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Sam- ples	$H_c$ (kA/m)	$M_r$ (kA/m)	$M_{\rm max}$ (kA/m)	χ <sub>m, max</sub>	k -
A1	38.42	0.00565	0.0309	0.013	0.178
A2	1.68	0.01369	0.0697	0.338	0.196
A3	0.35	0.02532	0.1680	0.580	0.151
B1	78.50	3.048	4.1046	16.8	0.743
B2	66.53	8.904	11.91	140.0	0.748
B3	52.69	13.90	18.96	235,0	0.733



Fig. 16. Variation of magnetic susceptivity with magnetic field strength for samples A.



Fig. 17. Variation of magnetic susceptivity with magnetic field strength for samples B.

Variations of magnetic susceptivity  $\chi_m$  with magnetic field strength *H* that correspond to the first magnetization curves (quadrant I) are presented in Figures 16 (for samples A) and 17 (for samples B). It comes out that, for samples A, values of  $\chi_{mA}$  decrease with *H*. Curves  $\chi_{mA}(H)$ present maxima for values of *H* that increase with the neodymium content of the samples (Fig. 16). For higher values of *H*, the values of  $\chi_{mA}$  are relative low. For samples B, curves  $\chi_{mB}(H)$  have greater values for low values of *H*, decrease to a minimum value (dependent on the filler content) and then increase to a maximum (for a value of *H* that increases more while the filler content is less).

Values of  $\chi_m$  are relatively small for both types of samples, due to the low volume filler content (below 2.1 %) [42, 44, 49]. On the other hand, the magnetic susceptivity has lower values for samples that contains neodymium (below 0.0031) than for neodymium-iron-boron samples (Table III). This is due to the magnetic interactions between orbitals 3d (Fe ions) and 4f (rare earth ions) that occur in the case of neodymium-iron-boron filler and that play a more important role on the magnetic properties values that those of rare earth ions [52].

On the other hand, the variations of the magnetic permeabilities  $\mu_{rA}$  and  $\mu_{rB}$  with the volume filler content  $c_{\nu}$  do not satisfy any of the equations (1)...(3). For example, in the case of samples A (for volume concentrations greater than 0.623 %), for maximum values of the susceptivity ( $\chi_{mA,max}$ ) and magnetic permeability ( $\mu_{rA,max}$ ) the following equations were obtained:

$$\chi_{mA,\max}(c_v) = -0.26 + 41.21c_v \tag{5}$$

$$\mu_{rA,\max}(c_v) = 0.74 + 41.21c_v, \tag{6}$$

The differencies between the equation (6) and the equations (1)...(3) are due to inhomogenities of shapes and filler particles dimensions, but also to lower values of the filler content.

The magnetization curves M(H) can be transformed in B(H) characteristics, taking into account the fundamental scalar relationship for an isotropic magnetic material:

$$B = \mu_0 (H + M). \tag{7}$$

The hysteresis losses  $\Delta w_m$  - variation of the volume magnetic energy density per cycle – can be computed according to Warburg' theorem:

$$\Delta w_m = \oint_{cycle} H \cdot \mathrm{d}B \tag{8}$$

Considering a linear variation between two consecutive measurement points, the integral has been approximated with a finite sum.

The hysteresis losses  $\Delta w_m$  values for A3 and B1, B2 and B3 samples are presented in Table IV. It is found that  $\Delta w_m$  takes higher values for neodymium-iron-boron filler. Thus, for the same value of volume filler content  $c_v =$ 2.064 %, the hysteresis losses are 676 times higher for Nd-Fe-B filler (samples B3) that for Nd ones (samples A3).

On the other hand, the increase of filler content leads, for both type of samples (A and B), to the increase of hysteresis losses. For example, in the case of B samples, if the filler content increases from 0.688 % to 2.064 %, the hysteresis losses increase from  $10.323 \text{ kJ/m}^3$  to  $48.231 \text{ kJ/m}^3$  (Table IV).

The significant increase of the hysteresis cycle area and hysteresis losses of polyethylene-based magnetic composites with Ne-Fe-B shows that, these composites can be used for permanent magnets manufacture. But, for such applications, the volume filler content of Nd-Fe-B must takes higher values (over 10 %) [55].

TABLE IV.

VALUES OF HYSTERESIS LOSSES *Aw*, FOR A AND B SAMPLES

Samples	$\Delta w_m [J/m^3]$
A3	71.38
B1	10322.71
B2	30281.33
B3	48231.26

# CONCLUSIONS

Neodymium particles (samples A) and neodymiumiron-boron (samples B) are grouped inside the bodies and give rise to metallic islands, where the polyethylene end chains are fixed. The dimensions of the islands vary between 95  $\mu$ m and 103  $\mu$ m for samples A and between 178.6  $\mu$ m and 228.5  $\mu$ m for samples B. The distances between clusters depend on the filler content and vary between 117.81  $\mu$ m and 67.5  $\mu$ m for samples A and between 510.55  $\mu$ m and 284.24  $\mu$ m for samples B.

The addition of Nd or Nd-Fe-B particles affects the rheological properties of the polyethylene matrix (by internal structural changes) and, consequently, the mechanical properties of the composites.

The magnetic properties of composites depend on the filler content nature, remanent and saturation magnetizations, coercive field and magnetic susceptivity taking values greater for samples with neodymium-ironboron than for neodymium samples.

Increase of the filler content leads to the increase of the magnetization, magnetic susceptivity and hysteresis losses, due to the feromagnetic interactions between the electrons from layers 3d and 4f (respectively Fe-Fe and Fe-Nd).

The values of the volume filler content used in this paper being lower (below 2.1 %), the magnetization is relative low and the histerezis cycles area and magnetic suscpetivity have also low values. Therefore, the magnets obtained from such composites have weak characteristics (comparing to the ferrites ones).

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