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Magnetic flexible material containing microcrystalline NdFeB powder

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A flexible magnet based on micro-crystaline NdFeB grinded powder and a polymeric matrix (EPDM) has been realized. The resulted composite shown high magnetic properties due to magnetic microcrystaline powder and good flexibility due to elastomer matrix. The influence of some parameters, such as grinding time and concentration of magnetic powder on magnetic and flexural properties are discussed in this paper.

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Keywords: Composite magnet, Microcrystalline NdFeB, Oxidation

1. Introduction

Flexible magnets are composite materials containing a magnetic component in an elastomer matrix [1,2]. These materials combine the elastic properties of rubbers with magnetic properties of the elastomer resulting flexible items useful in various technical applications. In this direction, high flexibility and high magnetic properties are usually required. Therefore, materials exhibiting high magnetic performance, such as Nd-Fe-B alloy become interesting for this kind of magnets.

This work is devoted to experimental features on a new material exhibiting both plastic (flexible) and high magnetic properties, due to its composition that includes an elastomer and a magnetic filler (Nd-Fe-B powder).

2. Experimental

2.1. Materials

Ethylene-propylene diene monomer copolymer (EPDM) used in this work, has been commercially available under trade name EPDM-C (ROMTER), Arpechim Piteşti. The main characteristics of this material are as it follows: unsaturated double bond content, 3.6%; Mooney viscosity, 36; polypropylene content, 40%; tensile strength, 13,6; elongation, 327; Shore durity, 67.5°Sh.

Magnetic powder was NdFeB MQP-A grade, with the characteristics shown in Table 1. Previously mixing with the elastomer, the microcrystalline powder has been submitted to a grinding process in order to reduce the particle size of the material. This operation has been performed in a 3 litter ball mill, using benzene as liquid for particle dispersion. Thus, 300g of NdFeB powder and 1 liter of benzene were been put inside and mixed for 30 minutes at 250 r.p.m. The resulted particle size has been around 80 microns as it has been evaluated by electron spectroscopy.

Several other magnetic materials, namely anisotropic barium ferrite, isotropic barium ferrite, and magnetite were used in order to compare the properties of the resulted NFeB magnets. All these substances were technical grade, for magnetic materials, and were used as received, without any supplementary transformation.

Table 1. Properties of MQP-A powder used to preparing the magnetic material.

Property	Units	Value
B_r	kGs	7.0
$_jH_c$	kOe	16.5
$_BH_c$	kOe	6.1
BH_{max}	MGOe	10.5

Mixing of magnetic component with elastomer has been preformed on a laboratory roller mixing device, the homogeneity of the resulted material being checked by picnometer density evaluation. Then, 50 mm in diameter and 0.5 mm or 1 mm in thickness plates were prepared using a hydraulic press (150 kgf/cm^2) at 160 °C for 5 minutes. Under these conditions, adequate thermal vulcanization of the product occurred.

The list and characteristics of the employed antioxidants is shown in Table 2. These substances were commercial available, technical grade, and were added during roller mixing treatment.

Table 2. Amine antioxidants tested in stabilization of EPDM rubber.

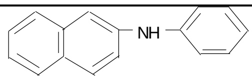
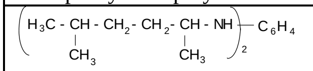
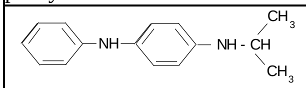
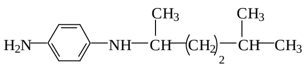
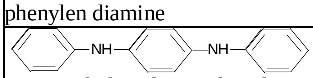
Chemical denomination	Trade name	M.w. (g / mol)	M.p. (°C)	Symbol
 N-phenyl-2-naphtyl amine	PBN	219	105	A_1
 N,N'-di-(1,4–dimethyl–pentyl)-*p* -phenylen diamine	Santoflex 77	304	-	A_2
 N-isopropyl, N'–phenyl-*p*–phenylen diamine	Santoflex IP	226	74	A_3
 N-(1,4 dimethyl–pentyl)-*p*–phenylen diamine	Santoflex 13	282	46	A_4
 N,N' - diphenyl - p - phenylen - diamine	DPPD	260	143	A_5

Table 3. Nd-Fe-B/ EPDM composites prepared in order to study the effect of concentration of magnetic phase on the functional properties.

Sample	Nd-Fe-B (g)	rubber (g)	% Nd-Fe-B (weight)	%rubber (weight)	% Nd-Fe-B (volume)	% rubber (volume)
M1	75	72	51	49	10	90
M2	150	64	70	30	20	80
M3	225	56	80	20	30	70
M4	300	48	86.2	13.8	40	60
M5	375	40	90.4	9.6	50	50
M6	450	32	93.4	6.6	60	40

A list of the flexible magnetic materials prepared in this work is shown in Table 3. Besides of above mentioned components, several other components were included in each formulation s it is shown in Table 4. All these components and mixing apparatus were kindly provided by ELDI Prod. SRL Pitești.

Table 4. Composition of the rubber formulations used in preparation of flexible NdFeB magnets

Component	gr	weight-parts
EPDM rubber (base elastomer)	80	100
Naftenic oil	40	50
Zinc oxide	4	5
Stearic acid	0.8	1
Sulphur	1.2	1.5
Tetramethyl thiuram disulphide	0.8	1
2-mercaptobenzotiazol	0.4	0.5
Antioxidant	1.2	1.5

2.2. Methods

Magnetization was carried out using impulse magnetization equipment, operating via capacitor set discharge on a coil. Magnetic measurements were performed on a hysteresis graph.

Evaluation of the flexibility has been performed with a device that is schematically presented in Fig. 1. Samples with the following dimensions: 30mm x 20mm x 1mm were cut from the vulcanized band to be used in these experiments. The sample has been fixed in the support and the free part (20 mm) has been moved as it is shown in Fig. 1, five times. The surface of the sample has been examined after each series of 5 flexions, using a magnifying glass, to observe the formation of cracks. Five similar samples were submitted to flexion test.

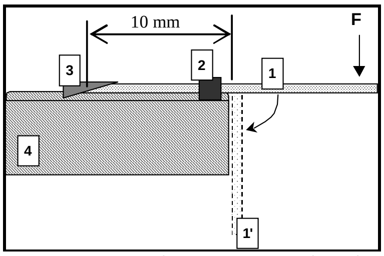


Fig. 1. The device used for assessment of the flexibility.

Thermal oxidation behavior has been monitored by a chemiluminescence CL 931 apparatus in isothermal mode, as it has been described in previous papers [3, 4].

3. Results and discussion

The comparative presentation shown in Fig. 2 illustrates the influence of the nature of magnetic component on, magnetic properties of the resulted composite. Nd-Fe-B induces very high magnetic properties as compared to conventionally used magnetic components for rubber magnets. The concentration of magnetic filler is presented in Table 1 in terms of volume or weight percent. Several samples of magnetic materials were prepared using various percent of Nd-Fe-B/ elastomer EPDM**.** The increasing in magnetic properties as increasing the concentration of magnetic component is shown in Fig. 3.

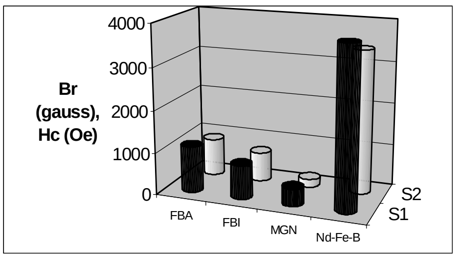


Fig. 2. Influence of the nature of magnetic component on the magnetic properties of some flexible magnets (concentration of the magnetic component = 30%): FBA = anisotropic barium ferrite; FBI = isotropic barium ferrite; MGN = magnetite; Nd-Fe-B = magnetic alloy MQP-A grade; $S_1 = B_r$ values; $S_2 = H_C$ values; elastomer matrix: vulcanized EPDM (ethylene-propylene diene monomer).

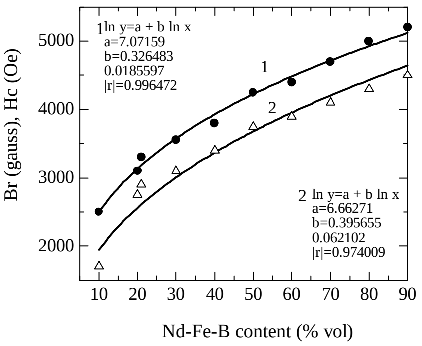


Fig. 3. B_r (1) and H_c (2) as functions of Nd-Fe-B content for various Nd-Fe-B/ EPDM compositions.

All samples tested for flexibility resisted more than 10 series of five flexions without observing any crack on their surface, so we can consider that the material shown a good flexibility. This flexibility could be assigned to the good mechanical properties of the rubber matrix as well as to the lubricating effect of some components, such as naftenic oil and stearic acid.

Oxidation behavior of EPDM and of NdFeB containing samples has been also studied being known that these materials undergo oxidation during exploitation, due to organic nature of the polymer component. To increase the stability of the organic materials, the effect of several amine antioxidants were tested Fig. 4. Their effectiveness could be described by the relative activity calculated with the formula:

$$A_r = \frac{(t_i)_s}{(t_i)_o} \quad (1)$$

where $(t_i)_S$ is the oxidation induction time of the stabilized sample, and $(t_i)_0$ is the same parameter in the case of unstabilized sample.

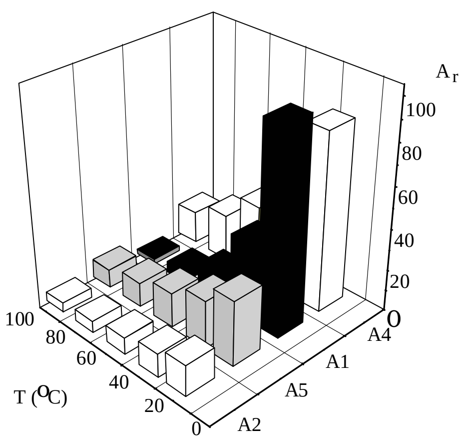


Fig. 4. Initial relative activity as a function of temperature for various amine antioxidants: A_2 - NR + Santoflex 77; A_5 - NR + DPPD; A_1 - NR + PBN; A_4 – NR + Santoflex IP.

Oxidative stabilization of the material is required because Nd-Fe-B alloy and other transitional metals facilitate the thermal oxidation of the various hydrocarbon polymers [5,6]. Thus, a relative small amount of Nd-Fe-B powder induced a significant decreasing in the oxidative stability of the material as it is shown in Fig. 5. The main effect seems to concern the oxidation induction time, which is reduced in the case of NdFeB containing sample. Also, the time to reach the maximum of CL signal is lower in the case of crystalline powder containing sample. In fact, great changes could be observed in the case of the shoulder on the increasing portion of CL curve. This shoulder decreased in the intensity, and it appears after shorter oxidation time in the case of EPDM containing NdFeB. Such a shoulder has been assigned to oxidation of some specific places of the polymer, which are more reactive than the whole polymer, such as chain branches or peculiar hydroperoxide structures [7]. These groups seem to be strongly influenced by presence of NdFeB particles.

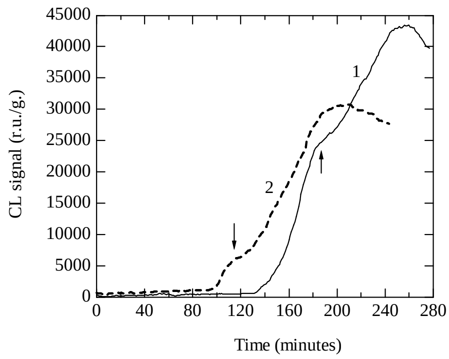


Fig. 5. CL curves at 190°C from EPDM (1) and EPDM + 0,5% NdFeB.

4. Conclusions

Flexible NdFeB plastic magnets prepared by mixing of ground NdFeB microcrystalline powder and a synthetic copolymer (ethylene-propylene-diene monomer) exhibited high magnetical properties as compared to other similar materials prepared by different magnetic components.

High magnetical properties and very good flexibility make this material interesting for several appliances, such as electrical engineering, micro-engineering, decorative applications a.s.o.

Catalytic effect of the metal component in thermal oxidation of EPDM matrix has been observed, that needs supplementary stabilization in order to ensure adequate lifetime of the resulted material.

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