MICROSTRUCTURE AND MAGNETIC PROPERTIES OF SINTERED ND-Fe-B MAGNETS

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Abstract

Sintered Nd-Fe-B permanent magnets with a medium coercive force were fabricated using a strip-casting alloy prepared at the VSB-TU Ostrava. Powders prepared by hydrogen decrepitation and subsequent milling were compacted in a magnetic field and sintered at T = 1340 K (for 2 h). The sintered blanks were subjected to optimum heat treatment at 500 °C for 2 h followed by quenching in gaseous nitrogen. Detailed studies of the microstructure, phase composition and magnetic properties of heat treated samples were performed. The composition and structure of samples were examined using X-ray diffraction (XRD) and scanning electron microscopy JEOL JSM-6610LV. Magnetic properties were measured using vibrating-sample magnetometer and hysteresisgraph AMT-4. The higher coercive force was reached after aging at 500 °C, which was performed after cooling from sintering temperature to room temperature.

Keywords: $Nd_2Fe_{14}B$ alloy, sintered magnets, heat treatment, coercive force

1. INTRODUCTION

Ever-growing applications of Nd-Fe-B permanent magnets, which, in particular, are the most promising materials for driving motors of the hybrid electric vehicles, determine widening the geography of their preparation and studies of their properties.

Since the operating temperature of magnets in these motors reaches 200 °C, the very high coercive force H_{ci} at room temperature is necessary. The strip-casting technique and powder metallurgical method, which is used for the preparation of powder of the required dispersity and with the maximum volume fraction of principal hard magnetic $Nd_2Fe_{14}B$ phase, are applied for the manufacture of high-energy Nd-Fe-B magnets [1, 2]. The effective way to increase H_{ci} is the substitution of Dy and Tb for Nd in the 2-14-1 phase and the replacing of the conventional Nd-rich constituents by small additions of a low-melting Dy,Tb/Co-based alloys [3, 4]. Additions of alloys harden the $Nd_2Fe_{14}B$ phase boundaries without considerable losses in the saturation magnetization of magnet. The H_{ci} value of sintered magnets can be further enhanced by a heat treatment around 500 °C [5, 6], but, up to now, the origin of such coercivity enhancement during annealing is under discussion.

The goal of this work was to prepare sintered Nd-Fe-B magnets from strip-casting alloy of basis composition for the subsequent possibility to improve their properties with various heat treatments and dysprosium hydride additions.

2. EXPERIMENTAL PROCEDURE

The $Nd_2Fe_{14}B$ magnets were prepared using the strip-casting alloy, whose composition, as determined by electron microprobe analysis after the alloy preparation, is given in **Table 1**. As is seen, the strip-casting alloy contains a marked amount of oxygen. The charge composition was 63.93 Fe; 34.76 Nd; 1.30 B (wt.%). Flakes with thickness from 0.3 to 0.5 mm (**Fig. 1**) were produced at the "Regional Materials Science and Technology Center" at VSB-TU Ostrava. The strip-casting technology is based on the induction melting of the material in vacuum followed by its pouring on water-cooled rotating copper cylinder (v = 2 - 2.5 m/s), thus avoiding creation of long dendrites of magnetically soft phase - Fe, and Nd - rich phase is finely and homogeneously distributed throughout the volume of material. All the process parameters were precisely controlled in order to achieve an optimum grain size, i.e. temperature of the molten alloy, the melt flow rate, the shape of the melt flow, coolant temperature, etc.

Table 1 The average chemical composition of strip-casting flakes

Phase	OK	AlK	NdL	FeK
	(wt.%)			
N1	3.47	0.64	33.97	61.92

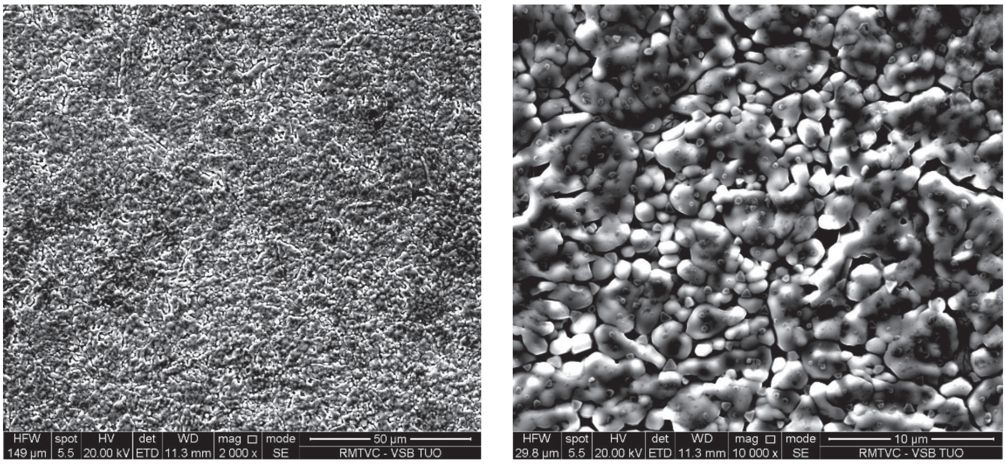


Fig. 1 SEM images of the surface of strip-cast flakes

The magnet preparation procedure includes the hydrogen decrepitation of flakes in dry hydrogen (at 375 K for 1 h) and subsequent passivation in gaseous nitrogen atmosphere. After cooling the powder to room temperature, the mixture was subjected to fine milling for 40 minutes to an average particle size of 3 μm using a vibratory mill and isopropyl alcohol medium. Samples were compacted in a magnetic field and sintered at T = 1340 K (for 2 h). The sintered blanks were subjected to heat treatment at 500 °C for 2 h followed by quenching in gaseous nitrogen.

The structures of the samples were examined using X-ray diffraction (XRD) and scanning electron microscopy. X-ray diffraction analyses were performed using a DRON X-ray diffractometer with Co $K\alpha$ radiation and graphite monochromator. The samples of initial and treated alloy were grinded into the powder (particles less than 100 μm) for X-ray diffraction patterns obtaining. The samples microstructure was analyzed using JEOL JSM-6610LV scanning electron microscope. Magnetic properties of strip-cast flakes and powder samples were measured using vibrating-sample magnetometer. The hysteretic properties of as-sintered magnets were measured by hysteresisgraph AMT-4 in magnetizing field up to 2000 kA/m.

3. RESULTS AND DISCUSSION

The microstructure of basic alloy in form of flakes, which were prepared by strip casting method, is documented in **Fig. 2a**. For comparison, **Fig. 2b** shows the structure of strip-casting alloy prepared at the VNIINM (company "A.A. Bochvar High-technology Research Institute of Inorganic Materials") and earlier used in our studies [7-

8]. As is seen, the dendrite-like structure is formed by two phases. Bright phase regions correspond to REM-rich phase and dark matrix is the Fe-rich phase whose composition is close to $Nd_2Fe_{14}B$. Both phase precipitates are rather dispersed. This fact determines the preparation of fine powder particles by hydrogen decrepitation and subsequent milling and sufficiently uniform distribution of the phases in powder particles.

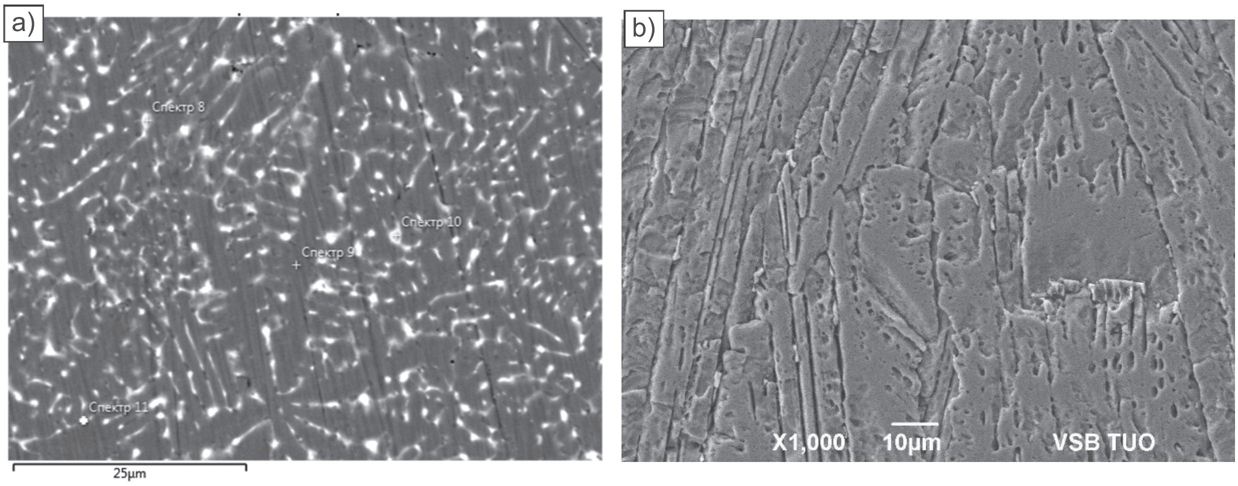


Fig. 2 The microstructure of flakes of main alloy prepared by strip casting method at the VSB-TU (a) and at the VNIINM (Moscow, Russia) (b)

Fig. 3 shows micrographs of (a) as-sintered and (b) heat-treated Nd-Fe-B magnets. As is seen from **Fig. 3a**, a bright phase enriched in Nd is present in the structure in the form of individual large 2-14-1 phase grains and precipitates at triple junctions of the the phase grains. Grain boundaries are decorated with oxides and are observed only between some grains of the phase. This fact indicates the insufficiently uniform distribution of Nd-rich phase over the volume of the sintered magnet. The annealing of sintered magnet at 500 °C for 2 h led to the spread of Nd-rich phase along grain boundaries and formation of thin inter-grain layers. The presence of such layers, as is known, improves the magnetic isolation of principal phase grains and leads to an increase in the coercive force.

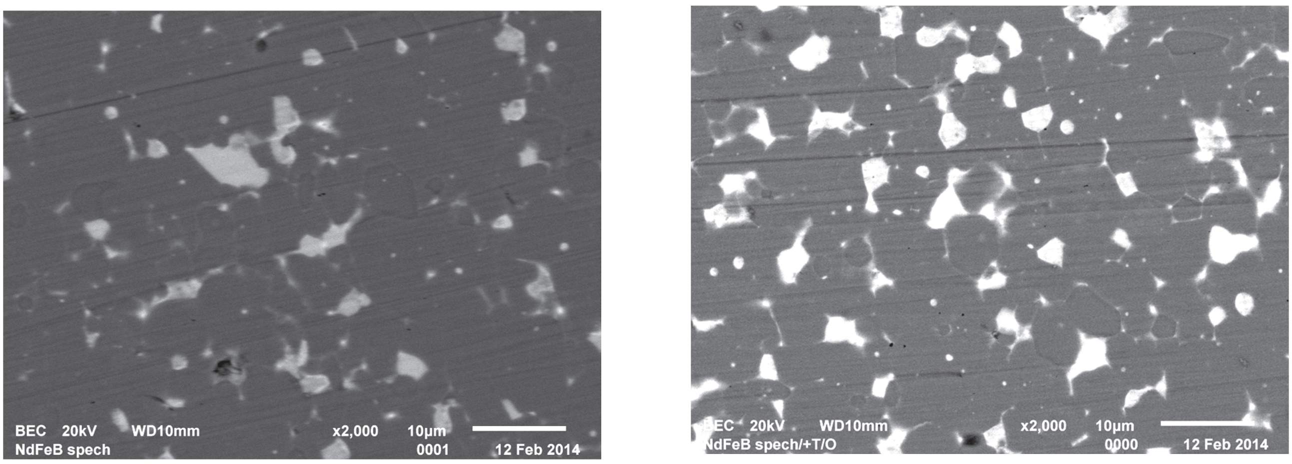
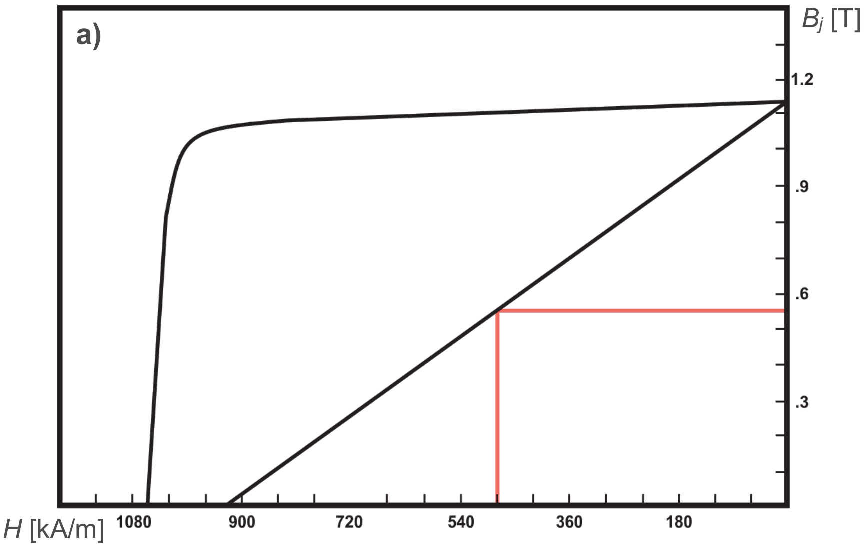


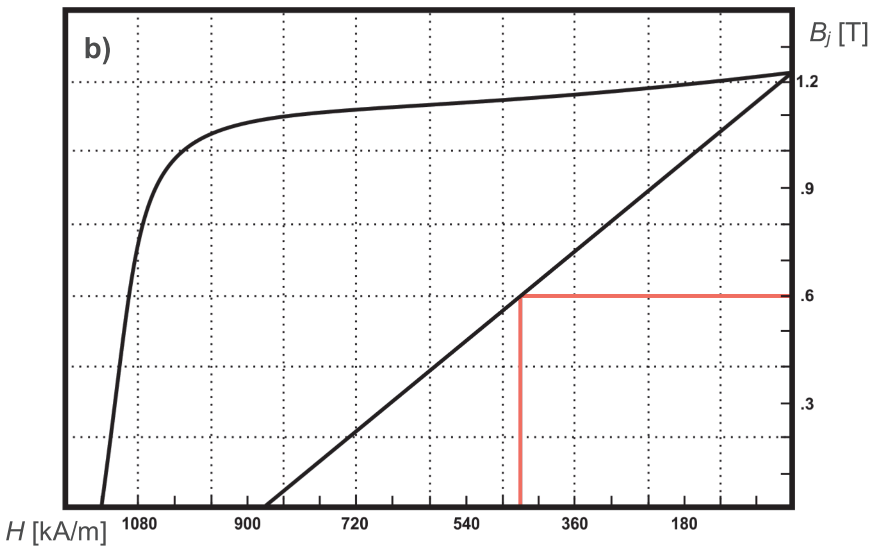
Fig. 3 Micrographs of sintered magnets: (a) after sintering and (b) after heat treatment at 500 °C for 2 h

Fig. 4 shows magnetization reversal curved for the (a) sintered and (b) subsequently heat-treated Nd-Fe-B magnets. Hysteretic properties derived from the curves are given near the curves.



Results:
B_r 1.2319 T
H_{cb} 914.79 kA/m
H_{cj} 1047.0 kA/m
$(BH)_{max}$ 286.07 kJ/m^3
H_k 973.45 kA/m
H_k/H_{cj} 93 %
H_d 469.25 kA/m
B_D 0.60927 T
H_m 1997.9 kA/m
Size:
Length 1.628 cm
Width 1.45 cm
Coil area 5.6 cmxcm
Wins 19

b)



Results:
B_r 1.2199 T
H_{cb} 868 kA/m
H_{cj} 1140.3 kA/m
$(BH)_{max}$ 266.5 kJ/m^3
H_k 843.76 kA/m
H_k/H_{cj} 74 %
H_d 447.26 kA/m
B_D 0.59584 T
H_m 2271.5 kA/m
Size:
Length 0.87 cm
Width 0.75 cm
Coil area 2.493 cmxcm
Wins 40

Fig. 4 Magnetization reversal portions of hysteresis loops for sintered (a) and heat treated (at 500 °C for 2 h) (b) magnets

As is seen, the annealing led to slight increase in the coercive force H_{cj}, but to decrease in the residual inductance B_r and maximum energy product $(BH)_{max}$ as well. The worsening of squareness of hysteresis loop is also taken place (the ratio H_k/H_{ci} decreases from 93 to 74 %). The found changes can be related to the presence of oxygen in the alloy composition and to the fact that the used annealing temperature was not optimum.

CONCLUSION

The data obtained in the present study for sintered magnets, which were manufactured from the strip-casting alloy, allow us to conclude the possibility of preparation of sintered Nd-Fe-B magnets in using the strip-cast alloy and improve their properties with additions of heavy rare-earth metal hydrides (in accordance with our previous studies) and Dy,Tb/Co-based alloys as well.

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