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NANOCOMPOSITE PERMANENT MAGNETIC MATERIALS Nd-Fe-B TYPE / THE INFLUENCE OF NANOCOMPOSITE ON MAGNETIC PROPERTIES

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Abstract

The influence on the magnetic properties of nanocristalline ribbons and powders has character of microstructure, between others – the grain size, volume of hard and soft magnetic phases and their distribution. Magnetic properties of ribbons and powders depend mainly on their chemical composition and parameters of their heat treatment [1]. Technology of magnets from nanocristalline ribbon consists of the following process: preparing the Nd-Fe-B alloy, preparing the ribbon, powdering of the ribbon, heat treatment of the powder and finally preparing the magnets. Nanocomposite permanent magnet materials based on Nd-Fe-B alloy with Nd low content are a new type of permanent magnetic material. The microstructure of this nanocomposite permanent magnet is composed of a mixture of magnetically soft and hard phases which provide so called exchange coupling effect.

Keywords: melt-spun Nd-Fe-B, heat treatment, nanocomposite, exchange coupling, magnetic properties

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1. Introduction

Nanocomposite permanent magnet materials based on Nd-Fe-B alloy with Nd low content are a new type of permanent magnetic material. The microstructure of this nanocomposite permanent magnet is composed of a mixture of magnetically hard and soft phases. Principle of exchange coupling between soft and hard magnetic grains is shown on Fig.1 [2]. Depending on the alloy composition, soft magnetic phases are one or two of α-Fe, Fe_3B and the hard magnetic phase is $Nd_2Fe_{14}B$. Essential conditions for microstructure of such material is uniform distribution of phases on a scale of the order of 5 - 20 nm, and exchange coupling between hard and soft phases. The nanocomposite magnet produced by this route exhibitet very high remanence, relatively high energy product $(BH)_{max} \sim 95 - 100$ kJ/m^3, in spite of the fact that the fraction of the hard magnetic $Nd_2Fe_{14}B$ phase was only 15% of the alloy. Nd content in nanocomposite magnetic materials is decreased for 50at% in relation to $Nd_2Fe_{14}B$ with stehiometric Nd content [3]. Among the advantages of these magnets is low material cost due to the reduction of the content of the expensive hard magnetic phase. These materials have a high

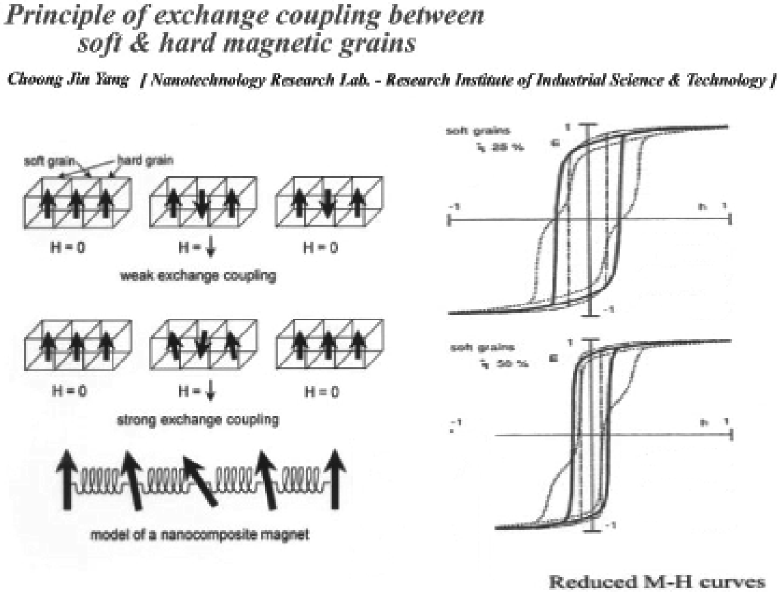

Fig. 1. Principle of exchange coupling between soft and hard magnetic grains

potential to be developed into high – performance permanent magnets with very high energy product.

During the research of high coercive magnetic materials based on Nd-Fe-B alloy, special activities in research are focused on nanocomposite Nd-Fe-B materials with Nd low content. The rapid quenching technology (melt-spun) for obtaining high-coercive magnets of this type gives the possibility to influence on the grain size and microstructure through the cooling rate. The cooling rate range in which optimal magnetic properties are achieved is rather narrow so that the heat treatment is needed in order to achieve the optimal magnetic microstructure. Optimization of microstructure is the key to improve the hard magnetic properties of these nanocomposite magnets [1].

2. Experimental

Investigated Nd-Fe-B alloy with Nd low content was prepared by rapid quenching (R/Q) under Ar atmosphere. The selected cooling rate was 20 m/s. In presented research different methods of analysis were used. The temperature behavior aimed at selection of a favorable regime of heat treatment of the investigated melt-spun alloy was examined by the DTA. The phase transformation in the function of cooling rate and heat treatment regime for defined initial chemical composition of melt-spun Nd-Fe-B alloy were investigated by determining the phase composition, by application of XRD and with Mössbauer spectroscopic phase analysis. For determining the critical temperature of phase and magnetic transformations, thermomagnetic measurements were carried out. The thermomagnetic curves were completed with hysteresis loops in stages before and after the TM treatment. Magnetic properties in the function of the investigated parameters were measured on the VSM (vibratory sample magnetometer) with magnetic field strength of 50 kOe.

Comparison of the experimental results obtained by different investigation techniques enabled more complete comprehension of the crystallization process and phase composition during the heat treatment.

Mössbauer spectra were taken in the standard transmission geometry with the Co^{57} (Rh) source at room temperature. The calibration was done against the a-iron foil data. For the spectra fitting and decomposition the CONFIT package was used. For thermomagnetic measurements weakly compacted material of the cylindrical shape with a diameter of 2 mm and thickness of about 1.5 mm was used. The thermomagnetic measurement was carried out in the field of 50 Oe with the temperature sweep of 4 K/min.

3. Results and discussion

Investigated Nd-Fe-B alloy with Nd low content was prepared by rapid quenching (R/Q) under Ar atmosphere with optimal selected cooling rate [1, 3, 4]. The as-quenched alloy had Nd-12.0 mass %, Pr-02 mass %, B-4.2 mass %, Fe-balance.

Thermal behavior of investigated alloy was done by DTA in the temperature range from 20 ^{0}C to 900 ^{0}C for the reason of getting optimal heat treatment regime. DTA curves are presented on Fig 2.

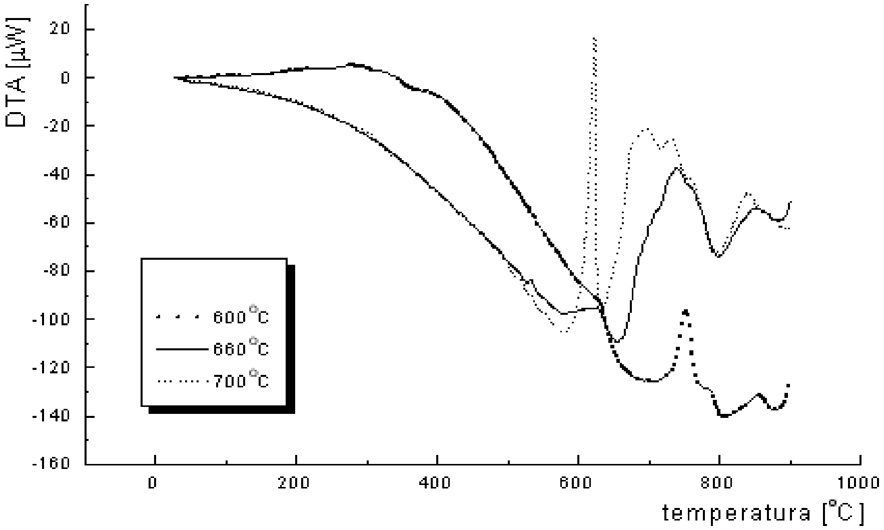

Fig. 2. - DTA curves of investigated melt-spun Nd-Fe- B alloy as a function of the annealing temperature. Annealing time: 5 min

According to the most of authors, in investigated alloy with low Nd content in temperature interval from 600^{0}C to 700^{0}C Fe_3B, $Nd_2Fe_{14}B$, $Nd_2Fe_{23}B_3$

phases and α-Fe phase are present [5, 6, 7]. On the DTA curve for investigated sample (Fig.2) first crystallization peak which corresponds with primary crystallization of Fe_3B phase (phase of high magnetization) can be observed.

That is proved by XRD analysis and MS analysis [1, 4]. Second peak on DTA curve of weaker intensity corresponds with crystallization of hard magnetic phase $Nd_2Fe_{14}B$. By increasing of temperature of heat treatment in investigated interval, it was shown that density of fine Fe_3B particles is increased, and that they are uniformly distributed in the amorphous matrix for the short time of heat treatment [7, 8, 9]. By analyzing of XRD diffractogram after heat treatment at 600^0C during 5 minutes, following phases are identified: $Nd_2Fe_{23}B_3$, Fe_3B, α-Fe and $Nd_2Fe_{14}B$ [10]. This corresponds with experimental results of DTA and with theoretical analysis of crystallization flow [8, 9, 10]. For the heat treatment at 660^0C during 5 minutes, same phases were identified but in different volume rations [4, 10]. Metastable phase $Nd_2Fe_{23}B_3$ crystallizes as intermediary phase, and in further process of crystallization during heating decompositions to other metastable phases: phase of high magnetization Fe_3B, main magnetic phase $Nd_2Fe_{14}B$, and small amount of soft magnetic α-Fe phase. Amount of $Nd_2Fe_{14}B$ phase comparing to Fe_3B increases with increase of temperature of heat treatment, and that leads to increase of coercive force [7, 8, 9]. By XRD analysis of investigated alloy heat treated at 700^0C during 5 minutes phases $Nd_2Fe_{14}B$, $Nd_{1.1}Fe_4B_4$, α-Fe and Fe_3B were identified [1]. Metastable $Nd_2Fe_{23}B_3$ phase decays completely at 700^0C which proves that crystallization of identified phase has been conducted completely [8, 9, 10]. According to the results XRD and MS analysis, because of relatively high boron content, $Nd_{1.1}Fe_4B_4$ phase is also present [1]. On the basis of DTA (Fig.2) and XRD results optimal heat treatment regime was selected (660°C for 5 min). On the Fig. 3 Mössbauer spectra is presented.

The Mössbauer spectra of this material after both thermomagnetic measurement cycles are very complex and not all components could be identified exactly. MS spectra illustrate the substantial difference between the state with optimised magnetic properties (Fig. 3.a) and the state after the decomposition induced by further heating above optimal temperature of heat treatment (Fig. 3. b).

MS spectra (Fig. 3.b) exhibit the decay of the $Nd_2Fe_{23}B_3$ phase and significant increase of the α-Fe phase content, above all. The α-Fe and the whole set of Fe-B phases prevail. This thermal decomposition will be the main reason for the quality loss of this hard magnetic material.

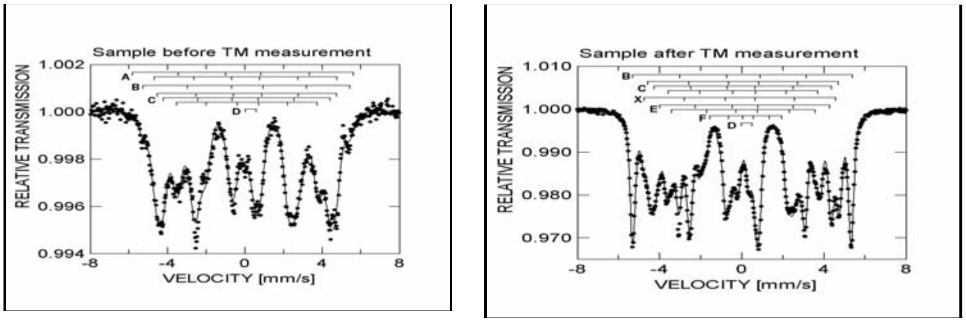

Fig. 3. Mössbauer spectra taken before (a) and after (b) thermomagnetic measurements [A – $Nd_2Fe_{23}B_3$, B – á-Fe, C – Fe_3B, D – $Nd_{1.1}Fe_4B_4$, E – Fe_2B, F – FeB; X – belongs probably to the $Nd_2Fe_{14}B$ phase]

In Table 1. B_r, H_c and BH_{max} values are presented in the function of the heat treatment regime. After heat treatment at 660^0C for 5 min maximal magnetic properties were obtained and therefore this regime was selected as an optimal regime of heat treatment for this type of magnetic materials with low Nd content.

Table 1. Magnetic properties in the function of the heat treatment regime

[1] HEAT TREATMENT REGIME	*[2] Br, kG*	*[3] Hc, kOe*	*[4] $(BH)_{max}$, MGOe*
[5] 600^0C / 5 min	*[6] 12.0*	*[7] 2.4*	*[8] 8.0*
[9] 660^0C / 5 min	*[10] 10.9*	*[11] 2.8*	*[12] 10.7*
[13] 700^0C / 5 min	*[14] 11.0*	*[15] 3.2*	*[16] 8.8*

Difference in optimum values may arise from the fact that those two different mechanisms: the exchange coupling and the phase content govern

Hci and Br. While exchange coupling is important for both Hci and Br, the amount of the soft phase competes with the exchange interaction in its effect on Br [11].

4. Conclusion

The magnetic coercivity and mechanical strength have analogous dependence on the grain size. Increase of Hc value during the heat treatment as a result has an increase of BH_{max}. For optimally selected heat treatment regime optimal values of BH_{max} were obtained. This indicates that decrease of grain size has occurred and the grain refinement thereby is strengthening the exchange interaction of soft and hard grains and increasing Hc.

Based on measured magnetic properties of investigated Nd-Fe-B alloy with reduced Nd content it can be concluded that it is very sensitive to the heat treatment which has direct influence on the magnetic microstructure and therefore coercivity.

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