Science of Sintering, **37** (2005) 139-145

UDK 537.622:550.835.6

Magnetic Properties of Nd Rich Melt-Spun Nd-Fe-B Alloy

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Abstract:
As a part of these experimental investigations of melt-spun Nd-Fe-B alloy with Nd rich content in relation to $Nd_2Fe_{14}B$ prepared by rapid quenching process for optimally selected cooling rate and heat treatment, the influence of the chosen chemical composition on magnetic properties was observed. The results of X-ray diffraction, Mössbauer spectroscopy phase analysis and magnetic measurement of investigated melt-spun $Nd_{14.5}Fe_{78.5}B_7$ alloy are presented to bring some new information concerning the relation between their structure and magnetic properties.
Keywords: *Melt-spun Nd-Fe-B; Phase composition; Magnetic properties; Mössbauer spectroscopy; XRD; SQUID.*

Introduction

Melt spun Nd-Fe-B alloys are an important class of permanent magnets containing $Nd_2Fe_{14}B$ as a principal phase [1]. The presence of soft magnetic phases like α-Fe and Fe_3B is usually detrimental to the hard magnetic properties [1, 2, 3]. Conventional alloy compositions are therefore selected to avoid these phases and form a final phase mixture of microcrystalline $Nd_2Fe_{14}B$ with Nd-rich grain boundary phases [1, 2].

The advantage of using the rapid quenching technology for obtaining high-coercive Nd-Fe-B magnets reflects in the possibility to influence directly on the grain size and microstructure through the cooling rate aimed at achieving a magnetic microstructure which provides maximal magnetic energy [1, 2, 3, 4]. The cooling rate range in which optimal results are achieved is rather narrow so that heat treatment of melt-spun Nd-Fe-B alloys is needed in order to achieve the maximal coercivity [4, 5, 6]. Arrangement of atoms in the short range (near single atomic radius) and defects of different levels have a very important role in the formation of magnetic properties [7]. The most important parameters which define magnetic microstructure of nanocrystaline magnetic alloys are determined by optimization of

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alloy composition, production route and heat treatment regime. For this reason the study is extended to investigate effects of the most convenient regime of heat treatment for the selected cooling rates on magnetic properties.

Experimental

The magnetic properties of melt-spun $Nd_{14.5}Fe_{78.5}B_7$ alloy prepared by rapid quenching (R/Q) on the external surface of a spinning copper wheel for selected cooling rates under Ar atmosphere were investigated. The investigated cooling rates were in the range from 15 – 20 m/s corresponding to the speed of the copper wheel. The starting alloy composition was Nd 32 mass%, Pr 0.5 mass%, B 1.2 mass%, Ti 1.0 mass% and Fe – balance. The heat treatment of as quenched ribbons was performed on 630^0C for 5 min. The phases present after heat treatment were determined by XRD and Mössbauer ^{57}Fe spectroscopic phase analysis (MS). X–ray diffraction was conducted on a Philips diffractometer with a copper anticathode (λ=0.151478nm) in the range of 2θ from 0^0 to 90^0. Mössbauer spectra were taken in standard transmission geometry using the Co^{57} (Rh) source at room temperature [8]. The calibration was done against the α-iron foil data. The CONFIT package was used for the spectra fitting and decomposition [9]. The MS phase analysis was based on the data published in [9]. Magnetic measurements at 300 K were performed on a SQUID magnetometer with an external magnetic field that can be varied from -5 to 5 T and on a vibration sample magnetometer (VSM) with a maximum external field of 50 kOe. Magnetic behavior was observed through the relative change of magnetic permeability. Measurements of the relative change of magnetic permeability were carried out by a modified Faraday method. Samples were pressed into pills of 8 mm diameter under pressure of 400 MPa. Pressed samples were placed in front of the solenoid where the gradient of the magnetic field was $\Delta H/\Delta z$ = 1.26 mT/mm. After magnetic measurements, XRD analysis and Mössbauer spectroscopy phase analysis were performed to correlate the microstructure and phase composition with magnetic properties for selected cooling rates and heat treatment regime.

Results and discussion

SEM micrographs of melt spun $Nd_{14.5}Fe_{78.5}B_7$ alloy prepared by rapid quenching at the selected cooling rate are shown in Fig 1. Platelets with long dimensions around 200 µm with a thickness of 25-35 µm are typical.

The X-ray diffractogram of $Nd_{14.5}Fe_{78.5}B_7$ annealed at 630^0, 5 min is shown in Fig 2. By observing the results of XRD analysis, the hard magnetic phase $Nd_2Fe_{14}B$ is identified as the primary phase. Due to low reflection intensity and a great number of reflections of the identified primary phase, it was not possible to define by this analysis to which phases the unidentified diffraction maximums belong.

For better insight in the phase composition of the investigated melt-spun $Nd_{14.5}Fe_{78.5}B_7$ alloy a MS analysis was conducted after the heat treatment. Analysis of the Mössbauer spectra [9] identified the main magnetic phase $Nd_2Fe_{14}B$ as the primary phase. Its content is estimated to 85 mass%. The presence of a non-fero magnetic phase $Nd_{1.1}Fe_4B_4$ (about 6 mass%) and α–Fe (at the level of about 3 mass%) was also noted. The appearance and identification, actually in small amounts, of a non- ferro magnetic boride phase type $Nd_{1.1}Fe_4B_4$ is the result of an increased B content of about 7.26 at% in the investigated alloy. Also, small amounts of unidentified components are detected. They obviously represent remnant minor paramagnetic phases, predominantly those of a high Nd content situated on grain boundaries [10, 11]. Most probably these undesirable phases are presented in nano size and therefore do not derogate magnetic properties. Comparing the experimentally obtained values of the lattice parameters a = 8.8052(3); c = 12.2061(6) for tetragonal $Nd_2Fe_{14}B$ phase with the standard values: a = 8.8050(50); c = 12.2050(50) [12], a very high closeness of these values can be ascertained (Table I).

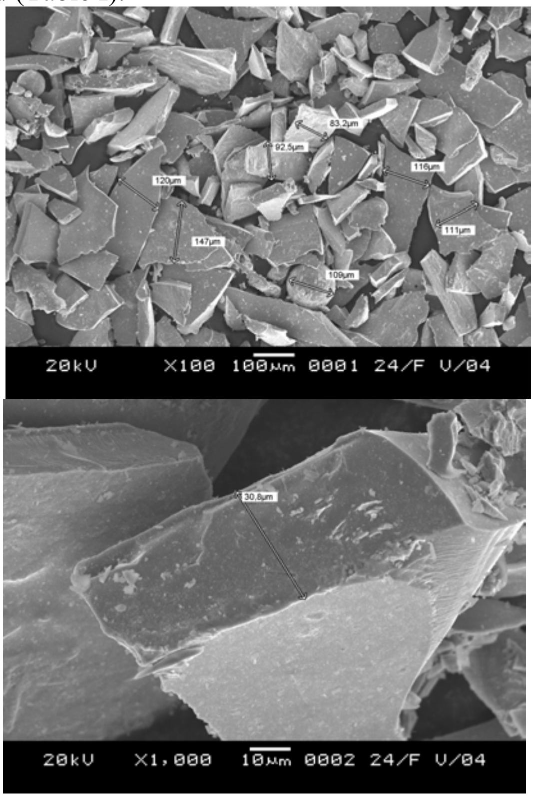


Fig. 1 SEM micrographs of melt-spun $Nd_{14.5}Fe_{78.5}B_7$ alloy

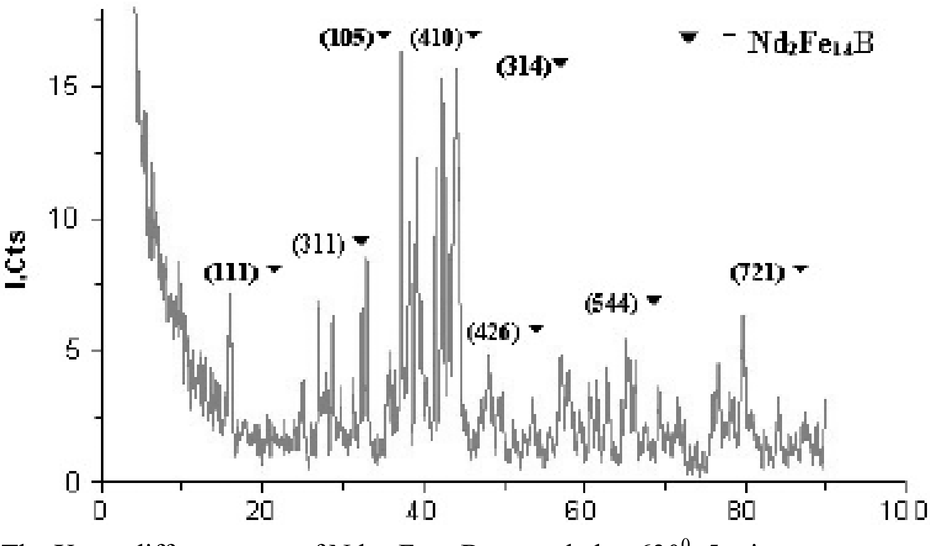


Fig. 2 The X-ray diffractogram of $Nd_{14.5}Fe_{78.5}B_7$ annealed at 630^0, 5 min

It can be concluded that other identified and unidentified phases, being in small volumetric fractions, had no substantial influence on the structural defects which could directly cause reduction of magnetic properties.

The relative change of magnetic permeability of $Nd_{14.5}Fe_{78.5}B_7$ alloy in the function of temperature for two heating cycles is presented on Fig. 3.

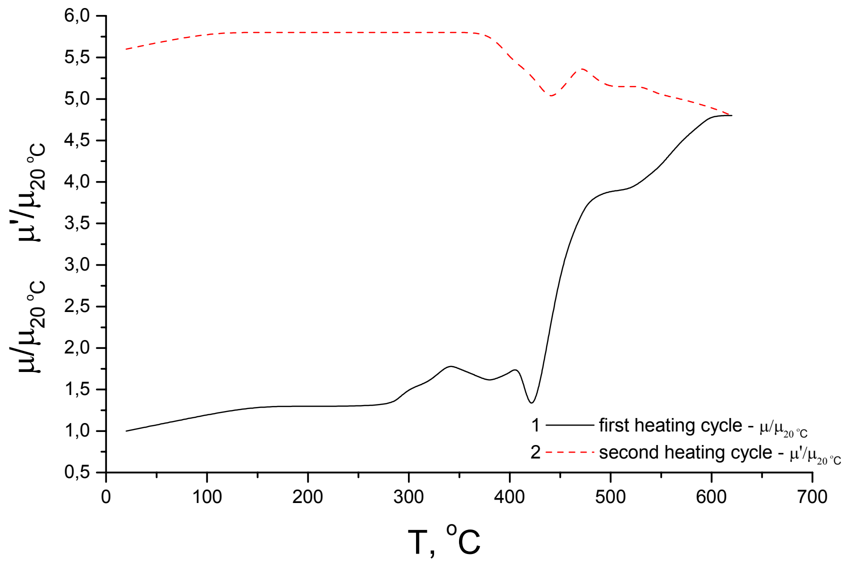


Fig. 3 Temperature dependence of relative change of magnetic permeability

The results show thermal stability of samples up to ~ 300°C. It can be assumed that the increase of relative change of magnetic permeability in the temperature range from 400°C – 620°C comes mainly as a result of larger domain wall mobility after the annealing treatment due to refinement of the microstructure [1, 3, 7]. Relative change of magnetic permeability of the samples heated on the maximal temperature of measurements (620°C) increased about five times (Fig. 3, curve 2). It is obvious that after cooling, during the second heating cycle magnetic permeability reaches the optimal value and retains this value up to 620°C.

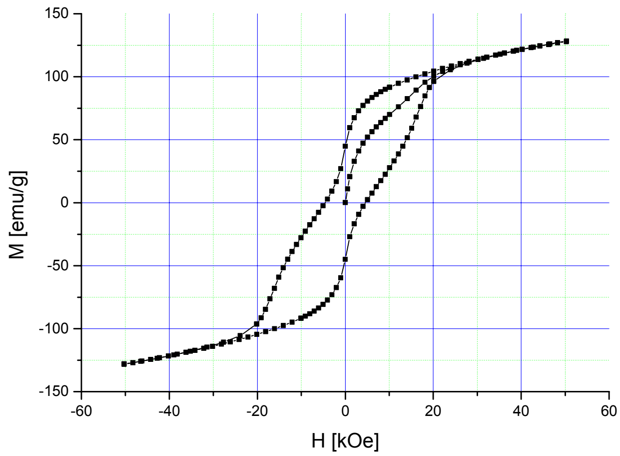
Fig. 4. shows hysteresis loops obtained by SQUID measurements of the investigated Nd-Fe-B alloy before and after heat treatment. The coercive force Hc after optimal heat treatment (630°C, 5 min) is increased about three times. Significant increase in the content of the hard magnetic phase $Nd_2Fe_{14}B$ (about 85 mass%) after heat treatment effectively enhances coercivity.

Summary results of XRD and Mössbauer spectroscopic phase analysis together with magnetic measurements on VSM are presented in Table I.

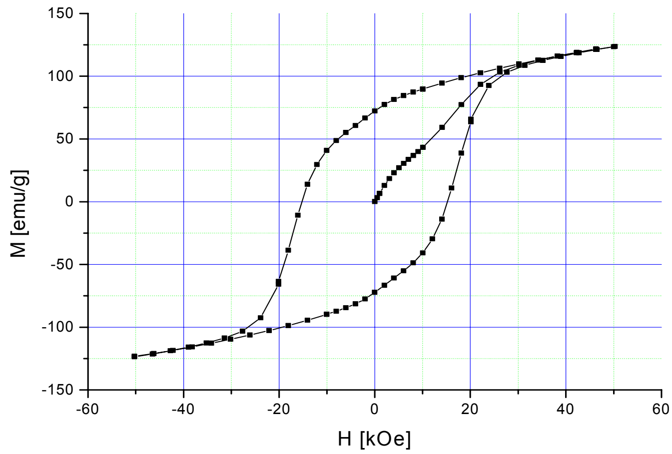
Tab. I Summary results of investigations of the $Nd_{14.5}Fe_{78.5}B_7$ alloy

Chemical composition mass%	Parameters of tetragonal crystal lattice of $Nd_2Fe_{14}B$		X-Ray analysis after heat treatment	Mossbauer phase analysis after heat treatment	Magnetic properties after heat treatment
	STANDARD VALUES	EXPERIMENTAL VALUES			
Nd 32.0 B 1.2 Pr 0.5 Al 0.3 Ti 0.0 Fe-balance	a=8.8052 c=12.2061	a=8.8050 c=12.2050	85% $Nd_2Fe_{14}B$ minimum	85% $Nd_2Fe_{14}B$ 6% $Nd_{1.1}Fe_{14}B$ 6%unidentified 3% α-Fe	Br 8.4kG Hci 13.5Oe Hcb 6.4kOe BH_{max} 12.6MGOe

The presented results of magnetic measurements obtained by a SQUID magnetometer (Fig 4b.) and VSM (Table I) are comparable.



a)



b)

Fig. 4 Hysteresis loops of $Nd_{14.5}Fe_{78.5}B_7$ alloy: a) as quenched, b) heat treated at 630°C for 5 min

Conclusion

The investigated melt-spun Nd rich alloy ($Nd_{14.5}Fe_{78.5}B_7$) with an almost $Nd_2Fe_{14}B$ monophase structure has shown good hard magnetic properties and not very high sensitivity to elevated temperatures and grain growth in the applied heat treatment regime. The basic hard magnetic phase $Nd_2Fe_{14}B$ has been identified as the main phase (85 mass%). Presence of the soft magnetic phase α-Fe and non-ferromagnetic phase $Nd_{1.1}Fe_4B_4$ are in minor quantities. These undesirable phases are most probably nanosized and therefore do not derogate the magnetic properties. The results of measurements of the relative change of magnetic permeability of the investigated $Nd_{14.5}Fe_{78.5}B_7$ alloy before and after annealing show that magnetic properties have changed as a consequence of refinement of the microstructure. This is comparable with the magnetic behavior of the investigated alloy after optimal heat treatment, which resulted in maximal values of magnetic properties.

Acknowledgement

This work has been supported by the Ministry of Science and Environment Protection of Serbia.

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Резюме: *Исследованный мелт-спун сплав $Nd_{14,5}Fe_{78,5}B_7$ с добавкой Nd, превышающей стехиометричиский состав $Nd_2Fe_{14}B$, получен методом быстрого охлаждения, оптимально отобранной скоростью охлаждения. Изучено влияние химического состава и режима термической обработки на магнитныие свойства. С целью толкования отношений микроструктура – магнитныие свойства экспериментально полученные результаты методами дифракции рентгеновских лучей и мессбауэровским фазовым анализом сопоставляли с результатами измерений магнитных свойств.*
Ключевые слова*: Фазовый состав, магнитные свойства, ДРЛ, мессбауэровская спектроскопия, SQUID.*

Садржај: *Истраживана мелт-спун $Nd_{14,5}Fe_{78,5}B_7$ легура са садржајем Nd изнад стехиометријског састава $Nd_2Fe_{14}B$, добијена је методом брзог хлађења, оптимално одабраном брзином хлађења. Размотрен је утицај хемијског састава и примењеног режима термичке обраде на магнетна својства. Експериментални резултати XRD*

анализе и Mössbauer спектроскопске фазне анализе корелисани су са измереним магнетним особинама у циљу добијања нових сазнања за тумачење релације микроструктура – магнетна својства.
Кључне речи: *Мелт-спун Nd-Fe-B, фазни састав, магнетна својства, XRD, Мњсбауерова спектроскопија, SQUID.*