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# PHASE COMPOSITION AND MAGNETIC PROPERTIES OF ANNEALED MELT-SPUN $Nd_{4.5}Fe_{77}B_{18.5}$ ALLOY

**Vladan R. Ćosović**
**Aleksandar S. Grujić**
**Jasna T. Stajić-Trošić**
**Nadežda M. Talijan**
**Institute of Chemistry, Technology and Metallurgy**
**Njegoševa 12, 11000 Belgrade**
**Serbia & Montenegro**

## ABSTRACT

*Phase transformations in the function of heat treatment regime for defined initial chemical composition of melt-spun* $Nd_{4.5}Fe_{77}B_{18.5}$ *ribbons were observed by X-Ray diffractometric analysis (XRD) and Mössbauer spectroscopy (MS). Magnetic properties of samples after applied heat treatment regime were measured on the vibrating sample magnetometer (VSM) with magnetic field strength of 50 kOe and on the SQUID magnetometer with magnetic field strength from -5 T up to 5 T. The heat treatment regime which provided nanocomposite structure* ($Fe_3B/Nd_2Fe_{14}B$) *of the investigated Nd-Fe-B alloy and obtained values of magnetic properties was presented and discussed.*

**Keywords:** nanocomposite, phase transformations, exchange coupling, magnetic properties

## 1. INTRODUCTION

Nanocomposite permanent magnetic materials based on Nd-Fe-B alloy with low Nd content are the new type of permanent magnetic materials [1–5]. The microstructure of these nanocomposite permanent magnets is composed of a mixture of magnetically soft and hard phases which provides so called exchange coupling interaction. Depending on the alloy composition, soft magnetic phases are one or both $\alpha$–Fe and $Fe_3B$, the hard magnetic phase is $Nd_2Fe_{14}B$. The rapid quenching technology is one of the major processing routes for the production of this type of permanent magnetic alloys. 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. Enhancement of magnetic properties of melt-spun Nd-Fe-B alloys with low Nd content after the heat treatment, according to most authors is the result of “exchange coupling” between neighboring grains of soft $Fe_3B$ and hard $Nd_2Fe_{14}B$ magnetic phases which gives a rise to remanence [6–9]. Necessary conditions for exchange coupling are that these phases should be crystalographically coherent and the mean grain size should be below 40 nm [8]. Therefore enhancement of remenence and thus the value of reduced remanence over the theoretical limit $Jr/Js = 0.5$ can be obtained by reducing the mean grain size. The effect becomes more pronounced with the decrease of the mean grain size which leads to the higher values of the energy product $(BH)_{max}$. Optimum dimension of the soft magnetic phase is considered to be twice the domain wall width of the hard magnetic phase $Nd_2Fe_{14}B$ ($\approx$ 5 nm). Theoretical considerations predict that very high values of maximum energy products may be achieved in the two phase exchange-coupled magnets [10].

The scope of this paper is investigation of the influence of annealing temperature on the magnetic properties of melt-spun Nd-Fe-B alloy with low Nd content. Thermal behavior is observed through phase transformations and the evolution of the microstructure during different regimes of heat treatment and parallel measurements of the magnetic properties. The goal is correlation of composition of the alloy, annealing temperature, microstructure and magnetic properties.

## 2. EXPERIMENTAL

Investigated alloy $Nd_{4.5}Fe_{77}B_{18.5}$ was produced by melt spinning method under argon atmosphere with optimal cooling rate, optimized in the previous investigations [11]. Influence of heat treatment on phase composition and magnetic properties of investigated melt-spun Nd-Fe-B alloy was studied in the temperature interval 600-700°C. Samples were annealed on 600, 660 and 700°C for 5 minutes. Phase composition of the investigated alloy after the heat treatment was observed by the XRD (X-Ray diffractometric analysis) and the Mössbauer spectroscopy (MS). For the spectra fitting and decomposition the CONFIT software package was used [12]. Identification of present phases was carried out by comparison of data obtained by the CONFIT package with the data found in literature [13]. Magnetic properties of heat treated samples were measured on the temperature of the ambient, on VSM (Vibrating Sample Magnetometer) with magnetic field strength of 50 kOe and on the SQUID magnetometer (Superconducting Quantum Interference Device) with magnetic field strength $\mu_0H$ from -5T up to 5T.

## 3. RESULTS AND DISCUSSION

Experimental results of XRD phase analysis and magnetic measurements on VSM after the different heat treatment regimes are presented in Table 1.

*Table 1. Experimental results of XRD phase analysis and magnetic measurements*

| Heat treatment regime | XRD | Magnetic properties (VSM) |
|---|---|---|
| 600°C / 5 min | $Nd_2Fe_{14}B$<br>$Nd_2Fe_{23}B_3$<br>$Fe_3B$<br>$Nd_{1.1}Fe_4B_4$<br>α - Fe | Br 12.0 kG<br>Hci 2.4 kOe<br>$(BH)_{max}$ 8.0 MGOe |
| 660°C / 5 min | $Fe_3B$<br>$Nd_2Fe_{14}B$<br>$Fe_{77.2}Nd_{22.8}$<br>α - Fe | Br 10.9 kG<br>Hci 2.8 kOe<br>$(BH)_{max}$ 10.7 MGOe |
| 700°C / 5 min | $Nd_2Fe_{14}B$<br>$Fe_3B$<br>$Nd_{1.1}Fe_4B_4$<br>α - Fe | Br 11.0 kG<br>Hci 2.7 kOe<br>$(BH)_{max}$ 8.8 MGOe |

Results of XRD phase analysis of the sample of the investigated alloy $Nd_{4.5}Fe_{77}B_{18.5}$ after the heat treatment at 600°C for 5 minutes show presence of $Fe_3B$, $Nd_2Fe_{23}B_3$ and α-Fe phases as well as minor quantities of $Nd_2Fe_{14}B$ and boride phase $Nd_{1.1}Fe_4B_4$. High value of remanence (12.0 kG) measured on VSM can be explained by large amount of soft magnetic phases: $Fe_3B$ and α-Fe. X-Ray diffractogram of the investigated alloy $Nd_{4.5}Fe_{77}B_{18.5}$ after annealing at 660°C for 5 min is presented on Figure 1.

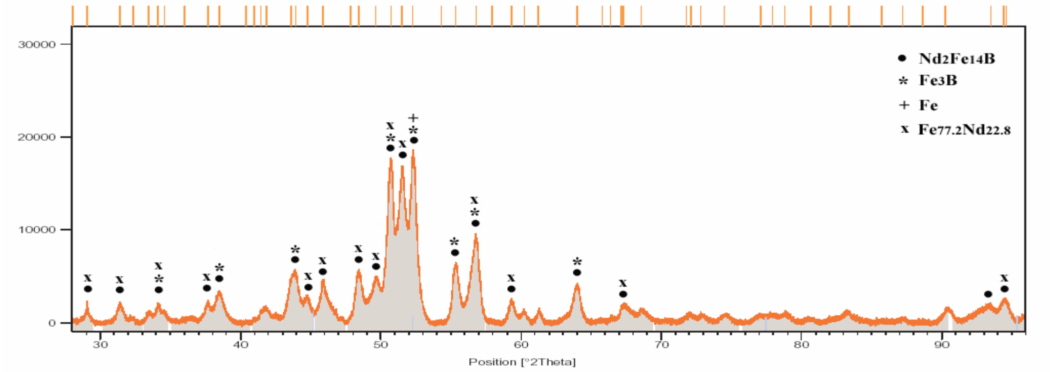

*Figure 1. X-Ray diffractogram of the investigated $Nd_{4.5}Fe_{77}B_{18.5}$ alloy after annealing at 660°C/5 min*

After annealing at 660°C for 5 minutes, as shown on X-Ray diffractogram on Figure 1, $Fe_3B$, $Nd_2Fe_{14}B$, $Fe_{77.2}Nd_{22.8}$ and α-Fe phases were identified. This sample possesses the highest coercivity due to formation of hard magnetic phase $Nd_2Fe_{14}B$. Intensities of corresponding diffraction peaks suggest that $Nd_2Fe_{23}B_3$ and α-Fe are present in traces. Metastable phase $Nd_2Fe_{23}B_3$ which was present in the first sample crystallizes as an intermedial phase and with the increase of the temperature of heat treatment it decomposes into soft magnetic phase $Fe_3B$, main hard magnetic phase $Nd_2Fe_{14}B$ and smaller amount of the soft magnetic phase α-Fe. Increase of the amount of the hard magnetic phase $Nd_2Fe_{14}B$ in alloy with the increase of the annealing temperature from 600°C to 660°C led to the higher coercivity (from 2.4 kOe on 600°C to 2.8 kOe on 660°C) and finally to the higher energy product $(BH)_{max}$ (from 8.0 MGOe on 600°C to 10.7 MGOe on 660°C).

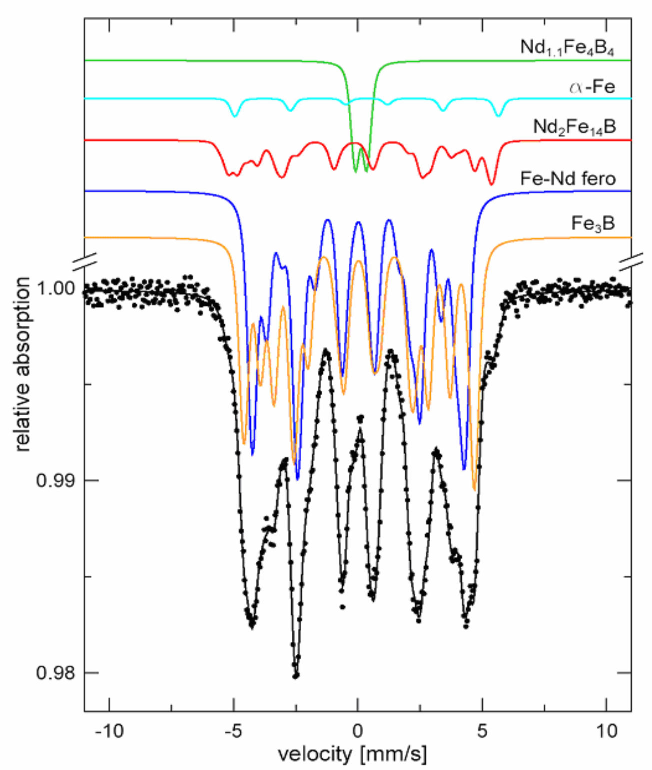

*Figure 2. Mössbauer spectra of investigated $Nd_{4.5}Fe_{77}B_{18.5}$ alloy after annealing at 660°C/5 min*

For the better insight into the phase composition of the investigated alloy after the heat treatment $^{57}Fe$ Mössbauer spectroscopic phase analysis was carried out. MS analysis provided identification and calculation of individual phase contents in the alloy. Analysis of the MS spectra of investigated $Nd_{4.5}Fe_{77}B_{18.5}$ alloy after annealing at 660°C for 5 minutes (Figure 2.) confirmed presence of the $Fe_3B$, $Nd_2Fe_{14}B$ and $Nd_{1.1}Fe_4B_4$ phases and the whole set of Fe-Nd ferromagnetic phases. Since the sample annealed at 660°C for 5 minutes has the most favorable magnetic properties, the sample was further investigated on SQUID magnetometer. Hysteresis loop of $Nd_{4.5}Fe_{77}B_{18.5}$ alloy after annealing at 660°C/5 minutes obtained by SQUID magnetometer is presented on Figure 3.

Remanence ratio Jr/Js=0.6 calculated from the hysteresis loop obtained by the SQUID magnetometer after the heat treatment at 660°C for 5 minutes suggests that the nanocomposite $Fe_3B/Nd_2Fe_{14}B$ was formed and that the magnetic properties of investigated alloy are directly influenced by the interaction of exchange coupling between the grains of the soft magnetic phase $Fe_3B$ and the grains of hard magnetic phase $Nd_2Fe_{14}B$. It can be assumed that this mechanism has influenced the enhancement of $(BH)_{max}$. Decrease of $(BH)_{max}$ after annealing at 700°C for 5 min is due to increased content of soft magnetic α-Fe phase as well as other identified and unidentified soft magnetic phases compared to content of the hard magnetic phase $Nd_2Fe_{14}B$. Considerable value of coercive force is consequence of still significant content of the $Nd_2Fe_{14}B$ phase. Remanences of the samples of investigated alloy with low Nd content after heat treatment on investigated temperatures (600°C; 660°C and 700°C) are significant (12.0; 10.9; and 11.0 kG), which has a direct influence on the obtained values of the magnetic energy $(BH)_{max}$ (8.0; 10.7 and 8.8 MGOe).

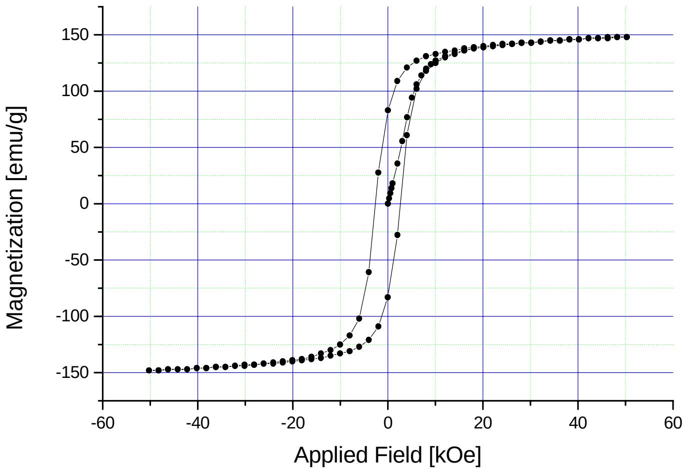

*Figure 3. Hysteresis loop of* $Nd_{4.5}Fe_{77}B_{18.5}$ *alloy after annealing at 660°C/5 min.*

Differences in obtained values of magnetic properties can be explained by the fact that those two mechanisms: exchange coupling and phase content govern the coercivity and remanence. While exchange coupling influences both coercivity and remenece, the content of the soft magnetic phase competes with the exchange interaction in its effect on remanence [14]. Presence of identified boride phase $Nd_{1.1}Fe_4B_4$ in all samples after annealing, although in small amounts, is consequence of high boron content, above 4.2 at% [15]. Measured values of magnetic properties suggest that this phase is probably formed in nanosize and that it has negligible influence on the magnetic properties of the investigated Nd-Fe-B alloy.

## 4. CONCLUSION

Optimal heat treatment regime was selected by correlation of phase composition in the function of heat treatment and magnetic properties. Based on high content of soft magnetic phase $Fe_3B$, presence of hard magnetic phase $Nd_2Fe_{14}B$ and comparison of measured magnetic properties it can be assumed that nanocomposite $Fe_3B/Nd_2Fe_{14}B$ was formed after annealing of the $Nd_{4.5}Fe_{77}B_{18.5}$ alloy at 660°C for 5 minutes. Calculated value of reduced remanence Jr/Js = 0.6 is higher that theoretical limit (0.5) which suggests that exchange coupling between grains of soft $Fe_3B$ and hard magnetic $Nd_2Fe_{14}B$ phases, identified by the phase analysis, had the direct influence on the improvement of magnetic properties of investigated alloy.

## 5. REFERENCES

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## 6. ACKNOWLEDGEMENT

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