Proceedings of the CSMAG'07 Conference, Košice, July 9–12, 2007

Thermomagnetic Behaviour and Microstructure of a Rapidly Quenched $Nd_{4.5}Fe_{77}B_{18.5}$ Alloy

N. Talijan, V. Ćosović, A. Grujić, J. Stajić-Trošić
Institute of Chemistry, Technology and Metallurgy, University of Belgrade
Njegoševa 12, 11000 Belgrade, Serbia

and T. Žák
Institute of Physics of Materials, Academy of Sciences of the Czech Rep., v.v.i.
Žižkova 22, 616 62 Brno, Czech Republic

Changes in the phase composition and crystallite size, as well as changes in the magnetic behaviour of $Nd_{4.5}Fe_{77}B_{18.5}$ alloy, caused by thermomagnetic measurements, was observed in regard of the optimal magnetic state of this alloy. In the optimized magnetic state, the formed nanocomposite consisted of $Fe_3B/Nd_2Fe_{14}B$ and partly of α-Fe with a mean crystallite size < 30 nm, as determined by X-ray diffraction and transmission electron microscopy. An increased amount of α-Fe, the presence of Nd_2O_3 and different Fe–B phases, as well as an increase in the mean crystallite size were observed after thermomagnetic measurements had caused a quality loss of the hard magnetic properties.

PACS numbers: 81.40.Rs

1. Introduction

The microstructure of Nd–Fe–B alloys with a low content of Nd consists of a hard magnetic phase: $Nd_2Fe_{14}B$, soft magnetic phases: α-Fe and/or Fe_3B phase and a variety of Fe–B-type phases. The main condition for obtaining a nano-composite structure is the homogeneous dispersion of a hard phase in a soft phase with a mean grain size in the nano scale (< 40 nm), because intergranullar coupling between the phases only becomes more pronounced in the nano scale [1]. The formed nanocomposites $Fe_3B/Nd_2Fe_{14}B$ and/or α-$Fe/Nd_2Fe_{14}B$ are directly responsible for an increase in remanence and magnetic energy. In this work, some of the results of size-strain analysis and quantitative phase analysis obtained by X-ray diffraction (XRD) of $Nd_{4.5}Fe_{77}B_{18.5}$ alloy in the optimized magnetic state and after thermomagnetic measurements (TM) and the corresponding magnetic behaviour are reported and discussed.

2. Experimental

The $Nd_{4.5}Fe_{77}B_{18.5}$ alloy was produced by centrifugal atomization and its magnetic characteristics in the optimized magnetic state were $H_c = 2.8$ kOe,

B_r = 10.9 kGs and $(BH)_{max}$ = 10.7 MGOe. TM in the temperature interval 20–800°C were performed using a vibrating sample magnetometer in a field of intensity 50 Oe under vacuum. The phase composition and crystallite size in the optimized magnetic state and after TM were determined by XRD analysis using an X'Pert PRO MPD diffractometer from PANanalytical with Co K_α radiation. The size-strain analysis and quantitative phase analysis of the obtained XRD data were realised using FullProf software [2]. The X-ray line broadenings were analysed through refinement of the TCH-pV function parameters. The microstructure of the alloy in the optimized state was observed by transmission electron microscopy (TEM). Magnetic properties before and after TM were measured previously using SQUID magnetometer with a magnetic field strength of 50 kOe [3].

3. Results and discussion

The thermomagnetic behaviour of $Nd_{4.5}Fe_{77}B_{18.5}$ alloy and the corresponding phase transformations were observed by TM. The obtained TM curves are presented in Fig. 1.

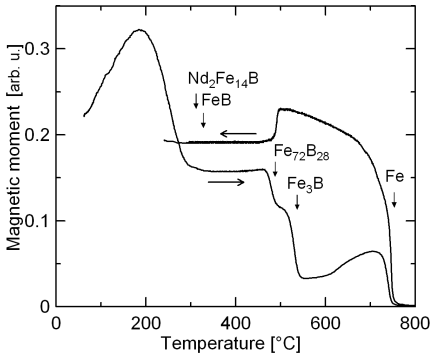

Fig. 1. TM curves of the investigated $Nd_{4.5}Fe_{77}B_{18.5}$ alloy.

To obtain a better understanding of the influence of phase composition and crystallite size of the identified phases on the magnetic properties before and after TM, size-strain and quantitative phase analyses of the XRD data were performed. Comparisons between observed and calculated intensities before and after TM are presented in Fig. 2. The vertical bars indicate the positions of the reflections and the difference patterns are given below. The amount of the hard and soft magnetic phases and their crystallite sizes, calculated by Full-Prof quantitative phase analysis and size-strain analysis of the XRD data are presented in Table.

In the optimised state of the alloy, Fe_3B, $Nd_2Fe_{14}B$, $Fe_{77.2}Nd_{22.8}$ and α-Fe phases were identified by XRD analysis. It also suggests that the presence of $Nd_2Fe_{23}B_3$ and α-Fe could not be neglected [3, 4]. The presence of a $Fe_{77.2}Nd_{22.8}$ phase is to be understood more as a representative of Fe(Nd,B) components. After the TM according to the XRD results (Fig. 2 and Table), the main decomposition

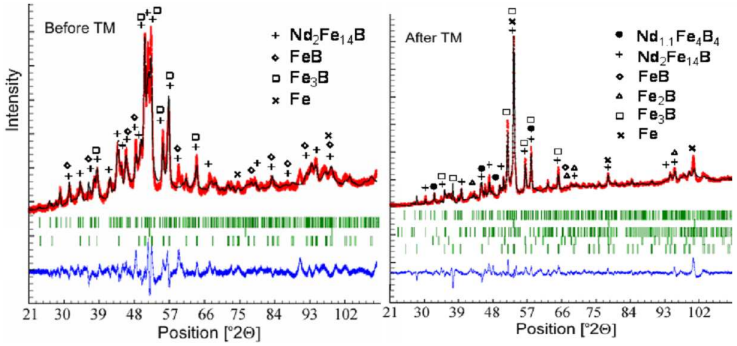

Fig. 2. XRD diffractograms with comparison of observed and calculated intensities.

TABLE

Mean crystallite sizes of phases in the $Nd_{4.5}Fe_{77}B_{18.5}$ alloy before and after TM.

Phase	Before TM		After TM	
	Amount [%]	Crystallite size [nm]	Amount [%]	Crystallite size [nm]
$Nd_2Fe_{14}B$	43.6	12	18.0	38
α-Fe	16.6	5	36.8	29
Fe_3B	39.8	24	28.2	34
Nd_2O_3	–	–	8.0	8
Fe(O)B	–	–	9.0	11

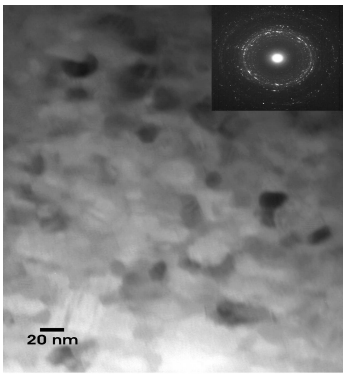

Fig. 3. Bright field TEM micrograph of $Nd_{4.5}Fe_{77}B_{18.5}$ alloy in the optimised state.

product is α-Fe. This phase is accompanied by an $Nd_2Fe_{14}B$ phase, Fe_3B, and a number of different Fe–B phases, as well as a boride phase $Nd_{1.1}Fe_4B_4$. The amount of soft magnetic phases had increased (mainly α-Fe) and the crystallite size of all identified phases had also increased. The presence of the Nd_2O_3 phase was confirmed, too. Practically, this all had a direct influence on the deterioration

of the magnetic properties. To clarify further the phase constitution, Mössbauer measurements are in progress.

The TEM micrograph (Fig. 3.), showing the average grain size in the optimized magnetic state below 30 nm, confirms the mean crystallite size calculated from the XRD data. Thus, on average, one grain is composed of one crystallite, i.e., all crystalline phases are well crystallized. A microdiffraction analysis gave evidence for the mixing of the nanocrystalline phases. This implies that the alloy has a nanocomposite structure of $Fe_3B/Nd_2Fe_{14}B$ and partly α-Fe. The phase transformations which occurred during TM are in correspondence with magnetic behaviour as measured by SQUID.

4. Conclusion

Both the mean crystallite size and grain size of the investigated alloy in optimal magnetic state determined by XRD and TEM analysis were below 30 nm indicating that in average one grain is composed of one crystallite i.e. all crystalline phases were well crystallized. Composed of $Nd_2Fe_{14}B$, Fe_3B and α-Fe phases, the alloy in the optimized magnetic state has mainly a nanocomposite structure. The value of the remanent ratio $J_r/J_s > 0.5$, calculated from the SQUID hysteresis loops [3], suggests that exchange coupling interactions between grains of the soft and hard magnetic phase exist. This assumption is supported by the higher value of the remanence and $(BH)_{max}$, which are typical of nano-composite structures of Nd–Fe–B alloys.

The increase in temperature during TM resulted in the growth of the crystallites and a decrease in the amount of $Nd_2Fe_{14}B$, due to an increase in the amounts of the α-Fe and Nd-oxide fractions, in respect of the optimized magnetic state.

In the light of these facts, it can be concluded that the structural and microstructural changes (phase compositions, crystallite size) have a direct influence on magnetic behaviour and they are the main reason for the deterioration of magnetic properties after TM.

Acknowledgments

This work was supported by the Serbian Ministry of Science, project OI 142035B and by the Czech Ministry of Education, Youth and Sports, project 1M0512.

References

[1] C.J. Yang, E.B. Park, *J. Magn. Magn. Mater.* **166**, 243 (1997).

[2] J. Rodriguez-Carvajal, 1998 FullProf computer program; ftp://charybde.saclay.cea.fr/pub/divers/fullprof.98/windows/winfp98.zip..

[3] V. Ćosović, T. Žák, N. Talijan, A. Grujić, J. Stajić-Trošić, *J. Alloys Comp.*, doi:10.1016/j.jallcom.2007.02.018; in press.

[4] V. Ćosović, A. Grujić, J. Stajić-Trošić, V. Spasojević, N. Talijan, *Mater. Sci. Forum* **555**, 527 (2007).