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MAGNETIC PROPERTIES OF THE NANOCRYSTALLINE $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ ALLOY RIBBONS

WŁAŚCIWOŚCI MAGNETYCZNE NANOKRYSTALICZNYCH TAŚM STOPU $Nd_{9,6}Fe_{64,32}Nb_4B_{22,08}$

Studies of magnetic properties and phase constitution of nanocrystalline $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbon samples are presented. In as-cast state, ribbon samples were fully amorphous and soft magnetic. Subsequent annealing resulted in an evolution of the phase constitution together with change of their magnetic properties. The main phase observed in the material is $Nd_2Fe_{14}B$ hard magnetic phase.
Keywords: X-ray diffractometry, magnetic properties NdFeB alloys, $Nd_2Fe_{14}B$, nanocrystalline ribbon

W pracy badano własności magnetyczne oraz skład fazowy taśm wytworzonych ze stopu $Nd_{9,6}Fe_{64,32}Nb_4B_{22,08}$. Taśma w stanie po odlaniu ma amorficzną budowę i wykazuje miękkie właściwości magnetyczne. Wygrzewanie taśm powoduje zmiany składu fazowego wraz ze zmianą ich właściwości magnetycznych. Podstawową fazą obserwowaną w badanym materiale jest faza magnetycznie twarda $Nd_2Fe_{14}B$.

1. Introduction

In the 80's of last century, the new generation of Nd-Fe-B permanent magnets, which exhibit excellent magnetic properties, were discovered [1]. Since than a numerous works were done in order to refine their microstructure and phase constitution, that resulted in refinement of magnetic properties leading to improvement of their coercivity field $_JH_c$, remanence B_r and maximum energy product $(BH)_{max}$. Especially, manufacturing technology plays important role in the microstructure and magnetic properties. The Nd-Fe-B magnets are usually produced by sintering [2], mechanical alloying [3] or HDDR (hydrogenation, disproportionation, desorption and recombination) [4]. Currently, the frequently used methods of processing the magnets is the melt-spinning of induction molten alloy into a ribbon under controlled atmosphere of an Ar. To obtain optimal magnetic properties, selection of the alloy composition as well as suitable heat treatment, are crucial [5].

In recent years, the bulk rapidly solidified nanocrystalline alloys, were produced in a form of amorphous rods, tubes or plates. These samples were manufactured for the doped Nd-Fe-B and Pr-Fe-B alloys, with a high boron content [6]. Interesting magnets produced from base Nd-Fe-B alloy doped with 4 at% of Nb, were studied in [7,8]. Samples studied by these authors were produced in a form of rods and annealed at one temperature T = 993K. Therefore, the aim of the present work was to investigate the $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy in a form of melt-spun ribbon annealed at various temperatures in order to optimize the magnetic properties. It was shown, that large content of B promotes the glass forming ability (GFA) of alloys. Also addition of Nb greatly improves the GFA and retards growth of grains formed during annealing [8]. Many other research are carried out for amorphous alloys containing Nb [9]. An important factor is the heat treatment process carried out to obtain optimum magnetic properties of magnets. For this reason it is important to determine the effect of annealing conditions on the phase constitution and magnetic properties of nanocrystalline ribbons produced from the $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy.

2. Experimental

A base alloy of nominal compositions $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ was prepared by arc melting the high purity elements with pre-alloyed Fe-B of known composition under an Ar atmosphere. Then ribbon samples were produced by single roll melt-spinning tech-

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nique under the Ar atmosphere at linear speed of the copper roll surface of 35m/s. Subsequently the ribbon samples were sealed off in a quartz tube under low pressure of argon to maintain the purity of atmosphere during heat treatment. In order to modify microstructure and magnetic properties, samples were annealed at various temperatures from 923K to 1023K for 5 min and quenched in water. The phase analysis of the samples was carried out using X-ray diffractometry (XRD) with Co Kα radiation. Room temperature hysteresis loops were measured by LakeShore 7307 vibrating sample magnetometer at external magnetic field up to 2T.

3. Results and discussion

The XRD patterns measured for the as-cast $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbons and for samples annealed at 923K and 943K for 5 min, are shown in Fig. 1.

Lack of peaks corresponding to crystalline phases suggests an amorphous structure of as-cast ribbon. Annealing at 923K and 943K for 5 minutes results in small changes in the phase constitution of the material. Broadened peaks corresponding to the crystalline phase are shown in the diffraction patterns. However, small intensity peaks compared to the width of the background does not allow clear identification of crystalline phases present in the samples.

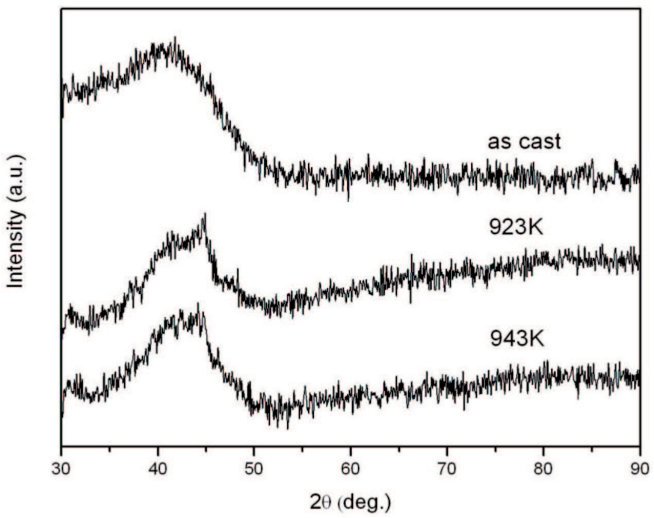

Fig. 1. X-ray diffraction patterns of as-cast $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbon sample and samples annealed at 923K and 943K for 5 min

The XRD patterns measured for the $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbon samples annealed at 963K, 983K, 1003K and 1023K for 5 min, are shown in Fig. 2.

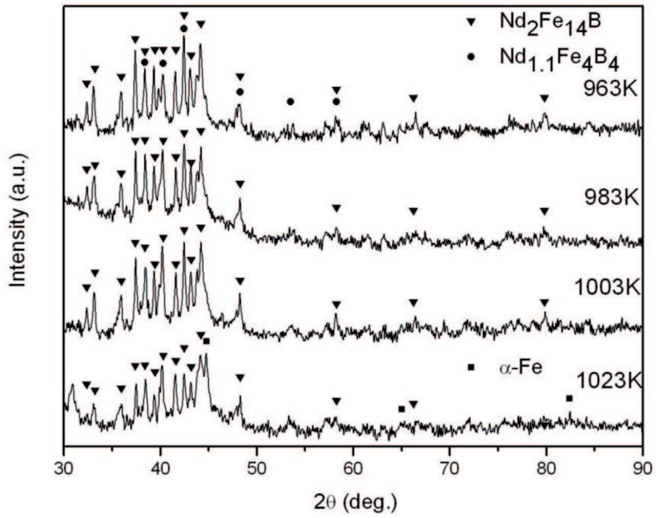

Fig. 2. X-ray diffraction patterns of $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbon samples annealed at 963K, 983K, 1003K and 1023K for 5 min

Short-time annealing of samples (for 5 min) at 963K and higher temperatures, led to nucleation and growth of the crystalline phases. The main crystalline phase observed in the investigated material is the hard magnetic $Nd_2Fe_{14}B$ phase. The phase analysis indicates also a presence of the $Nd_{1.1}Fe_4B_4$ paramagnetic phase. However, due to the proximity of the peaks from the $Nd_2Fe_{14}B$ phase, the presence of $Nd_{1.1}Fe_4B_4$ phase should be confirmed by other methods. For samples annealed at and higher temperatures than 963K, no change in phase constitution, was observed. Also the peak intensities corresponding to the crystalline phases are not significantly changed. However, annealing at 1023K for 5 min results in further changes in phase composition of the samples, where except the dominant $Nd_2Fe_{14}B$ hard magnetic phase, diffraction peaks from soft magnetic α-Fe phase are observed.

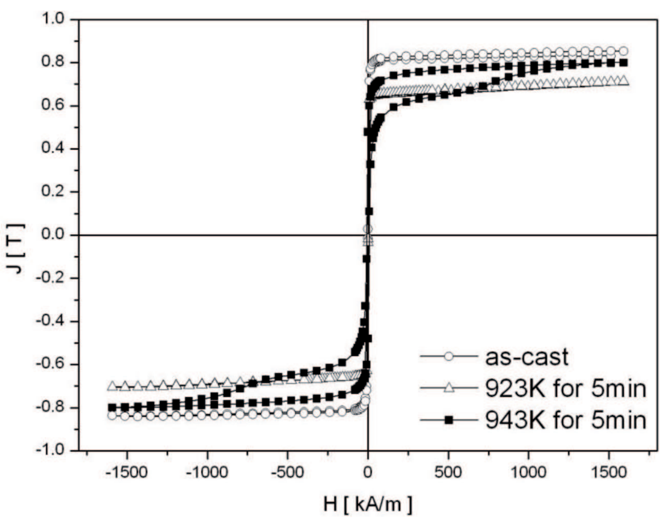

Fig. 3. The hysteresis loops measured for the as-cast $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ and alloy ribbon samples annealed at 923K and 943K for 5 min

In Fig. 3 the hysteresis loops of ribbon in as-cast state and after annealing at 923K and 943K for 5 min, are shown. Soft magnetic properties of as-cast ribbons confirm their amorphous microstructure. Annealing at 923K and 943K results in a change in shape of the hysteresis loops, due to a small fraction of crystalline phases.

The hysteresis loops measured for annealed ribbon samples in the temperature range from 963K to 1023K for 5 min, are shown in Fig. 4.

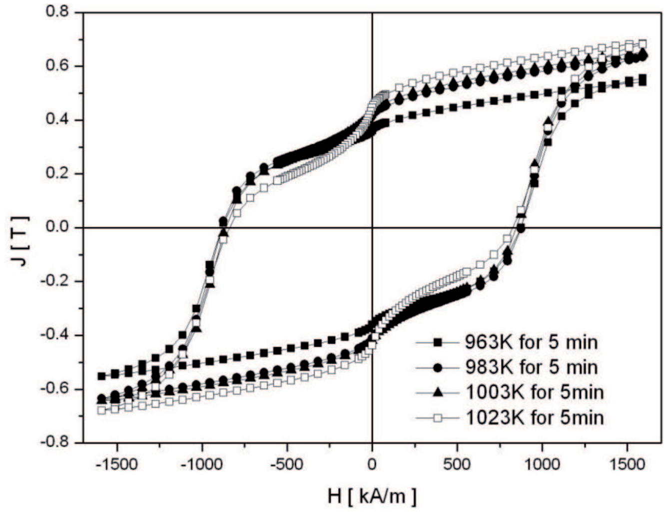

Fig. 4. The hysteresis loops measured for $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbon samples annealed at 963K, 983K, 1003K and 1023K for 5 min

Magnetic hysteresis loops measured for these samples are characteristic for hard magnetic materials. With the increase of annealing conditions a change of remanence, while almost constant coercitivity, were observed. Changes in the shape of magnetic hysteresis loops are related to different microstructure of samples annealed at various temperatures. Significant differences in demagnetizing curve of ribbon annealed at 1023K for 5 min are related to different phase constitution of this samples. Magnetic properties of the investigated materials are collected in Table 1.

TABLE 1
Magnetic properties of $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbon samples annealed at 963K, 983K, 1003K and 1023K for 5 min

	$_JH_c$ *(kA/m)*	J_r *(T)*	BH_{max} *(kJ/m³)*
963K	880±9	0,36±0,01	94±2
983K	881±9	0,41±0,01	99±2
1003K	851±9	0,43±0,01	92±2
1023K	833±9	0,44±0,01	96±2

The maximum value of coercitivity $_JH_c$ = 881kA/m was reached for the ribbon annealed at 983K for 5 min. It is lower than this given by Zhang et al. [8], which was ~1100kA/m. The polarization remanence J_r gradually increases with the increase of the annealing temperature. However, the values of the maximum magnetic energy product is about three times higher than those given by Zhang [8] for rods of the same alloy composition and comparable with the $(BH)_{max}$ obtained by Tamura [7] for $Nd_9Fe_{73}B_{14}Nb_4$, $Nd_9Fe_{77}B_{10}Nb_4$ alloys composition. Values of $(BH)_{max}$ are the same for all samples annealed at temperatures higher than 963K.

4. Conclusion

In the present work, the phase constitution and magnetic properties of the $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbons annealed at various temperatures from 923K to 1023K for 5 min, were shown.

It was found that the $Nd_{9.6}Fe_{64.32}Nb_4B_{22.08}$ alloy ribbon, produced by free jet melt-spinning technique had fully amorphous structure and soft magnetic properties. Heat treatment of these ribbons led to nucleation and growth of the $Nd_2Fe_{14}B$ hard magnetic phase, however, additional studies should be performed in order to confirm the presence of the $Nd_{1.1}Fe_4B_4$ paramagnetic phase. Furthermore, the X-ray diffraction patterns obtained for ribbons annealed at 1023K shown presence of additional peaks originating from the soft magnetic α-Fe phase. It was shown that the heat treatment leads to nanocrystalline structure of ribbon samples and good magnetic properties.

Acknowledgements

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