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The influence of densification temperature and the size of powder particles on the properties of isotropic Nd–Fe–B magnets made of quenched ribbons

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The influence of densification temperature and the size of powder particles on magnetization of remanence J_r, coercivity ${}_JH_c$ of hot-densified Nd–Fe–B magnets made of MQP-A quenched ribbons was investigated. A connection between magnetic properties, parameters of production process and microstructure of the magnet was confirmed. It was found that the remanence of isotropic magnet depends on temperature of hot densification process but is practically independent of the size of powder particles. On the other hand, the temperature of hot densification process and the size of powder particles have a substantial influence on coercivity.

Keywords: *Nd–Fe–B magnets, hot densification process, powder particle size, microstructure, quenched ribbons*

1. Introduction

Hot-densified isotropic Nd–Fe–B magnets made of quenched ribbons are semi-finished products in the production process of anisotropic magnets in which the texture is induced by hot plastic deformation [1–9]. A perfect preparation of isotropic precursor is a necessary condition to obtain a high quality anisotropic magnet. However, it is difficult to find any information in literature concerning the influence of such basic technical parameters as the size of powder particles or densification temperature applied in the production process of isotropic magnets on their properties.

Raja K. *et al.* [10] examined the influence of time of heat treatment of Nd–Fe–B isotropic magnets at 800 °C on their microstructure and magnetic properties but they did not examine the influence of densification temperature on microstructure and magnetic properties of the magnets.

Another group of researchers [11] studied the influence of the size of powder particles on the anisotropic properties of Nd–Fe–B magnets produced by single–stage hot deformation but they did not recognize the influence of this parameter on the properties of isotropic magnets assigned for two–step hot processing to produce anisotropic magnets.

This work aims at investigating the mentioned questions. It has been presumed that the properties of isotropic precursors have a significant impact on the properties of the final product – anisotropic die–upset magnets.

2. Experimental

To show the influence of densification temperature and the size of powder particles on the microstructure and magnetic properties of magnet material, various densification temperatures and sizes of powder particles were applied. To produce samples of the magnets, a commercial MQP-A powder (by Magnequench) with the chemical composition $Nd_{16}Fe_{78}B_6$ was used as a raw material. The powder was comminuted in a hand mortar and fractionated using sieves. These operations were carried out in a glove box in the presence of argon of 99.999 % purity. For densification, a 4 g portion of the powder was placed in a graphite mold and densified in following conditions (the same for all samples): vacuum 10^{-6} hPa, pressure 100 MPa and time 10 s. Such a short time of densification process was applied to reduce the grain growth. It was the shortest

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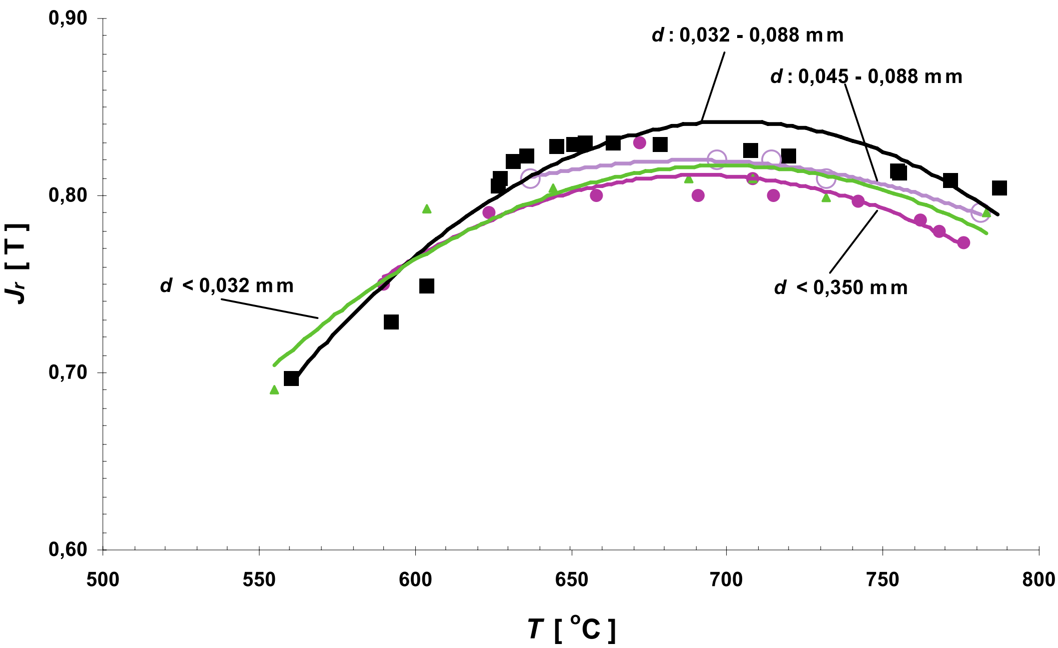

Fig. 1. Influence of temperature of powder densification on remanence of isotropic magnets made of fractions with different particle sizes d: < 32; 32–88; 45–88; < 350 μm.

time available for the used fabrication equipment. During the process the densification temperature was measured. As a result of the production process the isotropic magnets with density of 91–99.6 % of theoretical value were obtained. The density was measured with the use of Archimedes method. Magnetic properties were examined using a VSM, after magnetization in a magnetic field of 5 T. The content of oxygen in the material was determined using a high–temperature extraction method. Images of microstructure were taken with a SEM microscope on the fractures made along the direction parallel to the press force during densification.

3. Results and discussion

Fig. 1 presents the influence of powder densification temperature on remanence of isotropic magnets made of the fractions with different sizes of powder particles d: < 32; 32–88; 45–88; < 350 μm. The initial increase of remanence with growing densification temperature results from the increase in material density (Fig. 2). In the SEM images of the fractures of the compacts densified at relatively low temperature (603 °C) there are visible powder particles with their boundaries and void spaces between them (Fig. 3a). Such features of material structure result from densification without participation of liquid phase which occurs at temperatures above 640 °C [12]. The absence of a liquid phase causes that the material is not plastic enough to fill the void spaces between the particles. In the material densified at the temperatures of 719 °C and 789 °C (Figs. 3b–3c) there are no void spaces. Above 650 °C, the remanence is practically constant until about 720 °C for all fractions of the powder. A small decrease of remanence above 720 °C is probably caused by the effect of coercivity drop, connected with recrystallization of microstructure. Generally, it can be said that the size of powder particles has no significant influence on the magnetization of remanence.

The dependence of coercivity of isotropic magnets on densification temperature of powders with different particle sizes d: < 32; 32–88, 45–88, < 350 μm is shown in Fig. 4. The coercivity of the magnets decreases with the increase of densification temperature. The reason of this phenomenon is the increasing content of big grains in the bulk material with increasing temperature. This increase concerns

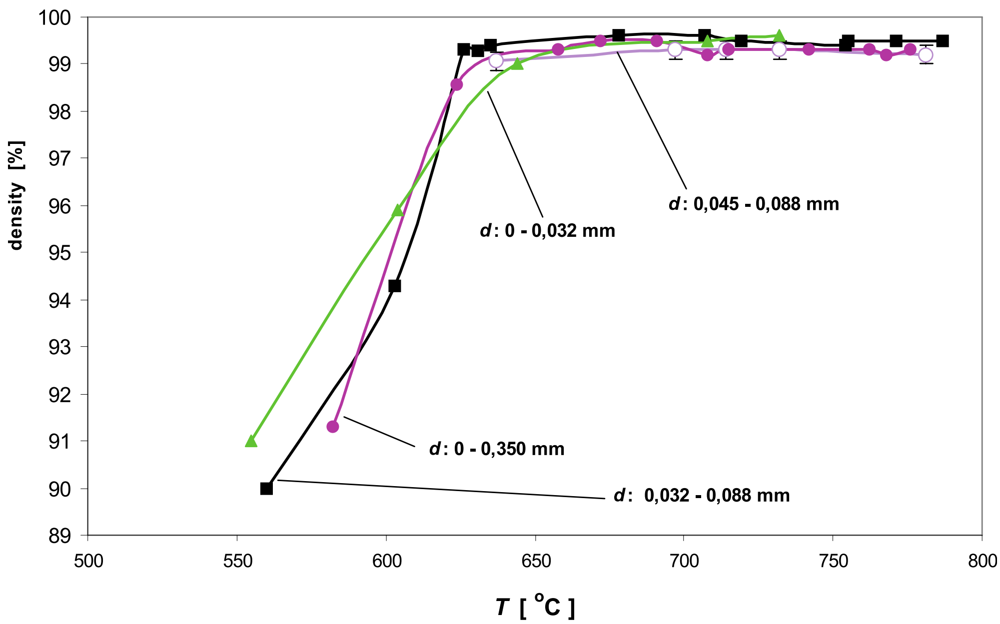

Fig. 2. Influence of temperature of powder densification on density of isotropic magnets made of fractions with different particle size d: < 32; 32–88; 45–88; < 350 μm. Density related to theoretical value (7.6 g/cm^3).

mainly the grains located on the boundaries of the powder particles (Figs. 3a–3c). At densification temperature which is close to the liquid phase transition, the fracture surfaces have clusters of big grains in a form of streaks. The sizes of streaks of the material densified at the temperature of 719 °C (Fig. 3b) are visibly smaller than those of the material densified at the temperature of 789 °C (Fig. 3c). Big grains occur also in the microstructure of the material densified at the temperature of 603 °C (Fig. 3a), but the participation of these grains in the whole bulk material is smaller, because they do not form large clusters (streaks). The smallest values of coercivity are characteristic of the samples with the powder fraction < 32 μm. This effect results from a disadvantageous relation of the surface area to the material volume for this fraction. When the material is comminuted, a free surface increases. During high temperature processing the free surface becomes the center of grain recrystallization because it is the area with higher energy in relation to the interior. The highest coercivities were shown by the samples obtained from fractions 32–88 μm and 45–88 μm. Both these fractions showed no special differences, except a slightly faster drop of coercivity for the fraction 45–88 μm at the temperature above 720 °C. The fraction < 350 μm produced magnets with intermediate coercivity in comparison to the other ones. Such effect is related to low participation of the smallest particles in this fraction, which have negative influence on coercivity.

The increase in the temperature of powder densification leads to a drop of oxygen content in the alloy (Fig. 5). This unexpected effect was obtained for the powder fractions with particle sizes d: < 32; 32–88; < 350 μm. This was caused by the disappearance of pores, which were sources of oxygen up to 650 °C. Above the temperature in which full densification is obtained, the content of oxygen is constant. It means that the decrease in coercivity with increasing densification temperature is not related to oxidation.

The factor which increases oxygen content in material is its comminution (Fig. 5). The highest amount of oxygen was found in the samples obtained from fractions with the smallest particles d < 32 μm. These samples showed also the smallest coercivity. This means that oxidation, apart from recrystalization phenomenon, is another factor decreasing coercivity.

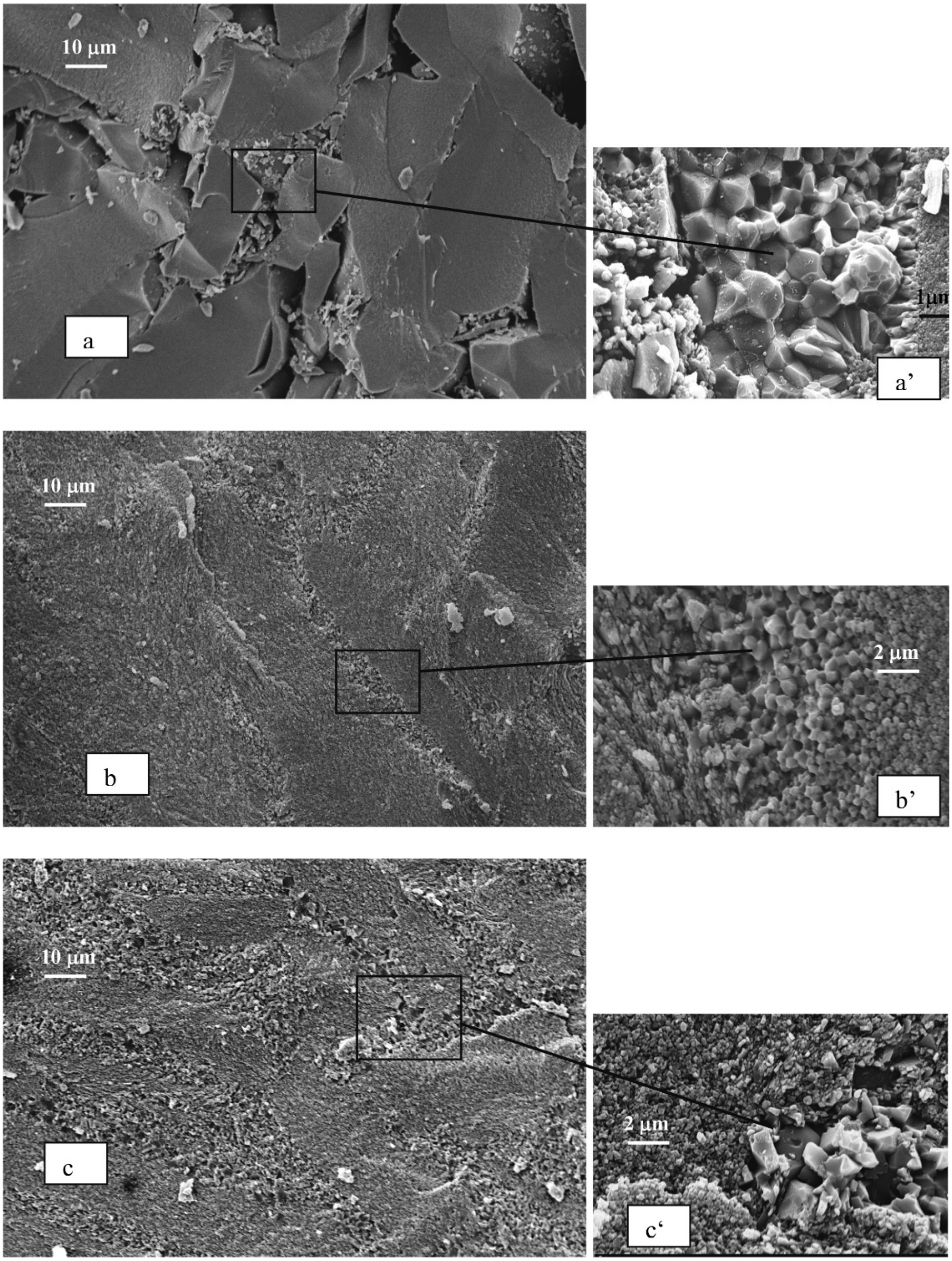

Fig. 3. Characteristic features of material structure depending on densification temperature: a) 603 °C; b) 719 °C; c) 789 °C, for powder fraction with particle size d: 32–88 μm.

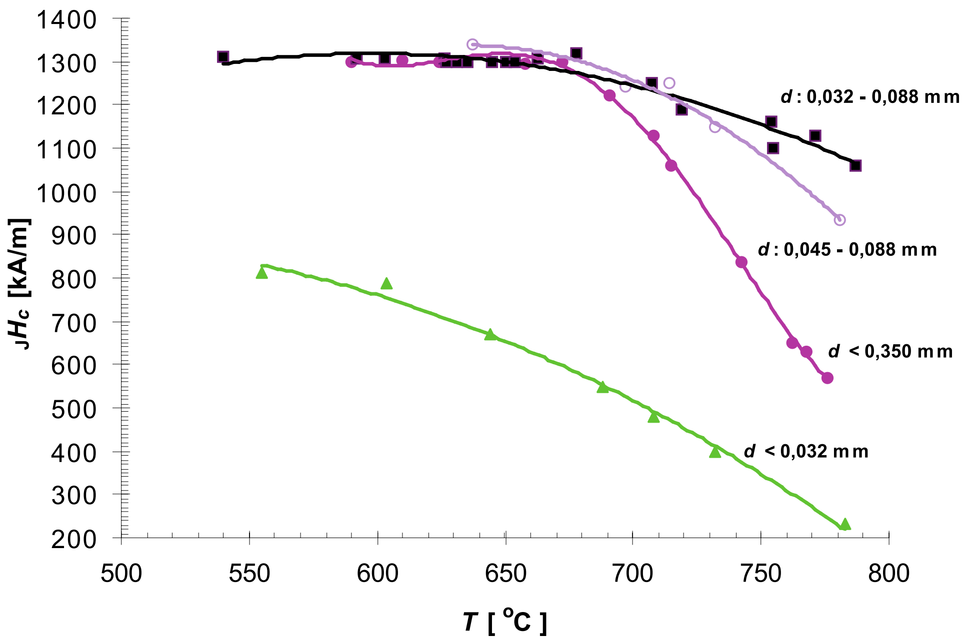

Fig. 4. Dependence of coercivity of isotropic magnets on densification temperature of powders for different particle sizes d: < 32; 32–88; 45–88; < 350 μm.

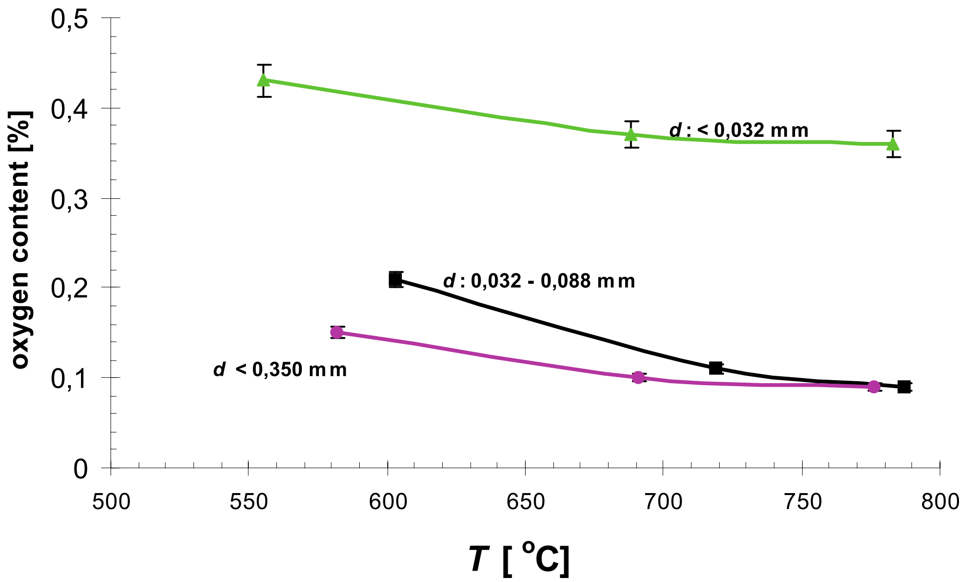

Fig. 5. Dependence of oxygen content in material of isotropic magnets on densification temperature of powders with different particle sizes d: < 32; 32–88; 45–88; < 350 μm.

4. Conclusions

1. The optimum densification temperature for MQP–A powder is about 650 °C. At this temperature the material reaches a density close to theoretical. At lower temperature it is not possible to obtain a required density and worse remanence is obtained. On the other hand, higher temperature leads to an increase in coarse-grain areas in particle boundaries and to a drop of coercivity.
2. The increase of densification temperature results in reduced oxidation of material because of disappearance of pores, which are the source of oxygen. It means that the decrease in coercivity with increasing densification temperature is not related to oxidation of the alloy but only to recrystalisation phenomenon.
3. The size of powder particles has not a significant influence on the remanence of hot–densified isotropic magnet but has a strong influence on coercivity. This is because the excessive comminution of powder increases the free surface of material which becomes a centre of nucleation of large grains in the process of high-temperature densification.
4. The increase of powder comminution leads to enhanced oxidation of material and causes a substantial worsening of coercivity.
5. From the investigated fractions, the optimum was the fraction with particle size d: 32–88 μm. The best sample of this fraction, densified at the temperature about 650 °C, was characterized by the following magnetic properties: J_r = 0,84 T; H_c = 1320 kA/m; $(BH)_{max}$ = 117 kJ/m^3.

References

[1] KIM H.T., KIM Y.B., KIM H.S., *J. Magn. Magn. Mater.*, *224* (2001), 173.
[2] GRÜNBERGER W., 15th Int. Workshop on Rare Earth Magnets and Their Applicatons, Dresden, RFN, (1998), 333.
[3] KRICHNER A., GRÜNBERGER W., GUTFLEISH O., NEU V., MÜLLER K.H., SCHULTZ L., 15th Int. Workshop on Rare Earth Magnets and Their Applicatons, Dresden, RFN, (1998), 373.
[4] SAITO T., FUJITA M., KUJI T., 15th Int. Workshop on Rare Earth Magnets and Their Applications, Dresden, RFN, (1998), 381.
[5] NARAYAN P.S., BASU K., 15th Int. Workshop on Rare Earth Magnets and Their Applicatons, Dresden, RFN, (1998), 349.
[6] KIM Y.B., KIM H. T., CHO S.H., KAPUSTIN G.A., *Mater. Sci.*, Vol. 21, No. 1, (2003), 141.
[7] KIM H.T., CHO S.H., KIM Y.B., KAPUSTIN G.A., *J. Appl. Phys.*, Vol. 93, No 10, (2003), 8137.
[8] VALIEV R.Z., GUNDEROV D.V., ZHILYAEV A.P., POPOV A.G., PUSHIN V.G., *J. Metastable Nanocristalline Materials*, Vol. 22 (2004), 21.
[9] BROWN D., MA B.M., CHEN Z., *J. Magn. Magn. Mater.*, 248 (2002), 432.
[10] MISHRA R.K., BREWER E.G., LEE R.W., *J. Appl. Phys.*, 63 (8), (1988), 3528.
[11] LI YING, KIM Y.B., WANG L., SUHR D.S., KIM T.K., KIM C.O., *J. Magn. Magn. Mater.*, 223 (2001), 279.
[12] LEWIS L.H., THURSTON T.R., WILDGRUBER U., WELCH D.O., *J. Appl. Phys.*, 82 (7), (1997), 3430.

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