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Microstructure and Properties of Magnets Obtained by Hydrostatic Extrusion of Nd–Fe–B Powder

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Nd–Fe–B MQ powder was initially densified in a copper capsule to reach about 60% of the theoretical density. Subsequently, three various processes of hydrostatic extrusion were conducted at room temperature. The values of true strain, obtained during the all three stages, were 1.38, 0.89, 0.94, respectively. The investigation performed showed that the coercivity of the material decreases as the strain increases. Decrease of the remanence was observed only for the highest strains. Size of the particles was strongly reduced during the extrusion processes. X-ray diffraction did not show changes in the phase structure of the material. The Mössbauer study, of the sample extruded within all the three stages, showed existence of the $Nd_2Fe_{14}B$ phase and 16% of other phase. Analysis of magnetization versus temperature confirmed that the additional phase was ferromagnetic.

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1. Introduction

Hydrostatic extrusion (HE) is a modern, advanced method of plastic deformation [1]. This method is widely applied for fabrication ultra-fine-grained materials in a form of wires or rods. The results presented in [2] confirm that HE can also be used for densification of powders. The characteristic feature of HE is high speed of the process, which retards grain growth in a case of extrusion at high temperature. Among major advantages of HE process one can include triaxial state of stress acting at the material in the course of extrusion, which in a case of extrusion of bulk materials retards formation and propagation of cracks and in application for powders allows their good densification.

So far, only one group reported the application of HE for densification of Nd–Fe–B powders [3]. It was shown that generation of high deformation ratios, in the HE method, enables fabrication of compacts having high density up to 90% of the theoretical value, for the powder extruded at room temperature (RT), with strain value of 3.17. Even better densities, 94 and 96% were achieved for extrusions at 700 and 800 °C, respectively, with lower strain values, 2 and 1.85, respectively. However, HE of powders at elevated temperatures led to grain growth, which resulted in deterioration of the magnetic properties.

Initial investigations showed that HE at room temperature also lowers the magnetic properties. Mechanism of this phenomenon is unknown. The major, hard magnetic $Nd_2Fe_{14}B$ phase is brittle and does not undergo deformation. X-ray phase analysis did not show changes in the phase constitution after extrusion.

The aim of the current studies is determination of changes in the microstructure of hydrostatically extruded Nd–Fe–B powder, which are responsible for the deterioration of the magnetic properties.

2. Experimental

For the studies the MQU-F42 high coercivity powder, applicable for hot working, was used. The powder had composition $Nd_{13.95}Fe_{73.27}B_{5.49}Co_{6.72}Ga_{0.57}$ (at.%) and was in a form of flakes, having thickness of 25 μm and much greater other dimensions. The powder particles consisted of nanocrystalline grains of the $Nd_2Fe_{14}B$ phase, having mean diameter of 30 nm. The theoretical density of the material was 7.5 g/cm^3.

The powder was initially densified in a copper capsule to about 60% of theoretical density. Subsequently the capsule was sealed under vacuum and welded with electron beam. The billet was hydrostatically extruded at room temperature, in the three-stage process, with application of various diameter reduction ratios, for each stage (Table). In the course of extrusion the copper billet was placed in a working chamber and surrounded by liquid pressured medium. Moving piston generated hydrostatic pressure. When the desired pressure was reached (it depends on the billet material and reduction ratio) the billet was extruded through the die. The final product was in a form of a rod with the Nd–Fe–B core.

The magnetic properties of the samples were examined using a Lake Shore vibrating sample magnetometer.

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TABLE

Selected parameters of hydrostatic extrusion and material properties after the process.

Process	Diameter reduction $[d_o^2/d_f^2]$	True strain ε_r [%]	Density ρ [g/cm^3]	Coercivity H_c [kA/m]	Remanence B_r [T]	Lattice deformation [%]
Initial powder	–	–	4.52	1690	0.750	–
Stage 1	2	1.38	5.48	1476	0.544	0.0485
Stage 2	1.56	0.89	6.84	1477	0.583	0.0974
Stage 3	1.6	0.94	6.89	1366	0.577	0.0989

Samples were characterized by XRD with Cu K_α radiation. The crystalline lattice deformation of the $Nd_2Fe_{14}B$ crystallites was determined using the Williamson–Hall method [4]. The Mössbauer results were recorded using a POLON spectrometer, working in a transmission configuration with the constant acceleration of the Mössbauer source ^{57}Co in an Rh matrix, having the source activity of 50 mCi. The quantitative analysis was done using the WinNormos for Igor package.

3. Results

The properties were studied for initial material and after each stage of extrusion. It was found that the remanence of the material increased with growing density (Table). However, for greatest deformation (stage 3) the remanence decreased even for the high value of density. The coercivity decreased with increasing density.

Analysis of the microstructure showed that the powder particles, in the course of extrusion, experienced noticeable refinement. Due to the change of size and shape, the particles, are able to relocate in the course of extrusion. As a result of densification the overall area of the powder particles and the area of boundaries between them increase. Materials processed by HE are characterized also by a strong lattice deformation. These phenomena can negatively affect the magnetic properties. Their influence is very difficult to quantitatively assess, however, one can assume that they should not be the major reason for the decrease of the magnetic properties. The X-ray phase analysis, performed after hydrostatic extrusion, showed neither changes in the phase constitution nor peaks intensity and locations.

For the investigations with the use of the Mössbauer spectroscopy, in order to record a representative spectrum for the entire volume, the specimens were pulverised. Both materials, the initial powder and the extruded rod, after the three-stage process, were examined. In the analysis of the Mössbauer spectra for the initial powder the presence of the main, hard magnetic $Nd_2Fe_{14}B$ phase was identified. For the $Nd_2Fe_{14}B$ component, a presence of the four crystalographically and six magnetically non-equivalent positions of the Fe atom, in the elementary unit of this phase, were considered (Fig. 1).

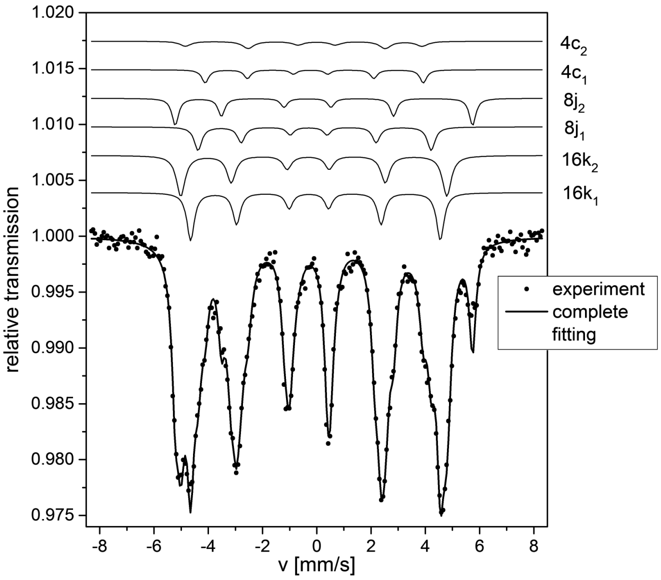

Fig. 1. Mössbauer spectra for the starting powder.

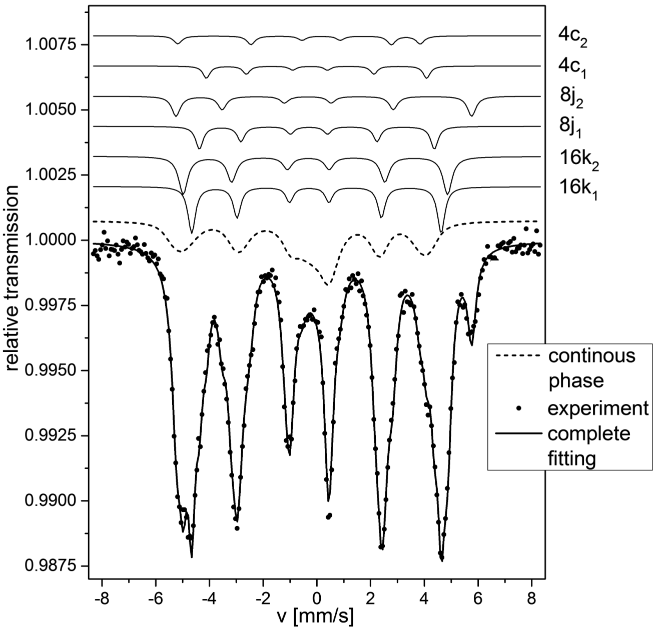

Fig. 2. Mössbauer spectra for the three-stage extruded specimen.

In fitting the experimental spectrum, for the three-stage extruded specimen, beside the six components, related to the hard magnetic $Nd_2Fe_{14}B$ phase, one continuous component, for which the distribution of the hyperfine fields was described, was taken into account. The presence of the continuous component can be related to either a presence of an amorphous or strongly disordered phase, which is formed during extrusion (Fig. 2). In Fig. 3 distribution of the hyperfine fields $P(B_{hf})$ for this phase is presented. In the plot one can distinguish two components of this distribution — low- and high-field, respectively. This evidences the existence of two various surroundings of the iron nuclei, in the disordered phase, having different local magnetic properties. In the extruded specimen, the values of the parameter such as B_{hf}, for the hard magnetic phase attain higher values than for the initial powder. This can evidence the presence of internal stresses in the elementary unit cell of this phase, related to the characteristics of the hydrostatic extrusion process. On the basis of the analyses of areas of the sextet's components we found that the fractions of the hard magnetic and disordered phases in the material amount to 84% and 16%, respectively.

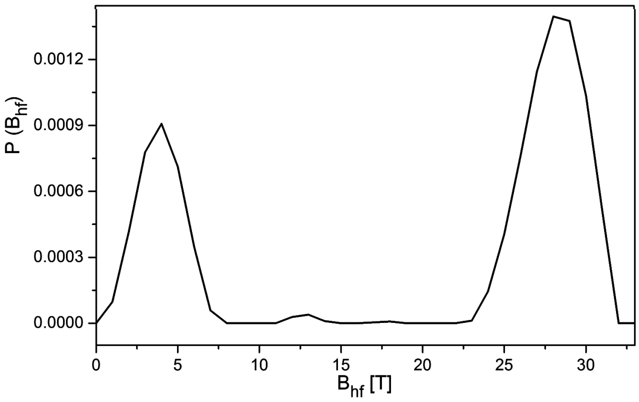

Fig. 3. Distribution of hyperfine fields for the three-stage extruded specimens.

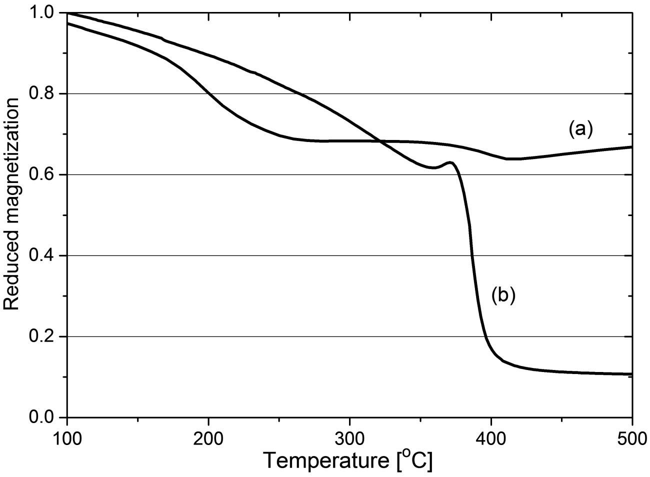

Fig. 4. Magnetization versus temperature for the three-stage extruded specimens (a) and initial powder (b).

In Fig. 4 magnetization versus temperature for the three-stage extruded specimens (a) and initial powder (b) are presented. Analysis of these dependences showed a presence of two, distinct deflection points for the extruded specimen (curve a). An abrupt drop of the magnetization, at a temperature of 390 °C is characteristic of the Curie temperature for the $Nd_2(Fe,Co)_{14}B$ phase. The other drop, observed around 200 °C is apparently correlated with the existence of the strongly disordered phase. The parallel deterioration of the magnetic properties at this temperature is an evidence of the formation of the new ferromagnetic phase during extrusion.

4. Conclusions

The experimental data evidence that hydrostatic extrusions of the Nd–Fe–B powder leads to formation of a new, strongly disordered phase, probably an alternative of the $Nd_2Fe_{14}B$ phase. Appearance of this phase in the material apparently leads to deterioration of the magnetic properties of the extruded material.

Acknowledgments

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