Topology of Nd-Fe-B Magnets with a high Energy Density

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Abstract—The topology and the effect of the B-content on the grain growth rate of sintered Nd-Fe-B magnets has been examined. Most of the grains have 5 to 6 corners. Normal grains grow slowly for B-contents <5.7 at.%, but grow faster for higher B-contents. Abnormal large grains grow almost as fast as grains in pure metals.

By optimizing the composition and refining the processing route sintered Nd-Fe-B magnets with a remanence of 1.528 T, a coercivity $H_{c2}$ of 840 kA/m (10.6 kOe) and a maximum energy density of 454 kJ/m³ (57 MGOe) could be prepared.

Index Terms—Topology, grain growth rate, Boron-content, energy density, magnets, temperature measurement.

I. INTRODUCTION

The excellent magnetic properties of the intermetallic Nd$_2$Fe$_{14}$B compound enable the production of permanent magnets with a high energy density by powder metallurgy [1]. Many R&D efforts are focused on the optimization of the composition as well as of the processing route in order to improve the maximum energy density of sintered magnets. The maximum energy density is mainly determined by the remanent polarization and the reversible permeability. The remanent polarization can be calculated from the saturation remanent polarization and the reversible permeability. The theoretical density of the alloy, the fraction of nonmagnetic constituents and the alignment coefficient [2, 3].

During sintering the irregularly shaped particles must be converted to a homogeneous and fine-grained microstructure, in order to achieve demagnetization curves $J(H)$ with an excellent squareness and strong coercivities $H_{c2}$. The basic requirement of space-filling and minimization of the grain surface energy results in a hexagonal lattice [4]. However, a typical microstructure consists of grains which have i corners, $3 < i < 10$ in general.

In order to maximize the volume fraction of the Nd$_2$Fe$_{14}$B compound, additions for improving the coercivity and impurities must be minimized. The fraction of RE-rich constituents, which are required for liquid phase sintering and for the magnetic decoupling of the individual grains [5] should be optimized. In general the fraction of RE-rich constituents ranges between 1.5 and 4 wt.% in sintered Nd-Fe-B magnets.

The dependence of the topology and the grain growth rate on the B-content of sintered Nd-Fe-B magnets has been investigated after different sintering conditions.

II. EXPERIMENTAL

For the investigations Nd$_{13.12}$Dy$_{0.46}$Fe$_{79.65-y}$TM$_{128}$B$_{5.51+y}$ magnets, TM: Co, Cu, Al, Ga, $y = 0, 0.06, 0.13, 0.23, 0.27$, were prepared by powder metallurgy from finely milled alloy-powders. The average particle size of the alloy-powders ranges between 3 and 5 µm. After the alignment in a dc magnetic field of 1300 kA/m - and in some tests additional alternating magnetic field pulses with a peak field strength of 6400 kA/m - the powder blends were pressed isostatically at a pressure of 200 MPa. The green compacts were sintered at temperatures between 1080 and 1120 °C for periods ranging from 2 to 24 h. Finally the magnets were annealed at 500 °C for 1 h.

Examination of the microstructure has been performed by metallographic analysis. The average grain size was determined by the three circular intercept method, according to ASTM E112.

The magnetic properties were deduced from demagnetization curves $J(H)$, recorded in a hysteresisgraph on test samples 9 mm in diameter, 6 mm in thickness, at temperatures between 20 and 80 °C.

III. RESULTS

Due to liquid phase sintering, Nd-Fe-B magnets achieve easily densities between 7.55 and 7.6 g/cm³ or $\rho/\rho_0 > 99 \%$, respectively.

A. Topology of Nd$_{13.12}$Dy$_{0.46}$Fe$_{79.65-y}$TM$_{128}$B$_{5.51+y}$ magnets, $y = 0, 0.06, 0.13, 0.23, 0.27$

In order to examine the topology of sintered Nd$_{13.12}$Dy$_{0.46}$Fe$_{79.65-y}$TM$_{128}$B$_{5.51+y}$ magnets with different B-contents the number of corners per grain has been counted of at least 100 neighbouring grains. The frequencies $P_i$ of grains with i corners are represented in Fig. 1 and 2. At a temperature
of 1100 °C the number of corners per grain does not change significantly for an extension of the sintering period from 2 up to 24 h. Most of the grains have 5 or 6 corners. The fraction of grains with 4 corners decreases from about 10 down to 3%, whereas the fraction of grains with 5 or 6 corners increases. There is only a small increase of the fraction of grains with 8 or more corners. Hence the microstructure of sintered Nd-Fe-B magnets is not very sensitive on the B-content.

Fig. 1. Fraction of grains \( P_i \) with \( i \) corners of Nd\(_{13.12}\)Dy\(_{0.46}\)Fe\(_{79.65-y}\)TM\(_{1.28}\)B\(_{5.51+y}\) magnets, sintered at 1100 °C for 2 h.

This is quite different to the topology of sintered Nd\(_{12.9+x}\)Dy\(_{0.46}\)Fe\(_{79.65-y}\)TM\(_{1.28}\)B\(_{5.51+y}\) magnets, \( x = 0, 0.25, 0.60, 1.00, 1.4 \), which have been sintered at 1080 °C for periods between 4 and 12 h. Due to the increased RE-content there was an increase of the fraction of grains with 8 or more corners for extended sintering periods at the expense of small grains. The fraction of grains with <4 corners and the fraction of grains with 5 and 6 corners decreased by about 5% each, indicating a substantial grain growth [6].

B. Grain Growth Exponent in Dependence on the Sintering Period

A homogeneous fine-grained microstructure is an essential prerequisite for achieving magnets with high coercivities and a proper squareness of the demagnetization curve \( J(H) \). According to the basic theory by J. E. Burke and D. Turnbull [7] the general parabolic grain growth equation reads:

\[
R_t = k \cdot t^{1/n}
\]

where \( R_t \) denotes the mean grain radius at time \( t \), \( n \) gives the grain growth exponent and \( k \) represents a constant. The grain growth exponents of pure metals range between 2 and 4.

In sintered Nd\(_{13.12}\)Dy\(_{0.46}\)Fe\(_{79.65-y}\)TM\(_{1.28}\)B\(_{5.51+y}\) magnets with different B-concentrations the grain growth exponent ranges between 16 and 20 for magnets containing <5.7 at.% B, but decreases to about 7.5 for B-contents >5.7 at.%, indicating some grain growth, see Fig. 3. In such magnets the average grain size ranges between 6 and 9 µm.

Fig. 2. Fraction of grains \( P_i \) with \( i \) corners of Nd\(_{13.12}\)Dy\(_{0.46}\)Fe\(_{79.65-y}\)TM\(_{1.28}\)B\(_{5.51+y}\) magnets, sintered at 1100 °C for 24 h.

C. Abnormal Grain Growth Exponent in Dependence on the Sintering Period

However, in all examined Nd-Fe-B magnets abnormal large grain growth occurred, see Fig. 4. The grain growth exponents for large grains range between 2 and 6, what is almost similar to the grain growth in pure metals. Since only a few large grains could be observed, the data scatter and the regression curves have got small reliability coefficients.

D. Magnetic Properties of Ternary Magnets

Minimization of the RE- and B-concentration, improvement of the alignment of the powder particles by 6 additional alternating field pulses, \( H_{\text{pulse}} = -/+6400 \text{ kA/m} \) [3], reduction of the impurities to about 1.5 % and of the transition metals

Fig. 4 Abnormal large grain radius \( R \) of sintered Nd-Fe-B magnets with
different B-concentrations in dependence on the sintering period \( t \). The sintering temperature was kept constant at 1100 °C. The exponents of the regression curves represent the inverse grain growth exponent \( n \) for abnormal grain growth.

TM to 0.8 and sintering to a fine-grained microstructure results in Nd\(_{12.5}\)B\(_{4}\)TM\(_{0.8}\)Fe\(_{80.7}\) magnets, TM: Al, Ga, Co, Cu, with an extraordinary maximum energy density.

Fig. 5 presents the demagnetization curves \( J(H) \) and \( B(H) \) of the best Nd-Fe-B magnet with a remanent polarization of 1.528 T, a coercivity \( H_c \) of the best Nd-Fe-B magnet with a remanent polarization of 840 kA/m (10.6 kOe) and a reversible permeability of 1.03 what results in a maximum energy density of 454.6 kJ/m\(^3\) (57 MGOe).

The temperature coefficients amount to -0.12 %/K for the remanent polarization and to -0.78 %/K for the coercivity in the temperature range 20 to 80 °C. The strong temperature dependence of the coercivity restricts the maximum application temperature to 80 °C for a loadline \( B/\mu_0 H = -2 \).

The grain growth exponent of sintered Nd-Fe-B magnets range between 16 and 20 for B-contents <5.7 at.%, but decreases to 7.5 for B-contents >5.7 at.%. That indicates an enhanced grain growth. At sintering temperatures of 1100 °C the volume of the liquid phase will increase for higher B-contents due to the eutectic reaction between the Nd\(_4\)Fe\(_{14}\)B and the Nd\(_{14}\)Fe\(_5\)B\(_2\) compounds [8].

Besides densification the increased volume of the liquid phase probably also enables grain growth by a solution precipitation process.

The grain growth exponent is quite similar to Nd-Fe-B magnets with different RE-rich constituents. In those magnets the grain growth exponents range between 30 and 40 for magnets with <4 wt.% RE-rich constituents and decrease to about 10 for magnets, which contain >4 wt.% RE-rich constituents [6]. The grain growth exponent for abnormal large grains ranges between 2 and 6 for magnets with different B-contents or a fraction of >4 wt.% RE-rich constituents.

Besides the sintering temperature the grain growth kinetics depend on the volume of the liquid phase at sintering temperature. By optimization of the composition and the manufacturing processes sintered Nd-Fe-B magnets with a maximum energy density of 454.6 kJ/m\(^3\) (57 MGOe) could be prepared.

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**REFERENCES**


