Role of Dy diffusion in sintered Nd-Fe-B-type hard magnets

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Introduction & Motivation

High coercivity Nd-Fe-B permanent magnets play an important role in rapidly-growing renewable energy sector, especially in electric vehicles or wind turbines. To retain the coercivity at high operating temperature, heavy-rare-earth elements (HRE), such as Dy and Tb, are added using grain-boundary diffusion (GBD) process. The addition of HRE results in a significant improvement of the coercivity due to the increase of the intrinsic resistance to demagnetization.

In the present study, we report on the correlation between the magnetic properties and the distribution of the Dy in the melt-spun $Nd_2Fe_{14}B$ ribbons. Ribbons were coated with the DyF_3 particles, pressed and spark-plasma sintered (SPS). The magnets were further annealed to assure the diffusion process which enhanced the magnetic properties.

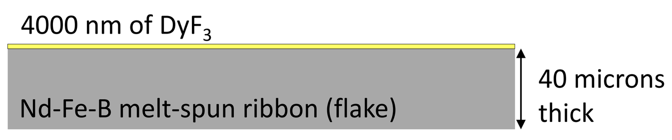
Results & Discussion

During SPS and annealing process Dy diffused along the grain boundaries (GB) into the outer parts of the Nd-Fe-B grains, thus forming core-shell grains with Dy-rich shell and Nd-Fe-B core. The magnetometry measurements (measured at room temperature using a Steingroever permeameter) showed that the addition of Dy increased the coercivity in heattreated samples by 25 %. For the structural and compositional studies, we used a Cs-corrected scanning transmission electron microscope (FEI Titan 80-200) equipped with SuperX electron dispersive X-ray (EDX) spectrometer and electron energy-loss (EEL) spectrometer (Gatan Enfinium ER model 977).

Processing and magnetometry measurements

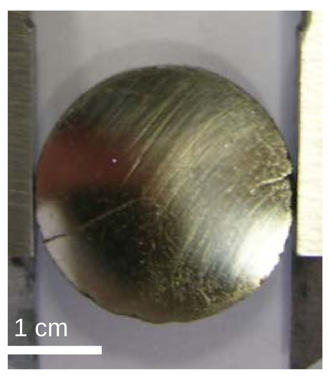
Dy-free Nd-Fe-B ribbons

1. Coating the ribbons with the DyF_3.

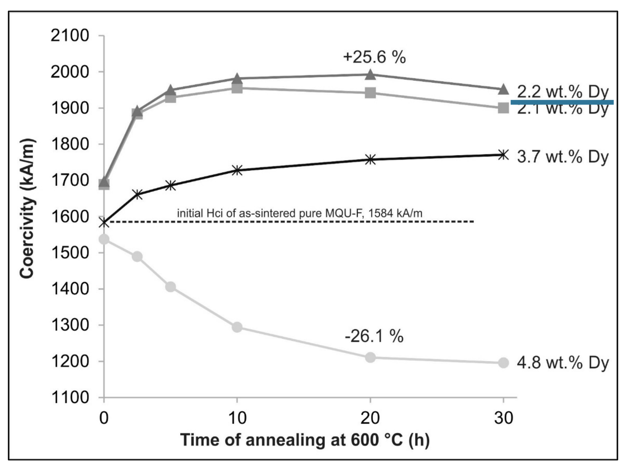


2. Spark-plasma sintering @ 700 °C for 1 h.
3. Annealing @ 600 °C.

(Dy,Nd)-Fe-B magnet

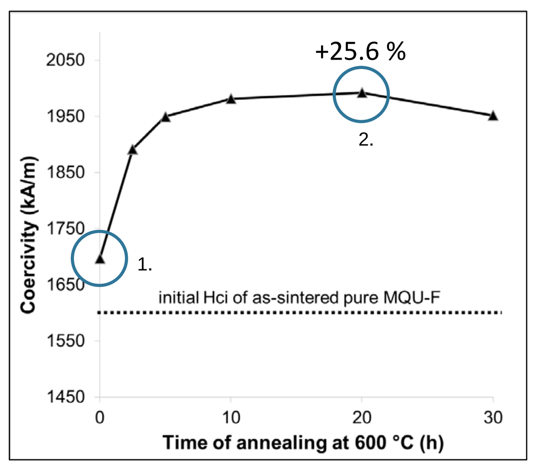


Magnetometry measurements



Coercivity values of SPS and annealed ribbons mixed with various amounts of DyF_3 powder.

Selected sample

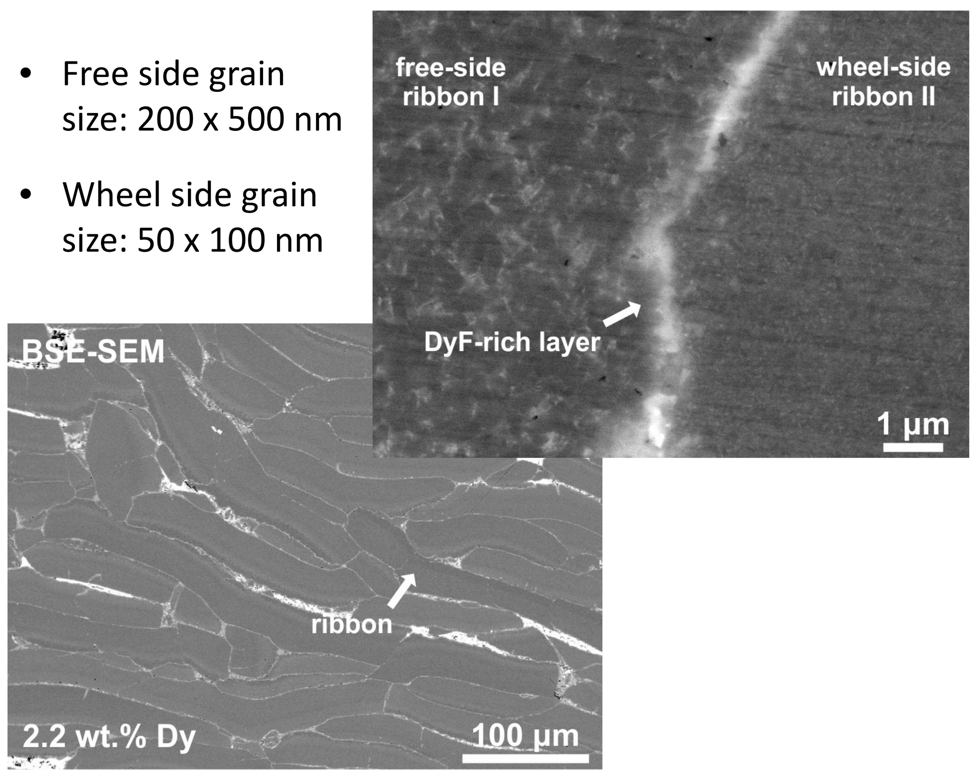


1. After spark-plasma sintering.
2. After spark-plasma sintering and annealing.

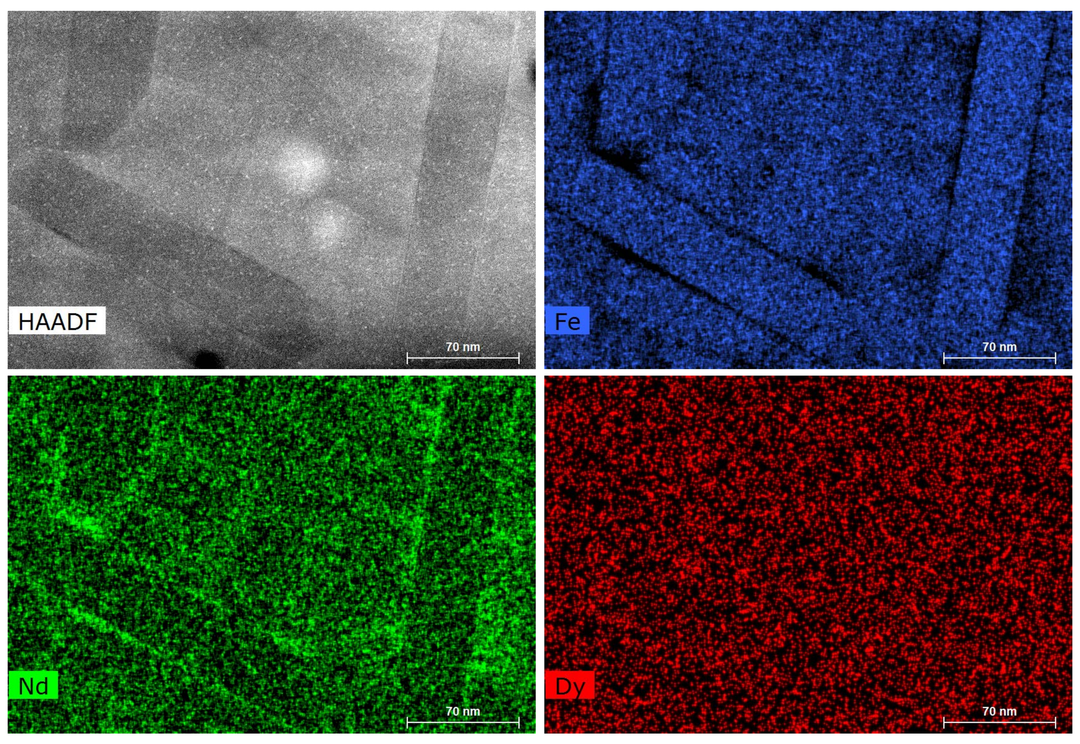
EDXS and EELS analyses on wheel side region of the Dy-treated Nd-Fe-B magnet

Investigated microstructural features

- Free side grain size: 200 x 500 nm
- Wheel side grain size: 50 x 100 nm

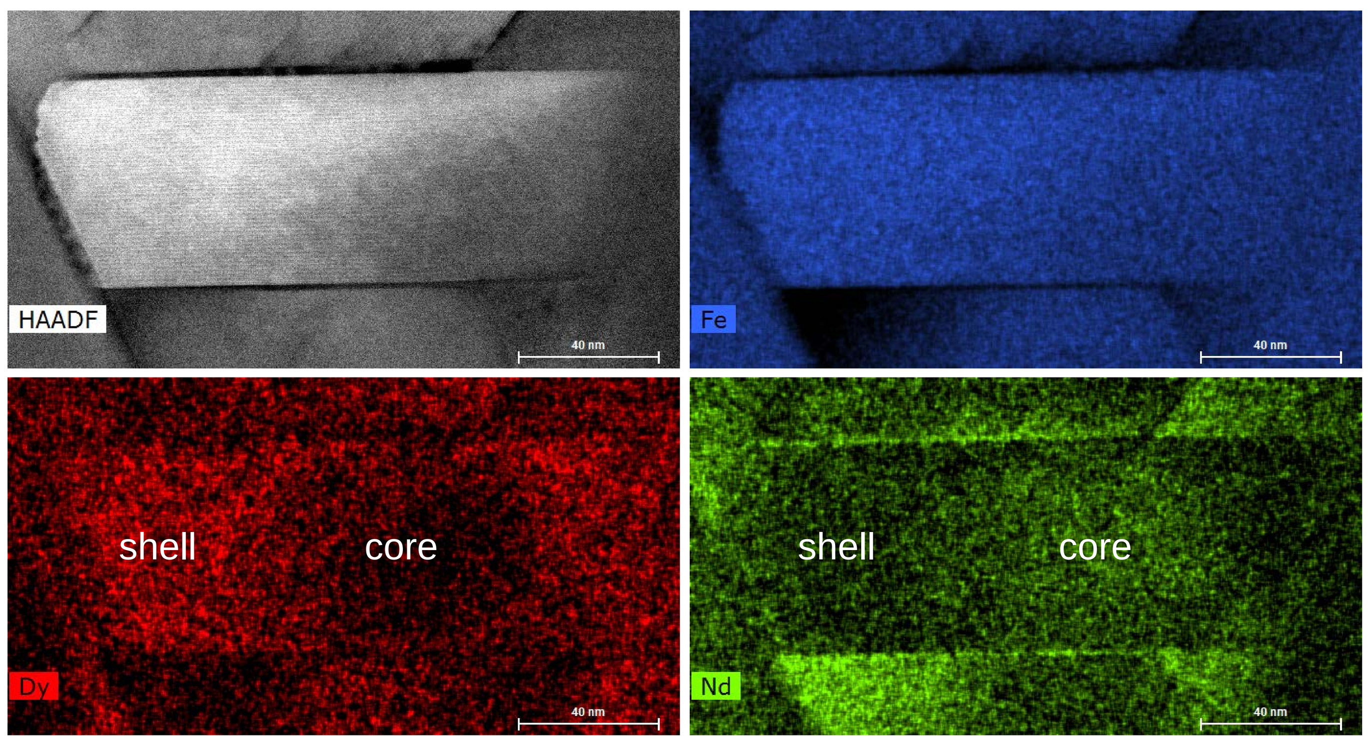


Spark-plasma sintered magnet - STEM and EDX maps



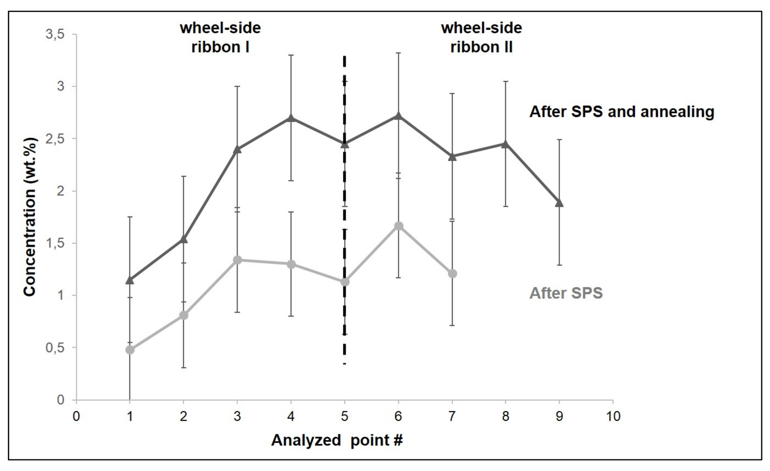
- No core-shell structures detected.
- Fe is homogeneously distributed through the Nd-Fe-B grains.
- Nd-rich grain boundaries and triple pockets.

Spark-plasma sintered and annealed magnet - STEM and EDX maps



- Core-shell-like structure: Nd-rich core and Dy-rich shell.
- Fe is homogeneously distributed through Nd-Fe-B grains.
- Nd-rich grain boundaries and triple pockets.

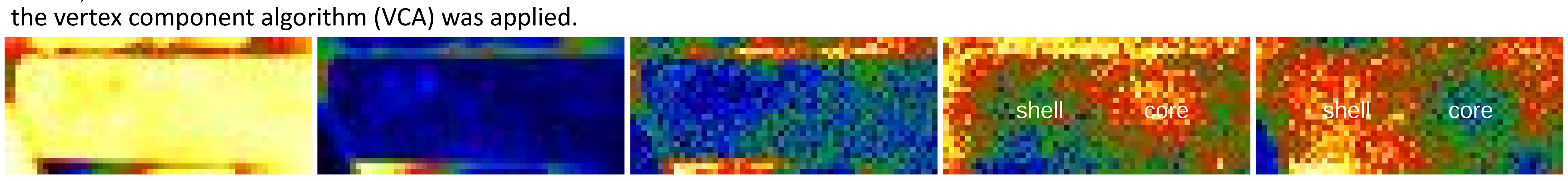
SPS and annealing – SEM/EDX line scan



Concentration profile of the Dy on **wheel** side is **2 times higher after annealing.**

Spark-plasma sintered and annealed magnet – STEM, EEL maps and vertex component algorithm (VCA)

The direct integration of the power law background-subtracted spectra did not allow us to fully separate the different contributions of elements. The Fe-L edge overlaps with the F-K edge tail which means overestimation of the Fe concentration. The overlap of the Nd-M_3 and Dy-$M_{4,5}$ edges is also present at higher energies and prevented an accurate determination of the Dy concentration. To unwrap the spectra, the vertex component algorithm (VCA) was applied.



Fe-$L_{2,3}$ / Co $L_{2,3}$ F-K O-K Nd-$M_{4,5}$ Dy-$M_{4,5}$

Conclusions

We concluded that the 25 % increase in coercivity is strongly linked to the heat-treatment process where the Dy diffuses along the grain boundaries and into the outer parts of the Nd-Fe-B grains where it partially substitutes Nd to form a core-shell-like grains. In the case of spark-plasma sintered sample we did not observe any diffusion of Dy, while the annealed sample revealed the (Dy,Nd)-Fe-B phase formation in the shell around the pure Nd-Fe-B (core) grains. Applying the vertex component algorithm was beneficial for the reliable detection of present elements in the samples. Further studies will focus on the quantitative analyses of the spark-plasma sintered and annealed sample.

Acknowledgements

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