Improving the properties of recycled rare earth permanent magnets with reduced heavy rare earth content through microstructural engineering

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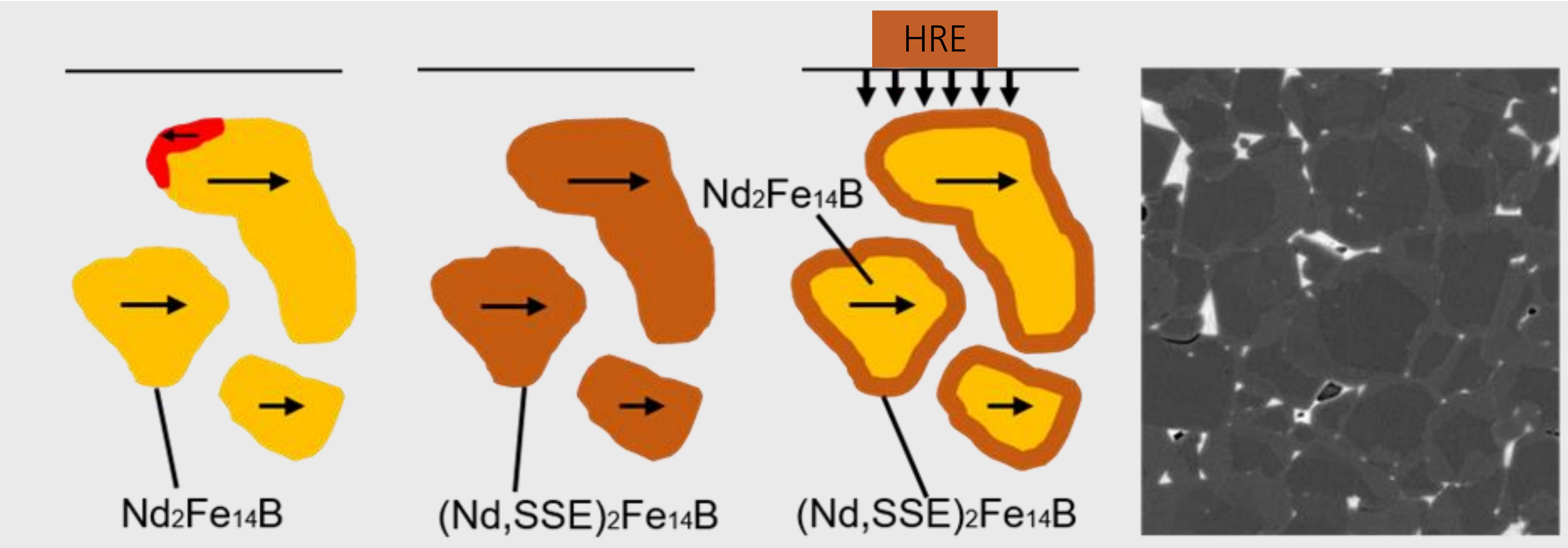
Today, rare earth (RE) permanent magnets based on Nd-Fe-B exhibit the highest energy densities of all magnetic materials. Due to their excellent properties, they are of strategic importance in key technologies like e-mobility, robotics and renewable energy technologies. However mining and conditioning of the RE-containing ores do not only have considerable negative effects on the environment but are also limited to a few locations worldwide. The reliability of supply is considered as highly critical*.
Industrial recycling of RE permanent magnets is one possibility to secure Europe's supply and to mitigate the criticality of these important materials. Additionally, magnets made of recycled material are ecologically favorable due to the elimination of mining and refining of rare earth elements.
For application in the mobility sector a high coercivity is required and the magnets often feature an increased content of very critical heavy rare earth elements (HRE) dysprosium or terbium. In order to optimize the required HRE content we analyzed in detail the element distribution and mechanism of coercivity increase at atomic level via atom probe microscopy for two different recycling approaches.

Functional Recycling of Nd-Fe-B permanent magnets

Through functional recycling the rare earth containing magnets can be processed directly into new magnets. The recycling process steps are the same which are used for the production of primary magnets: Hydrogen Decrepitation (1), Milling (2), Allingment & Pressing (3), Sintering & Annealing (4). Compared to primary magnets it´s not necessary to melt raw materials which saves energy and reduces production costs.

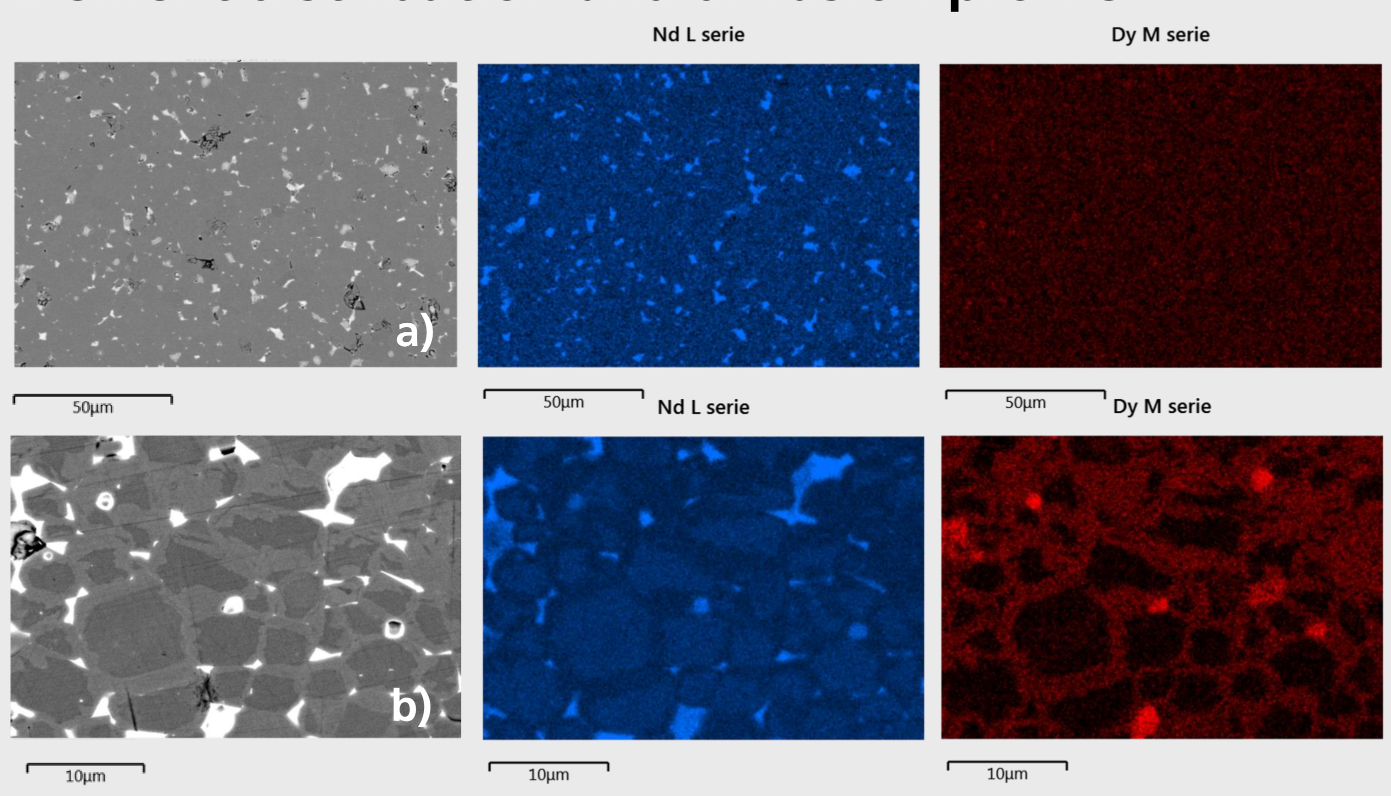
For increasing the coercivity often HRE elements are needed. With GBD (see right picture) the required HRE content can be reduced.



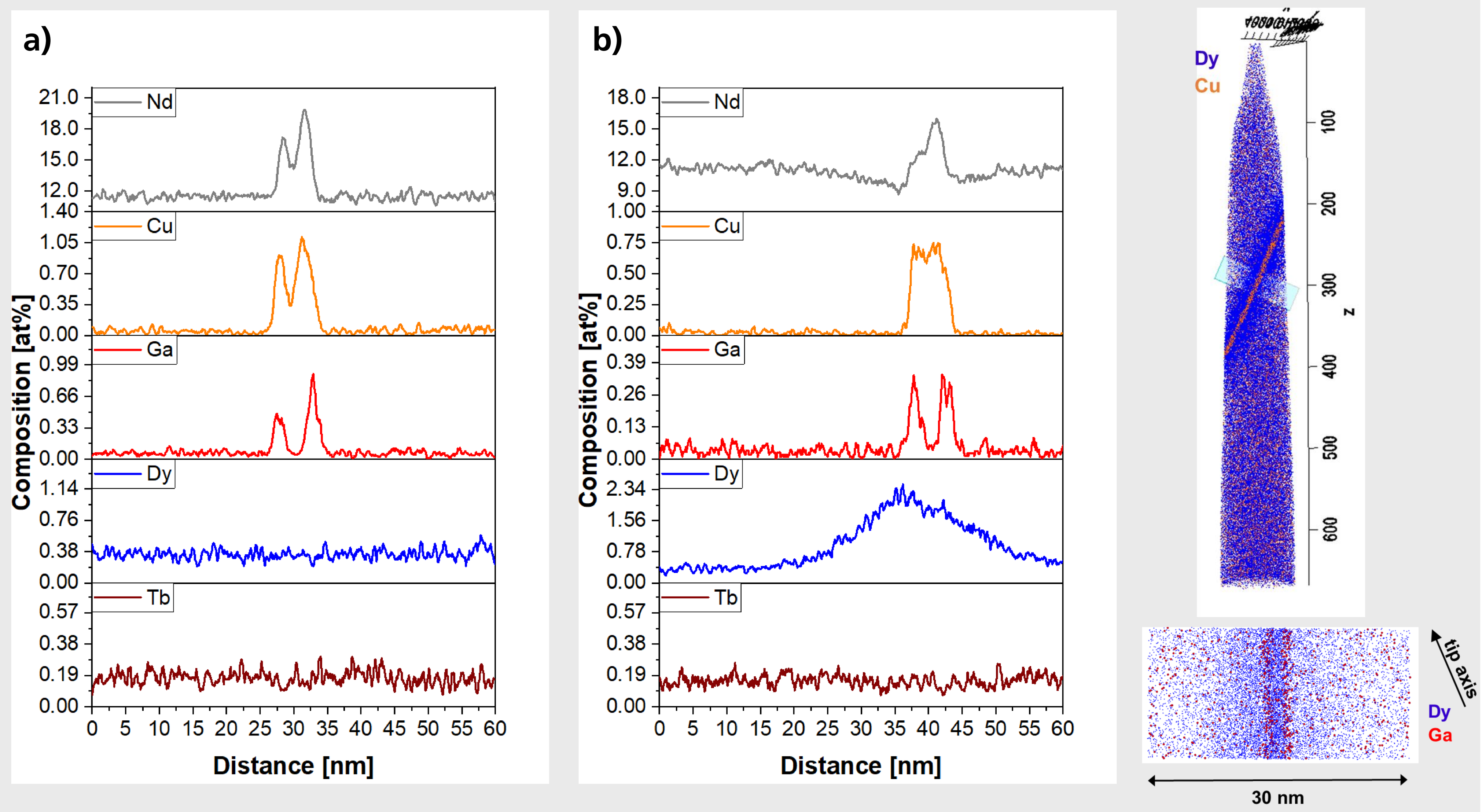
(a) Magnet without HRE **(b)** Magnet with HRE addition (e.g. hydrides) **(c)** Magnet with HRE addition via GBD

Adding HRE elements increases the coercivity of Nd-Fe-B magnets. The HRE are only effective at the outer region of the grains through magnetic hardening. Adding of HRE hydrides leads to a homogenous distribution of HRE in the whole grain. With Grain Boundary Diffusion (GBD) a core-shell-structure can be achieved with HRE location only in the outer regions of the grains**.

Element distribution and diffusion profile

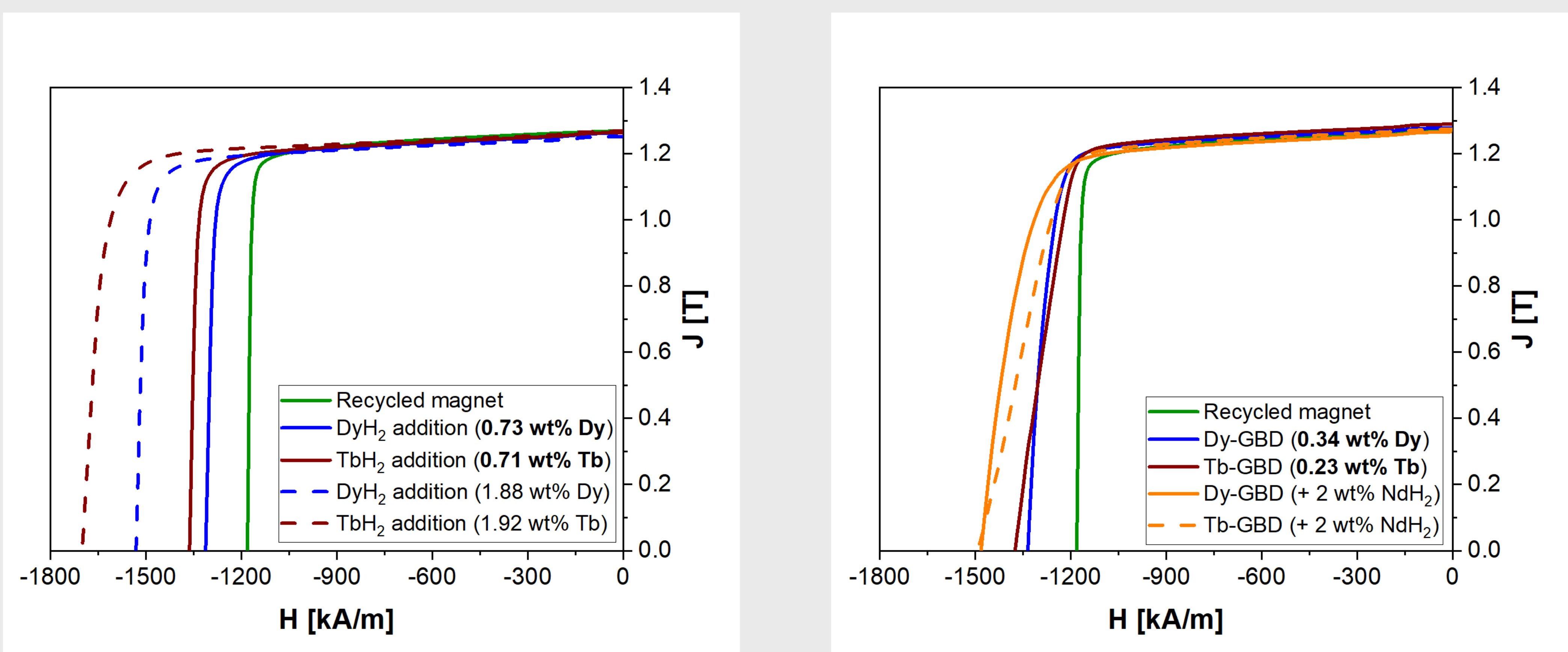


EDX images of a recycled magnet with addition of DyH_2 (a) and Dy-GBD (b). While the magnet which is produced via hydride addition shows a homogenous distribution of the HRE element Dy within the grains, the GBD magnet show a core-shell-structure where Dy is located at the outer regions of the $Nd_2Fe_{14}B$ grains.



3D atom probe investigation on different recycled magnets. The reference (a) without any addition of elements shows a uniform Dy distribution and an enrichment of Cu and Ga at each grain-boundary-interface. The Dy-GBD sample (b) show the same Cu and Ga enrichment. The thickness of Dy-shell is 15 nm while thickness of grain boundary is 9 nm.

Magnetic properties of recycled magnets



The difference in element distribution has also an influence on the magnetic properties. With GBD and core-shell-structure only the half of very critical HRE Dy is needed to achieve the same coercivity Hc compared to hydride addition. A higher amount of Dy or Tb increase the coercivity, too. Both methods (hydride addition and GBD) can be combined for a further Hc increase (right picture) which is caused by an improved diffusion behavior and diffusion depth (not shown here).

Summary

Functional recycling of Nd-Fe-B permanent magnets is one possibility to reduce the criticality and ecological footprint of this important material which has many applications in several key technologies. The recycling process can be easily implemented in the pimary production route.

Directly recycled magnets have properties which are comparable to primary magnets. Controlling the microstructure and element distribution of magnets can lead to optimized magnetic properties and an ideal usage of critical raw materials such HRE.

In the ongoing project „FUNMAG – Funktionelles Magnetrecycling für eine nachhaltige E-Mobilität" we will further analyze and improve the microstructure and properties of recycled magnets for different applications.

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*Study on the review of the list of Critical Raw Materials 2017, European Commission.
**Own depiction according to S. Sawatzki, „Der Korngrenzendiffusionsprozess in nanokristallinen Nd-Fe-B-Permanentmagneten", Technische Universität Darmstadt, 2016 and K. Löwe et al., „Temperature-dependent Dy diffusion processes in Nd-Fe-B permanent magnets", Acta Materialia, 2015.