

Optimizing the production process of NdFeB magnets

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Abstract

Permanent magnets and especially NdFeB (Neodymium Iron Boron) magnets are one of the most critical components in the global manufacture of modern electric motors. The production process of NdFeB magnets contains a large number of different procedures, all of which can have an influence on the properties of the end product. Although at first glance, relatively similar to the classic PM process, the manufacturing route involves some special challenges that will be examined in more detail in this paper. Starting with the optimization of the hydrogen decrepitation of the base material this contribution will additionally describe the effect of different milling parameters on microstructure and magnetic properties, the procedure of dimensionally precise compaction and sintering of magnet preforms and the magnetic testing of finished magnets.

Introduction

Neodymium iron boron (NdFeB) magnets are among the most powerful permanent magnets currently available. They are vital components in numerous modern technologies, including electric vehicles, wind turbines, and different electronic devices. Their exceptional magnetic properties enable efficient energy conversion, contributing to the development of sustainable energy solutions. Global production of rare earth magnets is key to meet the growing demand for clean energy technologies and advanced electronic devices. Securing diverse and reliable supply chains for rare earth magnets is essential for national security and economic stability, highlighting the strategic importance of producing these critical materials. Contrary to what the name suggests, rare earth elements are relatively widespread in the earth's crust. However, they are usually present in low concentrations. Currently, around 60% of global mining of rare earth ores takes place in China. This proportion is even higher if only the heavy rare earth elements (HREE) like Dysprosium and Terbium are considered which are essential for high performance magnet grades. The processing and metallization of rare earth ores also takes place in Asia for more than 98% of global production. Consequently, in the recent decades the parts production of rare earth magnets followed the availability of raw materials and is concentrating in Asia, so that more than 99% of the global NdFeB magnets production is taking place in this area (compare figure 1) [1].

Encouraged by North American or European programmes like the US REEShore Act of 2022 [2] and the EU Critical Raw Materials Act [3] motor manufacturers are looking more and more for local-to-local and flexible supply chains. Due to the tight supply situation in the rare earths sector together with the key technologies of e-mobility and renewable energy, which are dependent on these materials, there is a strong drive to increase production capacities for NdFeB permanent magnets globally.

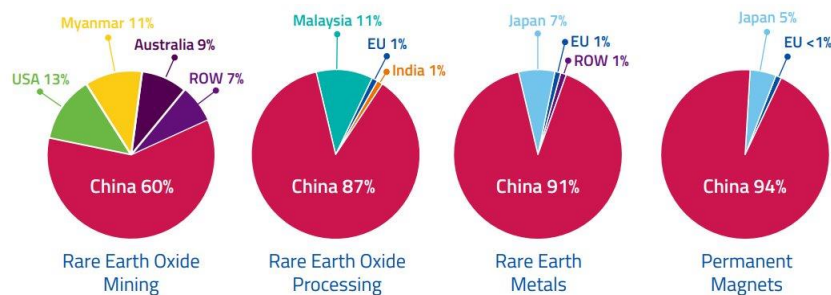


Figure 1: Estimated market shares in 2019 for rare earth materials and magnets [1].

Although the production of sintered NdFeB magnets appears to be very similar to the conventional PM route at first glance, there are significant differences in the processing of these products. All individual steps in the manufacturing process are aimed at tailoring the alloy focusing on its function, which means achieving targeted magnetic properties by best possible alignment of the magnetic domains in the finished magnet at lowest consumption of Rare Earth (RE). An overview of the manufacturing process steps for NdFeB magnets is shown in Figure 2. In this paper, we will describe the various process steps and present some experiments and optimization measures that were carried out for some individual sub-steps.

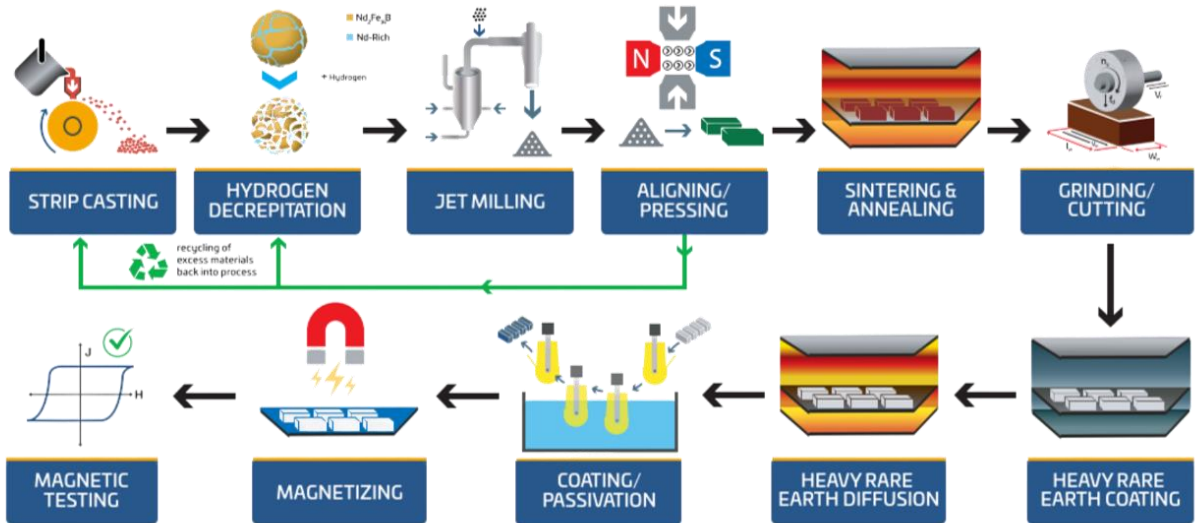


Figure 2: Production route for sintered NdFeB magnets.

Strip Casting

The main phase in NdFeB magnets consists of the magnetic ϕ -phase $\text{Nd}_2\text{Fe}_{14}\text{B}$, whose grains are surrounded by a seam of an Nd-rich secondary phase in the microstructure. The ϕ -phase forms a tetragonal crystal structure in the space group $P4_2/mnm$. The magnetic dipole moment runs along the crystallographic c-axis, the so-called "easy axis". In addition to the elements of the main phase and the additional rare earth components of the secondary phase, the alloys usually contain other alloying elements to adjust the Curie temperature, the other magnetic parameters, corrosion resistance or the grain size. In order to produce a small grain size in the material, the alloys are rapidly cooled using the strip casting process. In this process, the melt is poured onto a rotating, water-cooled copper wheel so that flakes with a thickness of approx. 0.3 mm and a length of a few centimeters are produced.

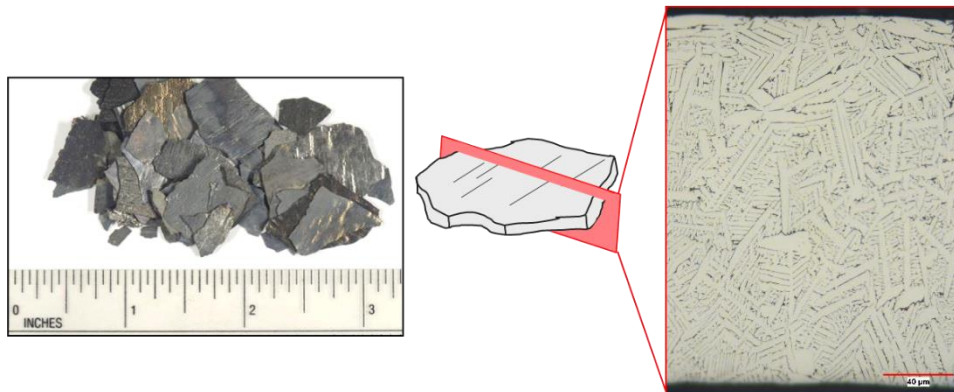


Figure 3: NdFeB-flakes produced by strip casting (left) and microstructure with light grey $\text{Nd}_2\text{Fe}_{14}\text{B}$ -phase and dark grey Nd-rich phase (right).

Hydrogen Decreptation

In order to process the flakes from the strip-casting process into a fine, if possible single-grain powder, a procedure called Hydrogen Decreptation (HD) is carried out. This utilizes the fact that neodymium and neodymium alloys form various hydrides with hydrogen, which have a lower density than the pure metal. Particularly the Nd-rich grain boundary phase expands considerably during the reaction with hydrogen, so that the material is broken up. The reaction takes place at temperatures below 100°C in a hydrogen atmosphere and leads to the formation of Nd hydrides or hydrides of the ϕ -phase with the composition $\text{Nd}_2\text{Fe}_{14}\text{BH}_x$. The material breaks apart in the process of hydrogenation. In the subsequent decomposition of the hydrides, the material is heated in a vacuum, whereby first the $\text{Nd}_2\text{Fe}_{14}\text{BH}_x$ decomposes (100-150°C) and then the NdH_3 is converted to NdH_2 (220-380°C) [4]. As the powder formed reacts strongly exothermically with oxygen and NdH_2 leads to a slight passivation of the material, the NdH_2 formed is left in the material until later sintering.

To optimize the process parameters, two different alloys were hydrogenated at a temperature of 100°C in a 1.15 bar hydrogen atmosphere. The dehydrogenation was then carried out under vacuum at different temperatures. The hydrogen and oxygen contents of the resulting powders were measured afterwards. The samples for the gas analyses were prepared in a glove box under argon atmosphere to prevent oxidation of the samples with atmospheric oxygen. This is particularly important as the material reacts directly with oxygen after the decreptation step due to its reactivity and larger specific surface area and can no longer be reduced in the further course of the production process. However, increased oxygen levels have a negative effect on the magnetic properties of the finished magnets. In addition, a reaction with atmospheric oxygen can lead to ignition of the material (pyrophoric). For this reason, all process steps from HD to sintering are carried out in a dry protective gas atmosphere (nitrogen or argon).

The results for the powders produced at different dehydrogenation temperatures are shown in Figure 4. For both alloys tested, an increase in the dehydrogenation temperature from 440°C to 470°C shows a significant reduction in the remaining hydrogen, which indicates an intended decomposition of the NdH_3 which is a compound that is undesirable for further processing. The measured hydrogen levels also indicate a conversion of NdH_3 to NdH_2 . It is also noteworthy that the decrease in the hydrogen content at this temperature is accompanied by a significant reduction in the oxygen content in the material. It can therefore be assumed that the atomic hydrogen (nascent) formed from the hydride decomposition has sufficient reducing power to decompose some of the rare earth oxides. Since the oxygen content is one of the key properties to produce high-performance magnets and cannot otherwise be reduced in the process, this fact is remarkable.

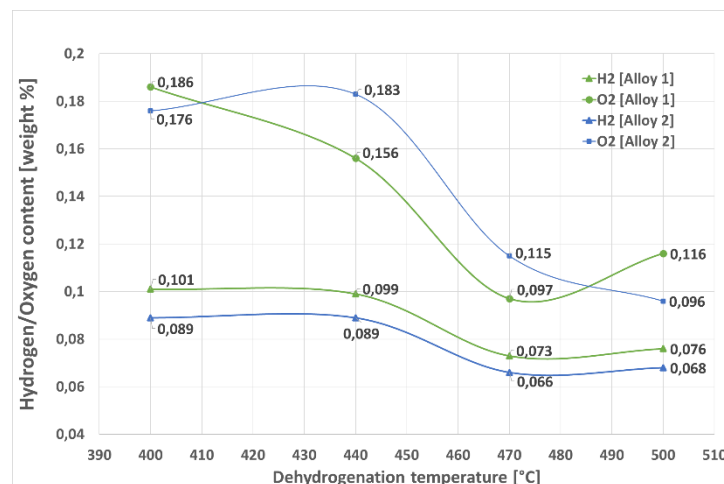


Figure 4: Variation of dehydrogenation temperature during hydrogen decreptation – Effect on hydrogen and oxygen content.

Jet Milling

In the next process step jet milling is used to grind the material efficiently to fine particles below 10 μm . This is required to generate single-crystalline powder particles with only one magnetic orientation per particle. This is required for a most effective alignment during the compaction step of the green parts. For milling high-speed gas streams (nitrogen or argon) impact and pulverize the hydrogen decrepitated powder. Jet milling is a common technology widely applied across industries like pharmaceuticals or chemicals and ensures precise particle size control and minimal contamination, making it ideal for heat-sensitive materials. The following optimization trials were performed in a Jet Mill 100 AFG from Hosokawa Alpine operating with nitrogen as processing gas. The mill is equipped with one classifier whose rotation speed defines the maximum particle size during milling. Furthermore, the variation of gas type, gas pressure, gas flow or powder feed can be used to tailor the particle size distribution. The desired particle size for the powder should have the narrowest possible distribution curve (uniform powder size). Furthermore, a small maximum particle size is aimed for ($d_{99} < 8 \mu\text{m}$) to reduce the number of multi-grain powder particles in which the alignment of the crystallographic axis (easy axis) is not given. On the other hand, the proportion of ultrafine particles should be as low as possible, as these can change the sintering activity during subsequent sintering, have negative impact on alignment, may lead to undesirable abnormal grain growth during sintering, and increases the flammability of the product powder.

Figure 5 documents various experiments to vary the milling pressure on the resulting particle size distribution of a ground NdFeB sample material. The ratio of d_{90}/d_{10} is used here to quantify the distribution width of the particle size distribution and should preferably have a value below 4. In addition, the proportion of unwanted ultrafine particles $< 0.9 \mu\text{m}$ is listed individually. The evaluation of the tests shows a positive influence of lower milling pressures on the particle size distribution and the quantity of ultrafine particles.

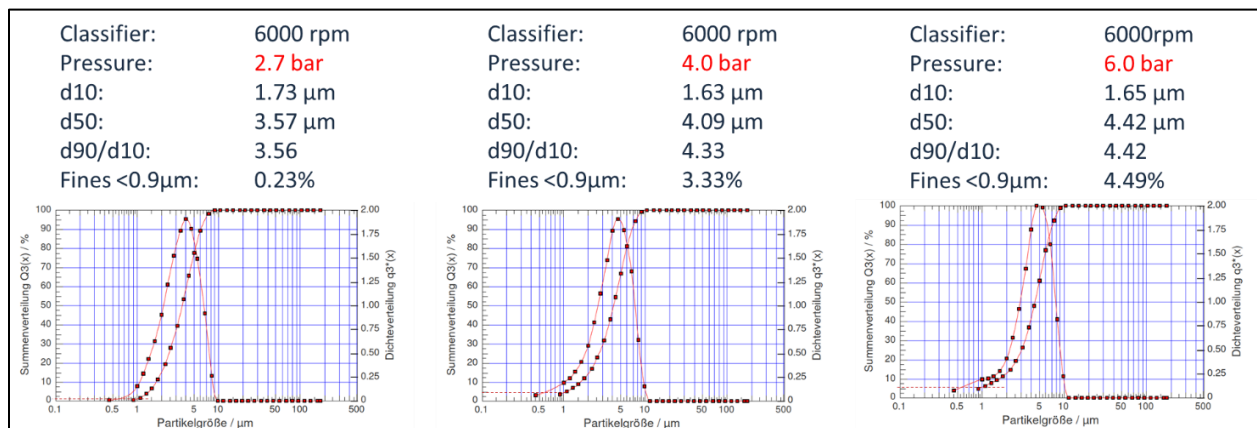


Figure 5: Effect of different milling pressures on particle size distribution of milled NdFeB powder (measured with Sympatec HELOS/KR).

The classifier rotation speed can be used to cut off the coarser particles and limit the maximum particle size. The effect of variations of this process parameter is shown in Figure 6 (left) and describes the reduction of particle size with increased classifier speed. It is noteworthy that the proportion of fine powder fractions (d_{10}) remains relatively constant with the variation.

As mentioned above, the base material is a two-phase material consisting of a ϕ -phase ($\text{Nd}_2\text{Fe}_{14}\text{B}$) and a rare earth-rich secondary phase. The milling behavior of these two constituents or phases differs, so that segregation or change in the rare earth concentration while milling is to be considered. A corresponding experiment is documented in Figure 6 (right), in which the Nd and total RE content was documented over a longer milling period. After a longer milling period and after the end of the product feed, there are clear reductions in the secondary phase (Nd-rich), which may have to be taken into account for the final products.

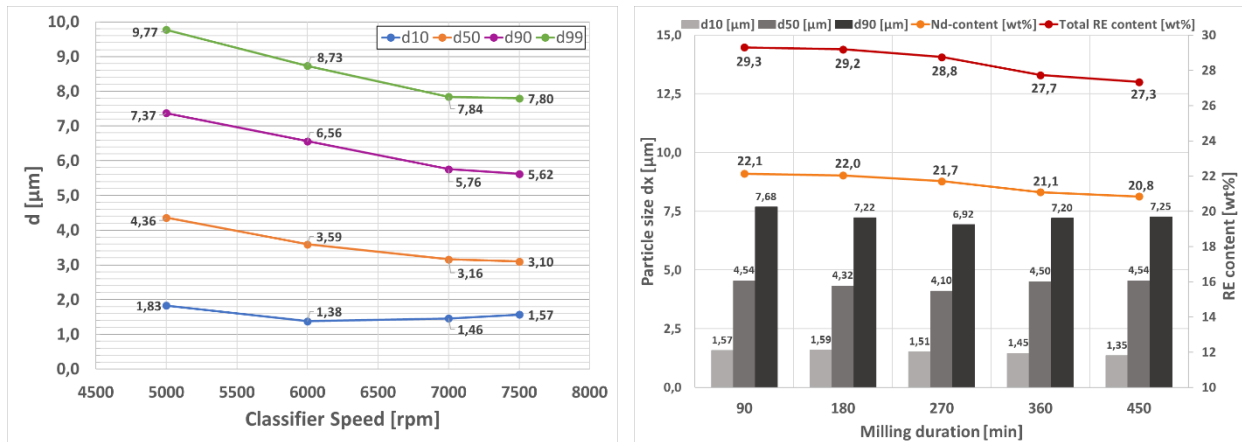


Figure 6: Impact of classifier speed on particle size distribution (left) and variations in RE-content and particle size during milling time (right).

Powder Compaction

To produce the formed parts, the NdFeB powder produced according to the process described above is compacted using a powder press. It should be noted that the entire process must take place under inert atmosphere due to the oxygen affinity of the powder and the green parts. At the start of the process, the powder is fed into the pressing tool via a dosing system. The correct amount of powder can be determined by volume or weight. Once the die has been filled, the cavity is closed with the upper punch without pressure and the powder in the closed tool is oriented by an externally applied magnetic field. In this way, all magnetic domains in the powder are aligned and the maximum remanence is obtained in the finished component. The orientation can be perpendicular to the pressing direction (transverse) or parallel to the pressing direction (axial). After orientation, the component is pressed to a green density of 4-5 g/cm³ and ejected.

Sintering

After forming, the green parts are sintered. Sintering takes place in a vacuum furnace at pressures below 10⁻³ mbar and temperatures between 1000 and 1130°C. During the heating process, the burn-out behavior of additives and the decomposition of rare earth hydrides still present must be considered. In all cases, reactions with atmospheric oxygen must also be avoided when loading the furnace and handling the green parts. The optimum sintering conditions depend on the chemical composition of the magnetic alloy, oxygen content and the particle size distribution. During sintering, the parts shrink by approx. 35% in the direction of the magnetic orientation and by approx. 10% in the other two spatial directions. The sintering temperature is selected so that sufficient shrinkage is achieved with a sintering density of more than 7.55 g/cm³. At the same time, a too high sintering temperature should be avoided, as this can lead to abnormal grain growth in the microstructure. These abnormal grains later act as starting points for demagnetization and thus impair the magnetic performance of the magnet. Figure 7 on the left shows a series of tests to determine the optimum sintering temperature as a function of density. The right part of the figure shows an exemplary microstructure with abnormal grain growth.

Machining

After sintering, the sintered components are ground and, if necessary, cut. When selecting the processing media, sufficient corrosion protection must be ensured in order to prevent dissolution of the secondary phase, which is rich in rare earths. Developments in the field of machining are not discussed in this report.

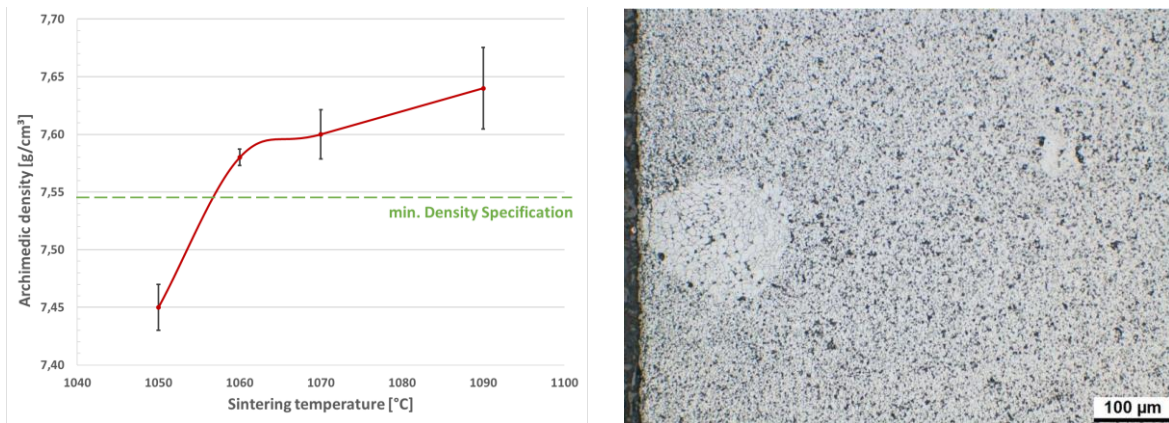


Figure 7: Sintering density of an NdFeB alloy depending on temperature (left) and microstructure with locally abnormal grain growth due to too high sintering temperature (right).

Grain Boundary Diffusion

The coercivity of NdFeB magnets experiences a notable decline at higher temperatures, attributed to the low Curie temperature T_c (approximately 312°C) and the substantial temperature sensitivity of the hard magnetic $\text{Nd}_2\text{Fe}_{14}\text{B}$ main phase [5,6]. One traditional method of producing high-coercivity Nd-Fe-B magnets for higher operating temperatures involves incorporating heavy rare earth elements (HREE) such as Dy and Tb as an alloying element during the melting process. Unfortunately, these elements are very expensive (Dy > 500 €/kg, Tb >1200 €/kg) and the consumption of HREE for the prealloying method is rather high. In order to overcome this situation, the grain boundary diffusion method (GBD) for NdFeB magnets was introduced in 2000 [7]. In this process, HRE permeates from the surface to the interior of the magnets during a heat treatment, predominantly reinforcing the secondary grain boundary phase and the surface near parts of ϕ -phase grains substituting Nd by Dy or Tb. With this method the quantity of Dy is only about 10% of the material used in the conventional prealloying method [8].

For this method, the surface of the magnet is cleaned and coated with an HREE source. This coating can be carried out using Dy or Tb compounds (oxides, fluorides or hydrides) or pure metals using various coating processes (painting, CVD, PVD, etc.). In a subsequent heat treatment step, the HREs are diffused into the material via the RE-rich secondary phase, if necessary, after decomposition of the precursor used. The diffusion depth and distribution of the HREs depends, among other things, on the treatment temperature and time. Due to the large number of parameters involved in this production step, this article will not go into detail and reference is made to the further literature on this process.

Corrosion protection

As already mentioned, several times, the material for NdFeB magnets is very reactive. This is mainly due to the high proportion of rare earth elements, which are very sensitive to oxidation in their metallic form. During the production process, this aspect is particularly relevant for intermediate stages with a large specific surface area. For example, the milled powders or the compacted green parts behave pyrophorically. However, corrosion protection is also required for the finished magnetic components. Various coatings are generally used here, such as epoxy resin or Nickel coatings. If the protection is only required for a short period of time, for example for transportation until the magnet is installed in a motor and encapsulated, passivation of the component surface in most cases is sufficient. Phosphating is generally used for this purpose.

During phosphating, a so-called conversion layer is created on the component surface by reacting the component material with phosphoric acid or phosphate solutions. Although a proportion of the material should react with the acid, a deeper corrosive attack must be avoided. The choice of suitable process media and parameters such as pH value, concentrations, exposure times and temperatures is therefore of crucial importance. The left part of Figure 8 shows an SEM image of the near-surface fracture of an incorrectly

coated magnet. Due to the BSE contrast (back scattering electrons), the secondary phase with the higher Nd content appears bright while the ϕ -phase ($\text{Nd}_2\text{Fe}_{14}\text{B}$) is shown darker. In the edge region of the microstructure (bottom of the image), a separation of the grains can be seen due to the absence of the grain boundary phase caused by corrosion. The right part of Figure 8 shows an EDX mapping of the same image section for the element neodymium and confirms the depletion of Nd-rich grain boundary phase in surface near areas caused during the passivation process.

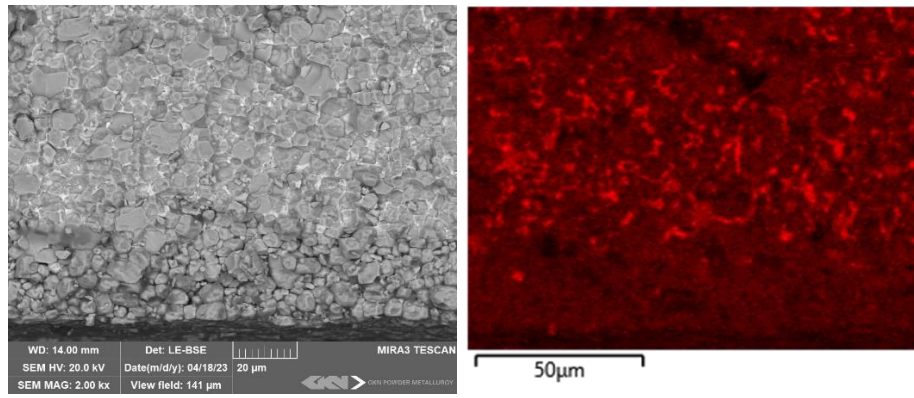


Figure 8: Fracture of surface-near area for incorrectly passivated magnet (left) and Nd distribution (EDX-mapping) of the same area (right).

If the process parameters for the coating are configured correctly, it is possible to passivate the surface without corrosive attack of the underlying material. Figure 9 shows the fracture structures in the surface area (left) and in the core area (right) of a correctly passivated magnetic sample. In the BSE images shown, a consistent distribution of the grain boundary phase with a high Nd content can be seen in all areas.

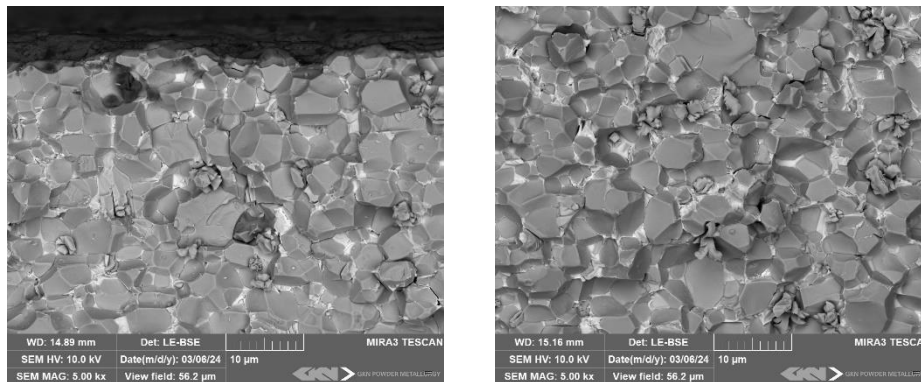


Figure 9: Fracture of surface-near area (left) and core area (right) for correctly passivated magnet.

Recycling

Considering the high demand for rare earth elements, the recycling of magnets is becoming increasingly important. Various material streams must be considered here. The materials produced during the manufacturing process from set-up operations and test components can generally be fed back into the process in a short-loop recycling cycle. The hydrogen decrepitation step and subsequent milling offer a simple way of returning scrap parts to the production cycle. Here, it is essential to ensure that the recycled material is sorted and free of impurities. In particular, increased oxygen levels or residues of additives used can significantly affect the purity of the material and make reuse impossible at least for high performance magnets.

If the recycled material streams have too high concentrations of impurities or are highly oxidized, complete oxidation of the material is unavoidable, which can usually be done by firing the material in air. In the subsequent recycling process separation of RE and iron oxides and, if necessary, separation of the various

RE oxides is inevitable. This long-loop process is significantly more complex and cost-intensive than the short-loop recycling described above. A similar recovery route would also be necessary for the recycling of end-of-life magnets, whereby one of the main challenges currently lies in the collection of these components and the creation of the necessary infrastructure. Moreover, in addition to the lack of sorting purity, other impurities in the material, consisting of residues from coatings or adhesives, must also be considered. Figure 10 shows an example of Nickel coated magnets before and after a hydrogen decrepitation step. Although it is possible to pulverize the NdFeB material, the coating residues remain and must be separated from the material at great expense. The complex topic of recycling is therefore currently the focus of several promising research projects that are investigating various process routes [9-11].



Figure 10: Nickel-coated magnets before (left) and after (right) hydrogen decrepitation.

Conclusion

Although the production of NdFeB magnets shows clear parallels to classic powder metallurgy, it involves a number of additional process steps. Throughout the manufacturing process, all process parameters must be aimed at the best functionality of the magnet material to ensure optimal magnetic performance. This paper provides an overview of the entire manufacturing process and illustrated some ways to define and optimize individual process steps, each of which has a substantial influence on the final magnetic properties and corrosion resistance.

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