DESIGN FOR RECYCLING OF NDFEB PERMANENT MAGNETS FROM ELECTRIC MACHINES

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Doctoral Dissertation
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IDEJNA ZASNOVA ZA RECIKLIRANJE TRAJNIH MAGNETOV NDFEB IZ ELEKTRIČNIH STROJEV

Doktorska disertacija

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To Mochi

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Abstract

This dissertation deals with increasing the recyclability of NdFeB-containing electric machines by design. Design for Recycling is re-defined in this work to comprise product design, process design and policy design to develop a coherent strategy for achieving high yields of good-quality material of recycled NdFeB at reasonable effort. Recycling of NdFeB is essential for three major reasons: To avoid the negative environmental effects of mining, to increase the resilience of the European rare earth permanent magnet supply chain and to utilize a source of material which is much closer to the desired composition than the mined minerals.

In the theoretical foundations and state of the art, NdFeB magnets are characterized in detail with an emphasis on sintered permanent magnets and the mechanisms of how the magnetic properties emerge to allow a better understanding of the effects of recycling-related changes in the material. Secondly, the recycling of NdFeB permanent magnets, with an emphasis on the short-loop route is explained, starting at the sorting and preprocessing and ending at resintering and postprocessing. Third, electric machines containing NdFeB are discussed to understand their variations and functions. The advantages and limitations of the use of NdFeB in electric machines are discussed. In the last sub-chapter, design for recycling is explained and defined. General and specific research on design for recycling and disassembly are presented and complemented by extensive patent research to show current recycling-friendly solutions.

In the practical investigations, three major studies are presented. The first study deals with in situ processing of electric machines, skipping the demounting and demagnetization and clarifies the possibilities and limitations by machine design. Furthermore, mechanical separation of contaminants like coating residues is discussed. The second study deals with another simplified processing route, using demagnetization to safely demount magnets and to combust adhesives and organic coatings, investigating the effects on the magnet materials and how to mitigate them. The third study complements the other two, investigating quantitatively what effects contaminants have on the magnetic properties at what level of contamination.

Finally, for implementation, a thorough analysis of the electric machine design from the housing down to the detailed magnet alloy is given. To make use of the work, a design for recycling guideline using a questionnaire and a conditional logic algorithm is designed and programmed to offer machine designers direct support to promote recycling-friendly design decisions. Finally, the creation of a technical specification within CEN/TC 472 on behalf of supporting the Critical Raw Material Act's Article 28 on labelling of magnet containing products is addressed. Leading this project allowed the partial implementation of the findings of this work at policy level, as the contents and technicalities of the label are defined there, which is meant to transfer the recycling-relevant knowledge from designers to recyclers.

Povzetek

Ta doktorska disertacija obravnava povečanje reciklabilnosti električnih strojev, ki vsebujejo magnete NdFeB, prek načrtovanja. V okviru raziskave je koncept »načrtovanja za recikliranje« na novo opredeljen tako, da vključuje načrtovanje izdelka, procesa in politike z namenom oblikovanja usklajene strategije za doseganje visokih izkoristkov kakovostnega recikliranega materiala NdFeB ob razumnem naporu. Recikliranje NdFeB je ključno iz treh razlogov: za zmanjšanje negativnih okoljskih vplivov rudarjenja, za povečanje odpornosti evropske dobavne verige trajnih magnetov iz redkih zemelj in za izkoriščanje vira materiala, ki je po sestavi bistveno bliže želeni končni zlitini kot primarni minerali.

V teoretičnih osnovah in pregledu stanja so magneti NdFeB podrobno opisani s poudarkom na sintranih trajnih magnetih in mehanizmih nastanka njihovih magnetnih lastnosti, da bi bolje razumeli učinke sprememb zaradi recikliranja. Nadalje je pojasnjen postopek recikliranja magnetov NdFeB s poudarkom na kratki zanki - od sortiranja in predobdelave do ponovnega sintranja in dodelave. Opisani so tudi električni stroji, ki vsebujejo NdFeB, njihove različice in funkcije ter prednosti in omejitve uporabe teh magnetov. Zadnje poglavje teoretičnega dela obravnava in definira načrtovanje za recikliranje. Predstavljene so splošne in specifične raziskave s tega področja ter analiza patentov, ki prikazuje obstoječe rešitve, usmerjene v večjo prijaznost do recikliranja.

V eksperimentalnem delu so predstavljene tri obsežne študije. Prva obravnava in situ obdelavo električnih strojev brez razstavljanja in razmagnetenja ter pojasnjuje možnosti in omejitve, ki jih določa konstrukcija stroja. Obravnavana je tudi mehanska odstranitev kontaminantov, kot so ostanki prevlek. Druga študija preučuje poenostavljeno procesno pot, ki uporablja razmagnetenje za varno odstranitev magnetov ter zgorevanje lepil in organskih prevlek; analizirani so učinki na magnete in možnosti njihove omilitve. Tretja študija kvantitativno analizira vpliv različnih kontaminantov na magnetne lastnosti pri različnih stopnjah onesnaženosti.

V implementacijskem delu je izvedena temeljita analiza zasnove električnega stroja – od ohišja do natančne sestave magnetne zlitine. Na podlagi ugotovitev je pripravljen in programiran priročnik za načrtovanje za recikliranje, ki z vprašalnikom in pogojno logiko neposredno podpira konstrukterje pri sprejemanju odločitev, prijaznih do recikliranja. Nazadnje je predstavljeno tudi oblikovanje tehnične specifikacije v okviru CEN/TC 472 za podporo izvajanju 28. člena Akta o kritičnih surovinah glede označevanja izdelkov, ki vsebujejo magnete. Vodenje tega projekta je omogočilo prenos rezultatov raziskave na raven politik, saj specifikacija določa vsebino in tehnične zahteve oznake, namenjene posredovanju informacij, ključnih za recikliranje, od načrtovalcev k reciklerjem.

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Abbreviations

AC	...	Alternating Current
AES	...	Auger Electron Spectroscopy
AFM	...	Axial Flux Machine
BEV	...	Battery Electric Vehicle
BLDC	...	Brushless Direct Current (Machine)
BSE	...	Backscattered Electron Mode
BSE COMP	...	Backscattered Electron Mode Compositional Mode
CAD	...	Computer-aided design
CEN	...	European Committee for Standardization (Comité Européen de Normalisation)
CRMA	...	Critical Raw Materials Act
CS	...	Carbon-Sulfur Analysis
D4R	...	Design for Recycling
DC	...	Direct Current
DG	...	Directorate-General
DIN	...	German Institute for Standardization (Deutsches Institut für Normung)
EC	...	European Commission
EDS	...	Energy Dispersive X-ray Spectroscopy
EELS	...	Electron Energy Loss Spectroscopy
EESM	...	Externally Excited Synchronous Machine
ELV	...	End of Life Vehicles
EMF	...	Electromotive Force
EOL	...	End of Life
ESPR	...	Ecodesign for Sustainable Products Regulation
EU	...	European Union
EV	...	Electric Vehicle
GBD	...	Grain Boundary Diffusion
GBE	...	Grain Boundary Engineering
GROW	...	Directorate General for Internal Market, Industry, Entrepreneurship and Small and Medium Enterprises
HDD	...	Hard Disk Drive
HDDR	...	Hydrogenation Disproportionation Desorption Recombination
HEV	...	Hybrid Electric Vehicle
HPMS	...	Hydrogen Processing of Magnetic Scrap
HRE(E)	...	Heavy rare earth (elements) including terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), lutetium (Lu), scandium (Sc), and yttrium (Y) [1]
ICEV	...	Internal Combustion Engine Vehicle
ICP-OES	...	Inductively coupled plasma - optical emission spectroscopy
IM	...	Induction Machine
IPM	...	Internal Permanent Magnet (Machine)
IPMSM	...	Internal Permanent Magnet Synchronous Machine
IPS	...	International Postgraduate School
IR	...	Infrared Radiation or Inner Rotor
JRC	...	Joint Research Centre

JSI	...	Jožef Stefan Institute
LCA	...	Life Cycle Analysis
LRE(E)	...	Light rare earth (elements) including lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd) and promethium (Pm) [1]
MRE(E)	...	Medium rare earth (elements) including samarium (Sm), europium (Eu) and gadolinium (Gd) [1]
NDIR	...	Non-dispersive Infrared Radiation
ONH	...	Oxygen-Nitrogen-Hydrogen Analysis
OR	...	Outer Rotor
PLM	...	Product lifecycle management (system)
PM	...	Permanent Magnet
PMSynRM	...	Permanent Magnet (assisted) Synchronous Reluctance Machine
QR	...	Quick Response (Code)
REE	...	Rare earth element
REPM	...	Rare Earth Permanent Magnet
RM	...	Reluctance Machine
RMIS	...	Raw Material Information System
SCIM	...	Squirrel Cage Induction Machine
SE	...	Secondary Electron Mode
SEM	...	Scanning Electron Microscopy
SIMS	...	Secondary Ion Mass Spectrometry
SM	...	Synchronous Machine
SMC	...	Soft Magnetic Composite
SPM	...	Surface Permanent Magnet (Machine)
SPMSM	...	Surface Permanent Magnet Synchronous Machine
SUV	...	Sports Utility Vehicle
SynRM	...	Synchronous Reluctance Machine
TC	...	Technical Committee or Thermal conductivity
TEM	...	Transmission Electron Microscopy
TS	...	Technical Specification
WEEE	...	Waste Electrical and Electronic Equipment
WG	...	Working Group
WRIM	...	Wound Rotor Induction Machine
XPS	...	X-ray Photoelectron Spectroscopy

Chapter 1

Introduction

The rare earth elements have been deemed as critical and strategic raw materials by the United States of America, the European Union, Japan, and even China, their main producer, for more than a decade [2], which is due to their importance for several key technologies, as illustrated in Figure 1.1. This distinct group of chemical elements usually encompasses the Lanthanide series as well as Scandium and Yttrium. Due to their unique electron structure, having a partially filled 4f orbital, they show very similar chemical properties, but highly distinct physical properties, especially regarding magnetism, making them extremely challenging to separate and to replace [3].

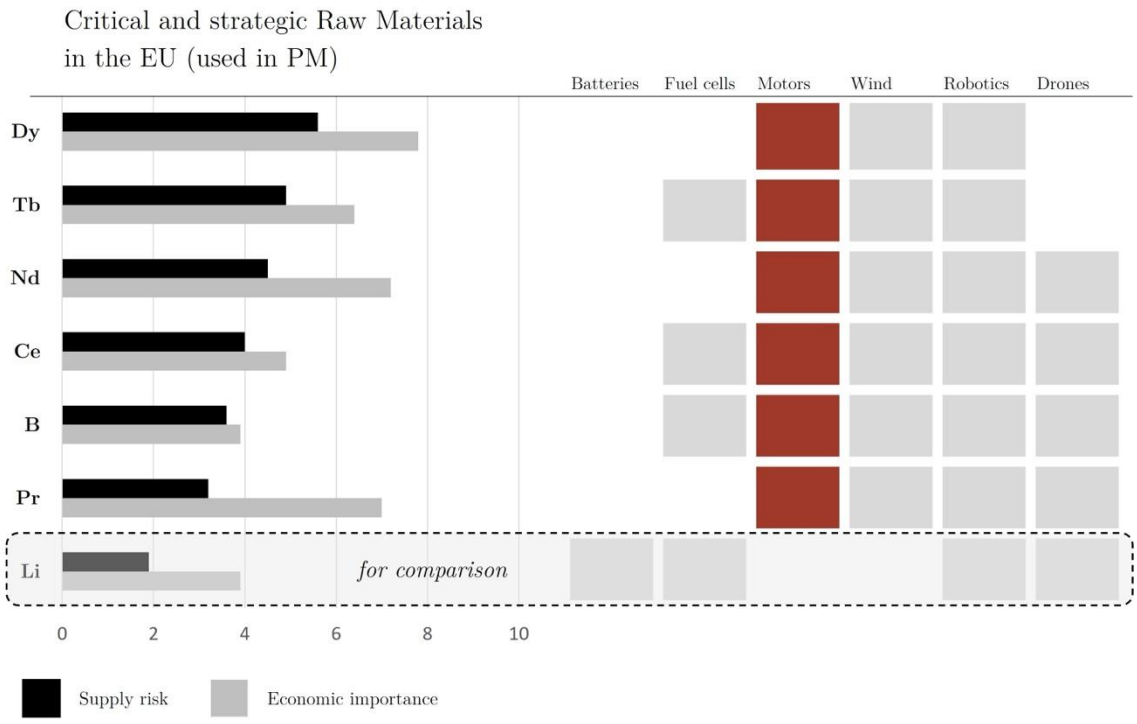


Figure 1.1: Strategic and critical raw materials required for NdFeB permanent magnets in comparison with Lithium. Based on data from the Raw Material Information System (RMIS) [4].

The Joint Research Center's Raw Material Information System states that Ce, Dy, Gd, Nd, Pr, Sm, and Tb, which are all used for permanent magnets, are on the proposed list of Strategic Raw Materials of the European Union, citing a 100 % import reliance [5].
In the 2024 Critical Raw Materials Act (CRMA), ambitious goals of producing 10 %, processing 40 %, and recycling 25 % of strategic raw materials in 2030 were set to combat

this high reliance [6]. As shown in Figure 1.2, most REEs are used in permanent magnets; and most permanent magnets are used in electric machines [7], highlighting the importance of establishing a circular economy for rare earth permanent magnets, specifically, NdFeB from electric machines.

Electric machines generally encompass motors, generators, and combinations of both, like they are used in hybrid electric vehicles [8]. They are applied in numerous key applications relevant to the green transition and digital transformation [9]. Ranging from miniature motors built into smartphones to applications in household appliances, industrial robots, electric vehicles, and wind turbines for energy generation, they are essential for modern living and industry.

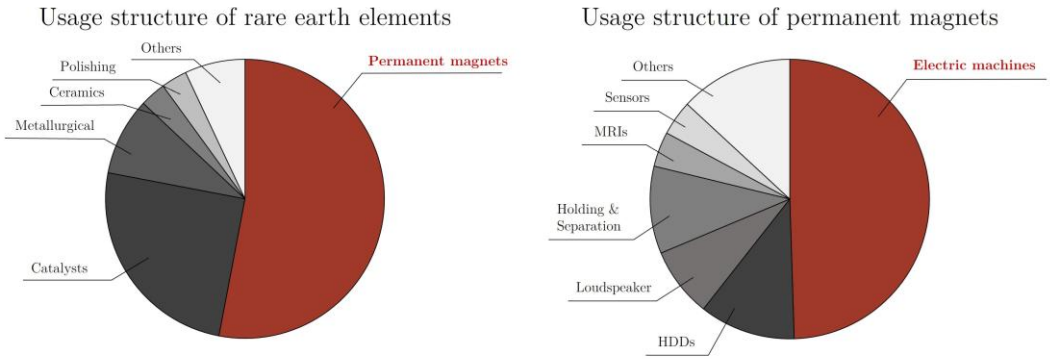


Figure 1.2: Relative demand of REE and permanent magnets per application. Based on data from RMIS [5] and Cui et al. [7].

It is essential to avoid environmentally and socially harmful mineral extraction as much as possible and to implement design for sustainability concepts for the aforementioned products. McAloone and Bey [10] state that around 80 % of a product's lifetime environmental impact can be determined during the conceptual stage.

However, the design for recycling NdFeB from electric motors is not considered a viable task in some literature sources. One guide towards design for recycling recommends not using permanent magnets at all, as they complicate recycling [11]. Another source explains that the design for recycling of electric machines seems “far-fetched” due to the narrow requirements caused by space restrictions and all adjacent components of the mechanical systems around them, as their design appears to be the priority of most designers [12].

The goals of machine design are already complex and varied: a high torque density and high efficiency over the whole speed range, a long lifetime, high power factor, low noise level, ease of control, and the possibility of field weakening, all while maintaining low material and manufacturing cost [12]. Adding recyclability-related requirements to this long set of prerequisites is complicated, even if designers are properly sensibilized and willing to adjust [13].

Furthermore, the majority of electric vehicles, which are suspected to be the most abundant NdFeB users in the future [14, 15] use either internal permanent magnet synchronous motors or permanent magnet assisted synchronous reluctance motors [16]. Both types have internal permanent magnets in the rotor, making them relatively challenging to extract compared to other permanent magnet-driven machines. The promising news, however, is that there has been empirical demonstration that electric machines equipped with recycled NdFeB magnets do not necessarily fall behind those using virgin magnets [17, 18].

The presented combination of a monopolized, insecure supply chain, strategic and environmental importance, and significant issues with recyclability calls for the

development of a thorough and sensible design for the recycling concept. The current situation's uncertainty allows for a high level of flexibility: As supply chains for recycled NdFeB material are being developed momentarily, the recycling processes, particularly the pre-processing stages, are not yet fully established. Specific design for recycling rules for NdFeB in electric machines are not currently available and European policymakers are working intensively on the topic. Therefore, a concept can be developed sensibly around an existing, highly variable, and highly established technology like electric machines, encompassing product design, process design, and policy design.

What is the ideal design for recycling NdFeB from electric machines?

While this question cannot be answered clearly, an approach to the answer could be: The one that is applied.

Some attempts to achieve 100 % recyclability have served as "interesting curiosities" [19]. Therefore, flexible approach that can be tailored to different designs is prioritized in this work, which is aiming to integrate design for recycling in a realistic and approachable manner, considering both ecological and economic factors. In order to design the desired flexible set of recommendations, it is important to take into account a large amount of variables. Furthermore, due to electric machines being a mature, highly diversified and performance-driven application and the recycling infrastructure being still under development, it is worth investigating the design feature in context with different, ideally simplified processing routes. This necessitates focusing on under-researched factors as well as on those design decisions which affect the efficiency of the recycling process and the quality of the recycled material the most. In this work, a combined design for recycling strategy taking into account leverage from the sides of product design, process design and policy design, is developed and presented to ensure efficient, high-quality recycling of NdFeB permanent magnets from electric machines.

Chapter 2

Motivation

Aside from NdFeB permanent magnet recycling being a highly interesting process, some aspects can be highlighted as key motivations to improve the recycling rates, efficiency, and quality.

On a global scale, the environmental and social effects of rare earth primary mining and processing should not be underestimated. The European Union also has a strong interest in ensuring the resilience of the supply chain, especially following the rare earth crisis in 2010 – 2011 [20]. Another reason for concern is the highly monopolized production and processing of rare earth elements [21, 22], which recently (2025) led to the threat of extreme export restrictions. Lastly, due to the natural composition of the mined ores, the supply of more scarce elements, which are relatively higher in demand, results in an oversupply of other, more abundant rare earth elements. In contrast, in the "urban mine", which holds scrap and end of life NdFeB permanent magnets, the balance of available elements is already close to the desired relative amounts.

2.1 Environmental and Social Issues

The detrimental effects of rare earth mining have been extensively documented in the media and consistently scrutinized by the public.

Common health problems reported near REE mining sites are diabetes, osteoporosis, respiratory diseases, as well as the irritation of skin, eyes, and the gastrointestinal tract, as well as birth defects and leukemia [23, 24]. The dust emitted from the mine can contain airborne radioactive elements [14] aside from the rare earth dust, which by itself is considered biopersistent and can cause organ damage, especially in the respiratory tract [25]. In the vicinity of the well-known Bayan Obo mine, farming practice ceded strongly, and the population of nearby villages faced a decline of almost 90 % [26]. In a 2019 social life cycle analysis (LCA), not only Bayan Obo, but also the Australian Mount Weld mine performed very poorly [27]. However, compared to the mining of ionic clays, classical mines are seen as less problematic. Ionic or ion absorbed clays are usually mined using so-called "slash and burn" leaching ponds, where the REE are leached in situ in the ground and oftentimes left behind afterwards [14, 28]. This impairs the integrity of the ground, provoking landslides while poisoning the groundwater and the soil [28]. Figures 2.1 and 2.2 visualize the effects of mining and the processing facilities for REE.

As the main mining area of REE until the early 2010s, China has invested tremendous sums into cleaning up the damage caused by the REE sector. Originally, 4 billion Yuan ($600 million) were foreseen when Chinese authorities stated that they are not willing to sacrifice the environment to develop the REE industry [26]. In a 2019 report, however, it was stated that the environmental clean-up bill for southern Jiangxi province alone would

amount to around 38 billion Yuan, according to China's Ministry of Industry and Information Technology [29]. Recently, rare earth mining via ionic clays has been transferred to other countries like Myanmar, where local residents experience the poisoning of their crops, livestock, and water, frequent landslides from mining practices, and violent displacement by paramilitary groups [28]. In the Mountain Pass mine in California, a federal-level investigation found a total of 60 spills of wastewater between 1984 and 1998, contaminating the already scarce groundwater under the desert floor [26].

Another issue is radioactive elements, which are often mined in conjunction with rare earth-rich ores, such as thorium and uranium [26]. Wastewater containing radioactive sludges, heavy metals, and toxic waste from processing chemicals is emitted into the groundwater [14]. Illegal mining practices aggravate the social and environmental problems associated with REE mining. For instance, in 2016 it was reported that illegal mining output in China exceeded the output of the major non-Chinese competitors and amounted to around 40 % of domestic Chinese REE production or 30 % of the global production [23]. Unsurprisingly, the working conditions and environmental protection in illegal mining activities are subpar to regulated production [23]. Counteractively, recent legal measures to combat illegal mining and trade, and to ensure safer working conditions, have been implemented in China [30].

Overall, while efforts to alleviate the impact of primary production and diversify the primary production supply chain through mining projects worldwide are underway [31], the production processes themselves should not be underestimated, both in terms of their environmental impact and the required expertise. The mining and processing of rare earths is notoriously complex, especially due to the REE's chemical similarity. The process is summarized by McNulty et al. [32] and Sprecher et al. [33]: The mined ores are first beneficiated into rare earth oxide concentrates by removing undesired ore constituents by utilizing the physical and chemical differences between the gangue (the to-be-removed minerals) and the rare earth minerals. The gangue is sent to tailings, where it is either processed separately or discarded as waste. With roasting and hydrometallurgical treatment, the REE minerals are "cracked", leached, and solvent-extracted, encompassing a multitude of different steps to produce REE oxides. Finally, the oxides are reduced to REE metals, which is also a challenging process, as REE are highly reactive and attract elements from the processing equipment, such as crucibles, which makes achieving high purity very challenging. For magnet production, this process is followed by alloying, strip casting, hydrogen decrepitation, powder preparation, aligning and pressing, sintering, grinding and slicing, passivation, coating, and finally, magnetization.



Figure 2.1: A toxic lake of mine and tailings in Baotou. Reprinted from Toby Smith /Unknown Fields. Permission kindly given by Toby Smith. www.tobysmith.com



Figure 2.2: Facilities for REE processing in Baotou. Top: Blast furnace. Bottom: Stage 1 Refinery. Reprinted from Toby Smith /Unknown Fields. Permission kindly given by Toby Smith. www.tobysmith.com

2.2 Resilience of the Supply Chain

Transitioning to a green economy requires the implementation of large-scale changes in power supply and in the design of numerous everyday products, which rely on technologies that involve critical raw materials [34]. These materials, including rare earths, which are deemed especially critical, face a high risk of supply disruption [34].

Between 2009 and 2012, a crisis occurred in the supply of rare earth elements, as China set tight export quotas to fulfill its rising domestic demand [21]. This led to a limited availability and a sharp price increase for its international supply [35, 36]. In 2013, just after the rare earth crisis, China still produced more than 90 % of all rare earths worldwide, while only having 40 % of the confirmed geological reserves [14, 21]. In 2020, it was estimated by the European Commission that the European demand for rare earths will increase tenfold by 2050 [37], multiplying the risk of supply chain disruption. Recent data showcasing the market dominance of China is shown in Figure 2.3.

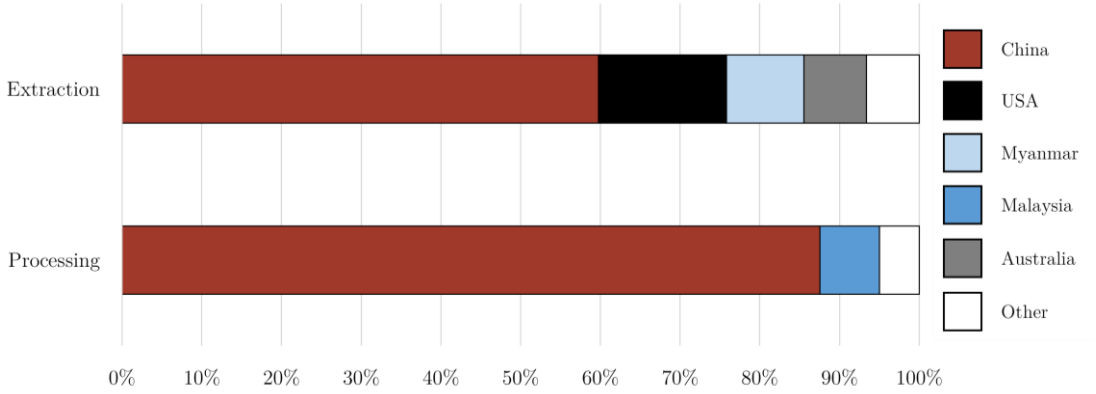


Figure 2.3: The scope of China's monopoly on rare earth extraction and processing set into perspective of the worldwide volumes. Based on data from RMIS [5].

The reason for this extreme market dominance is found in the specialization of the rare earth extraction and downstream mining activities [21]. This led to increasing efforts in Western countries to both mine and process rare earth elements, as well as recycling REE-containing materials like rare earth permanent magnets (REPM). A secondary material supply from anthropogenic sources is especially important for regions with no rare earth mining [15].

2.3 Balance Problem

Another dimension of the supply issues is the so-called “balance problem”. This refers to the issue that mining the desirable elements, which are required in much higher proportions than others, results in other, less desirable elements being co-mined, often in much higher amounts [38, 39]. The rare earth-containing minerals mined most commonly are bastnäsite, monazite, and ionic clays [31]. The distribution of REE in commonly mined ores is compared to the demand and average magnet composition in Figure 2.4.

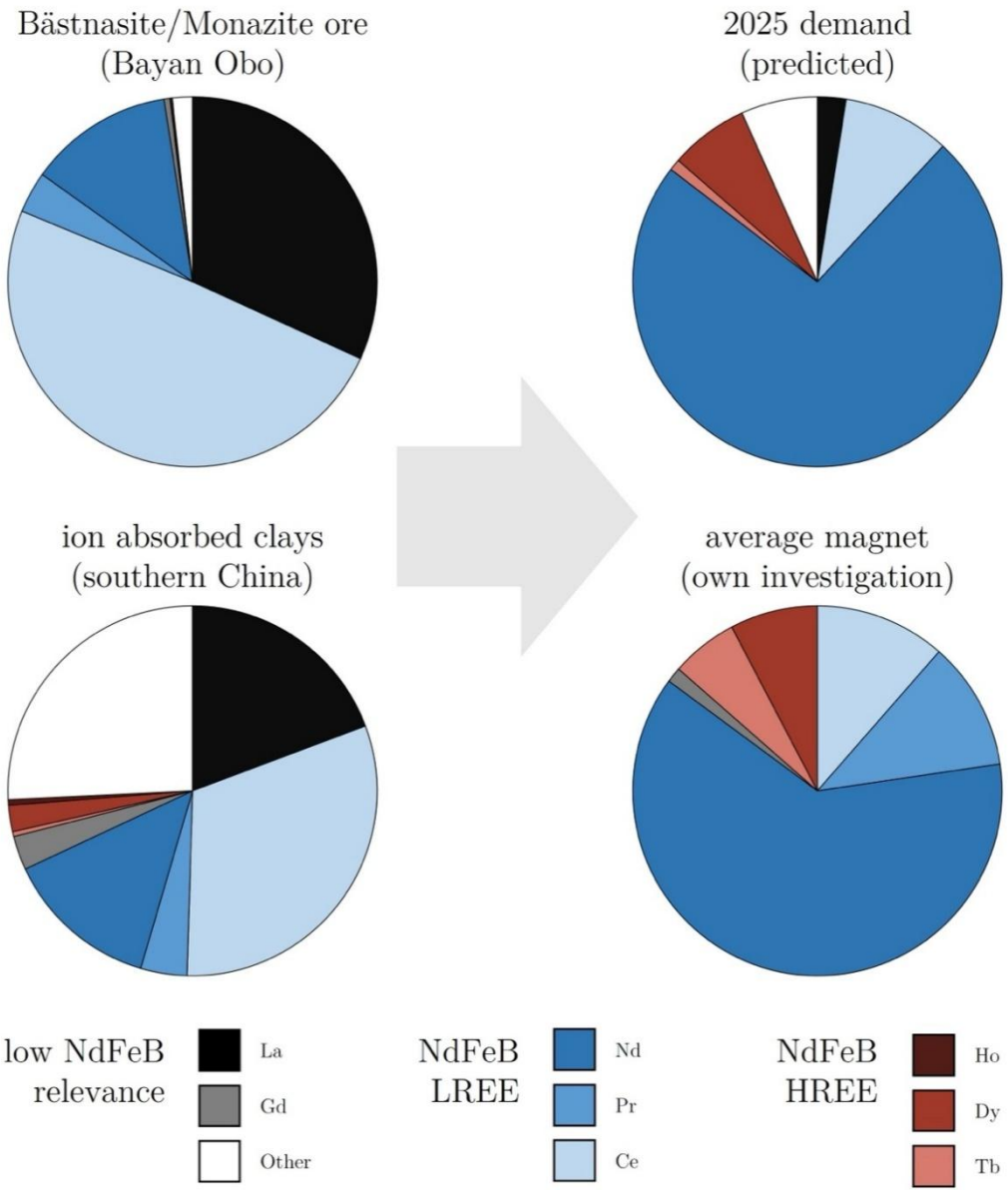


Figure 2.4: Comparison of the distribution of magnet-relevant REE in common primary sources (left) as reported by Eggert et al. [40], versus the demand (right), which is represented by a prediction for 2025 by Zhou et al. [41] and the average magnet composition measured in 196 NdFeB magnets at the Institute of Strategic Precious and Technology Metals, summarized by the author.

Looking at Figure 2.4, the advantage of using pre-separated and pre-alloyed anthropogenic sources rather than virgin material becomes evident. In fact, even countries like China, which are rich in REE reserves have started to look into recycling even before the rare earth crisis in 2010 - 2011 [42]. Nonetheless, it is clear that regions without exploitable geological REE reserves are more dependent on recycling to secure a stable supply chain in the future [15], which is why REPM recycling has received significant scientific interest and public funding in the last decade.

Chapter 3

Strategy and Hypotheses

Classically, Design for Recycling is understood as an approach to alter product design in a way that considerations regarding the recyclability are made during conceptualization. As mentioned, 80 % of the environmental impact is decided in the concept creation according to McAloone and Bey [10], which underlines clearly that the long-term solution of adapting product designs from the earliest stages of product development is worth the effort.

For this, a suitable guideline for enhancing the recyclability of NdFeB from electric machines, should be provided as a part of this dissertation. However, while specific research on how to deal with NdFeB or permanent magnets in design for recycling is very sparse, significant amounts of research has already been conducted on all kinds of products generally, especially regarding disassembly. Therefore, the focus should be on NdFeB or permanent magnet specific aspects.

During the preparation of the thesis, the opportunity arose to also engage in policy design regarding the labelling of permanent magnet-containing products by preparing a technical specification within the standardization committee CEN/TC 472. Here, the operational lead of a group of 70 stakeholders from industry, academia and policy was taken. The technical specification aims to support the creation of the implementing act for Article 28 of the European Critical Raw Materials Act [6].

As illustrated in Figure 3.1 it is important to understand that design for recycling on the product level or labelling only yields the desired results after the average lifetime of the redesigned product, which can range from approximately 2 years to 30 years [39].

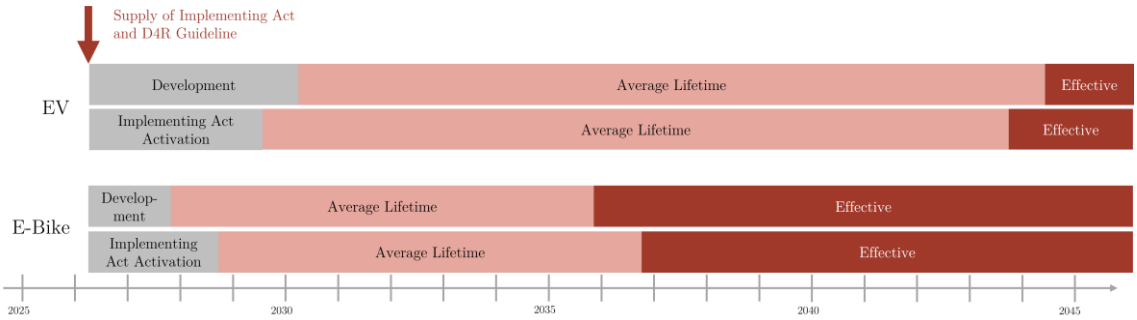


Figure 3.1: The timeline of product design- and labelling related efforts and subsequent results of improved recycling rates.

While product design guidance as mentioned above should certainly not be neglected, design for recycling should also comprise short-term solutions, which are mostly focused on process design and processing the oftentimes less-than-ideal current designs. The cooperation of stakeholders in end of life management and product designers is imperative,

which has also been stated by Li et al. [43] regarding implementing a circular economy for electric motors in electric vehicles.

Thus, the overall strategy becomes clear: For achieving ideal recyclability, specific long-term solutions based on a tailored product design guide and support of policy design are required. In the meantime, to ensure the intermediate supply of recycled NdFeB, to fulfill the European goals outlined in the CRMA [6] and to support the scale-up of recycling facilities in Europe, recyclers should be supported with research data. This data should be translated into implementable, realistic process design recommendations. The overall, simplified strategy is summarized graphically in Figure 3.2, where the areas are linked to the corresponding sections of the document.

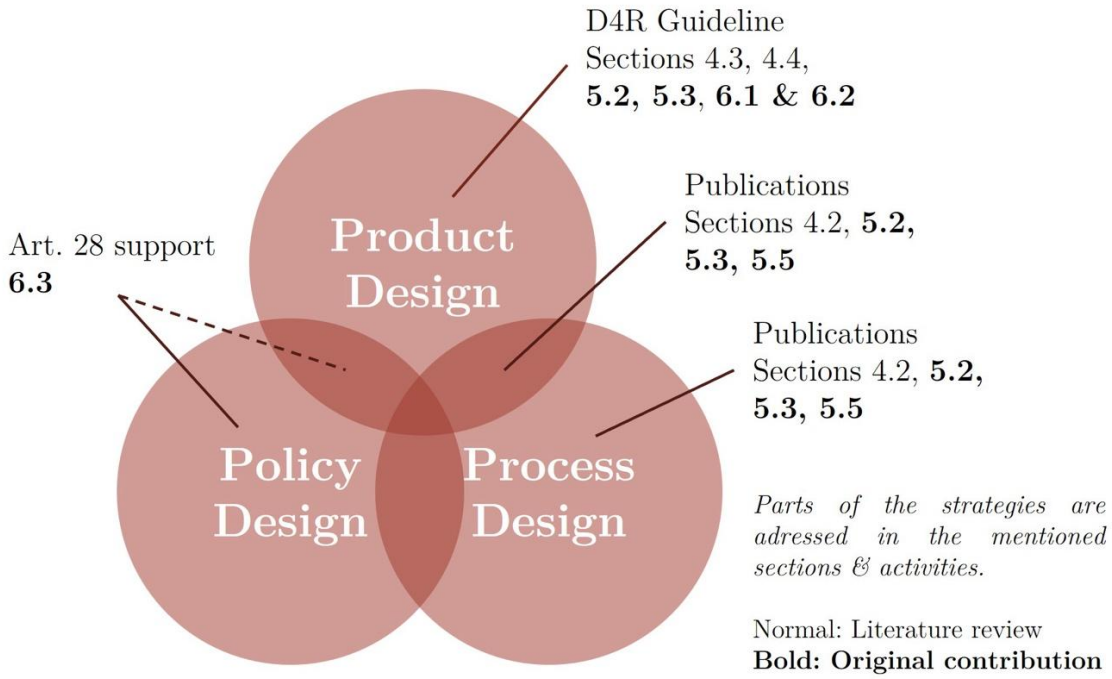


Figure 3.2: Visualization of the overall strategy to improve the recycling rate of NdFeB via product design, process design and policy design.

Another important annotation at this point is that the research of the thesis is aimed at recycling via the patented method of hydrogen processing of magnetic scrap [44, 45] as it is currently the most straight-forward and ecological method [46–48]. Other recycling methods should be employed when the material is unsuitable to be recycled via HPMS, where purification is limited. However, part of this work is to avoid the deterioration of material so most of it can go into a regular HPMS process.

The practical research to fill the gaps regarding product design features and their processibility, giving guidance to product and process designers alike, is based on the following hypotheses:

Good recyclability is achieved with suitable combinations of product features and process features – the suitable product features strongly depend on the process.

This is a likely correct assumption made by product designers – to know how to design, they must know, what processes they are designing for. On the other hand, recyclers will

base their processes on the product designs. If this hypothesis proves wrong, there is one ideal design to work with. If it is right, a design for recycling guideline must be adaptable to flexibility on both sides.

Processing of rotors without demagnetizing and disassembling the magnets is viable in most cases. The removal of coating and adhesive residues from powder is viable in most cases.

Literature shows that it is possible in at least some cases [49, 50]. It would be very beneficial to skip demounting and demagnetizing of magnets from rotors, especially for smaller assemblies. However, as it is also known that the reaction on which the HPMS process is based shows a significant volume expansion [39, 51], it is possible that it does not work with some assemblies. Another factor are possible contaminants from coatings and adhesives, which may or may not be removable from the powder, however, there is also promising information in research and patents [49, 52]. This would be especially helpful for internally mounted magnets.

Thermal demagnetization can be a means to disassemble and demagnetize adhesive-fixed magnets, including segmented magnets.

When organic substances are heated above the thermal demagnetization temperature of NdFeB, they can decompose. If the right temperature, time and atmosphere are provided, a nearly complete combustion should be possible. Through these conditions, adhesive connections should be broken. This would be especially helpful for surface mounted magnets.

The separability of magnet material and nearby other materials is essential for achieving high quality recycled material. However, for all contaminants, there should be an acceptable limit whereas perfect separation is not necessary.

This should be validated by testing the effects of contaminants from adhesives, coatings and corrosion. The need for finding acceptable limits was already identified in earlier publications by the author's research group [53].

Suitable labelling and provision of information on the to be recycled magnets is superior to testing and the time and money required for labelling and reading the label are well invested.

This is mostly an economic consideration, as it depends on whether all relevant features of the magnet and the assembly can be detected quicker than a label could be read. Labelling has been explored in earlier publications by the author's research group within the MaXycle project [54] and the 2024 CRMA mandates the implementation of a labelling system [6].

Chapter 4

Theoretical Foundations and State of the Art

4.1 NdFeB Permanent Magnets

This sub-chapter serves to establish an understanding of NdFeB permanent magnets and their magnetic properties, which are essential to retrace the effects of design for recycling related decisions: Design for recycling does not only influence the efficiency and viability of the recycling process itself, it also serves to ensure the quality of the recycled material. Therefore, it is vital to understand how a NdFeB magnet with competitive quality is manufactured, what distinguishes it, and what may impair its quality. Given that contamination is a significant issue in NdFeB recycling, it is important to explain the mechanisms that link the introduction of contaminants to the loss of magnetic properties.

4.1.1 Characterization, application, and history

4.1.1.1 Permanent magnets: applications and types

Permanent magnets are commonly used in applications where energy conversion between mechanical and electrical forms is needed, such as motors and generators. Both types are included under the umbrella of electric machines, along with combinations of both (e.g. traction motors which are also used to regenerate energy from braking). Electric machines are required in applications like wind turbines, electric vehicles, e-bikes and scooters, industrial robots, white goods, power tools, and consumer electronics, among others. Magnets are also used for reading and writing to electromechanical data storage devices such as hard disk drives. Acoustic technology uses magnets to produce defined vibrations in speakers. They are needed in many sensor applications, ranging from magnetic resonance imaging in medical technology to analytical chemistry. Just like they are used as toys, to fix postcards on the fridge, or for the smooth closure of handbags or dishwashers. The placement of NdFeB on the global market and its magnetic properties are shown in Figure 4.1.

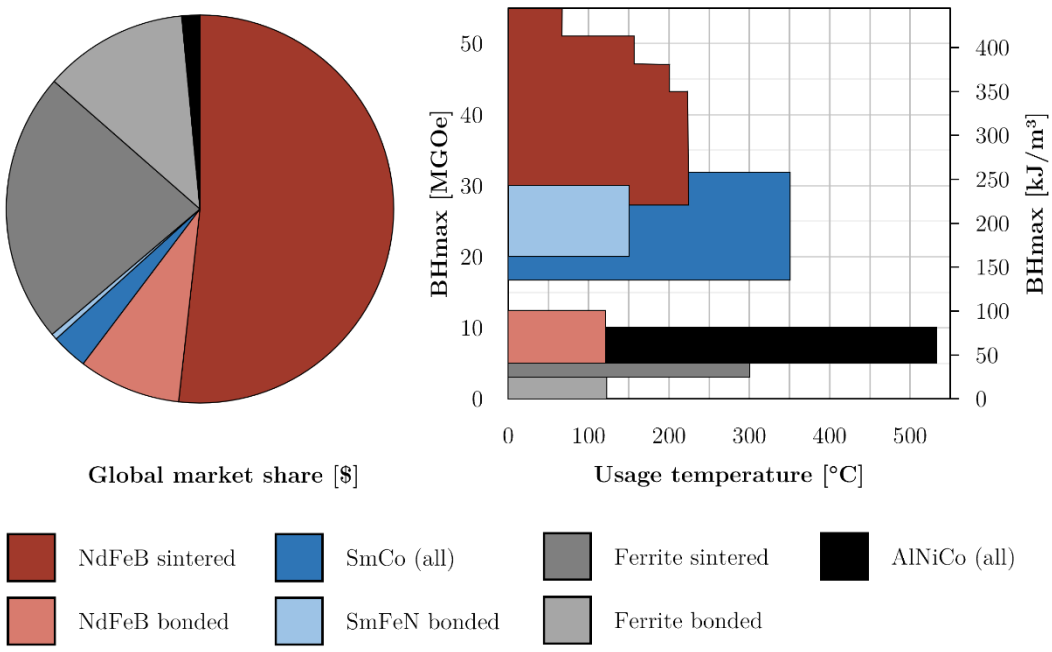


Figure 4.1: NdFeB permanent magnets in context with other permanent magnets. On the left, the market volumes ($) of different permanent magnets are given according to Coey [55]. Mind that in terms of mass, more ferrites are on the market than the significantly more expensive NdFeB. On the right, the maximum energy product and usage temperatures of common permanent magnet materials are compared.

NdFeB permanent magnets, often called Neodymium magnets are the strongest commercially mass-produced permanent magnets with a maximum energy product of up to 440 KJ/m³ for sintered NdFeB. This significantly sets them apart from other magnet types. However, the caveat of using NdFeB is its relatively low use temperature.

This is shown in Figure 4.1, where the market shares of the different magnet types are shown along with their maximum energy product and usage temperature ranges.

NdFeB is produced using several methods, including sintering and polymer bonding, which are the most common ones. Due to the modifications of the microstructure, the doping of the base alloy, and different manufacturing methods, different magnet grades can be achieved.

4.1.1.2 History of the development of NdFeB

Two unrelated research teams developed NdFeB at around the same time: Sumitomo Metals, led by physicist Masato Sagawa, who advanced research on sintered NdFeB magnets [56], and General Motors, which focused on melt-spun nanocrystalline NdFeB used in bonded magnets [57].

These simultaneous discoveries were driven by the need for a more cost-effective alternative to the SmCo magnets, which were the “gold-standard” magnets at the time, however, with a “gold-standard” price as well. Compared to SmCo, which was developed around 1960, NdFeB has higher remanence but a significantly lower Curie temperature and lower tolerance to higher operating temperatures.

Due to eddy currents and friction in electric machines, which cause the magnets to heat above a tolerable level for the base alloy, strategies were developed to create higher-coercivity, temperature-resistant alloys. These strategies include, e.g., alloying with heavy

rare-earth elements like Dy and Tb, improving the coercivity-related microstructure by annealing, and, more recently, using special manufacturing techniques such as segmenting [58]. Due to the constrained supply of heavy rare earths, grain boundary diffusion (GBD) was introduced as a method to locally reinforce coercivity, requiring less heavy rare earth content when combined with the diffusion treatment [59–61].

Furthermore, the technology for aligning, pressing, and sintering has evolved over time, including improvements in atmosphere control, to create “cleaner” magnets with fewer defects. Another field of research is alternative magnet types, such as nanocrystalline exchange-coupled magnets [62]. Further, significant amounts of research have been conducted on NdFeB recycling recently due to the rare earths being classified as critical raw materials.

4.1.1.3 Magnet grades and qualities

NdFeB permanent magnets are available in different magnet grades, which depend on the specific alloy used and microstructural parameters. The magnet grade, though, does not provide any information about these; instead, it is merely indicative of the maximum energy product of the magnet, expressed in MGOe (Mega Gauss Oersted), which describes the magnet's strength relative to its volume, and the heat resistance. There is no information on other essential properties, such as corrosion protection or the magnet's manufacturing methods used.

The nomenclature, e.g., is N42SH for a NdFeB magnet (N) with a maximum energy product of 42 MGOe (334 kJ/m^3) and an operating temperature of 150 °C (SH).

Different applications of NdFeB require different magnet grades, depending on the design specifics, e.g., rotational speeds in electric machines, cooling, magnet placement, and possible magnet volume.

Approximate magnet grade ranges for different applications are shown in Figure 4.2, where data from various manufacturers and own measurements are compiled and compared to example applications [63].

It is important to note that exceptionally high magnet grades, both in terms of energy product and heat resistance, usually require higher heavy rare-earth contents. The NdFeB-relevant light rare earths (LRE(E)) include La, Ce, Pr, and Nd, and the heavy rare earths (HRE(E)) are Gd, Tb, Dy, and Ho [1].

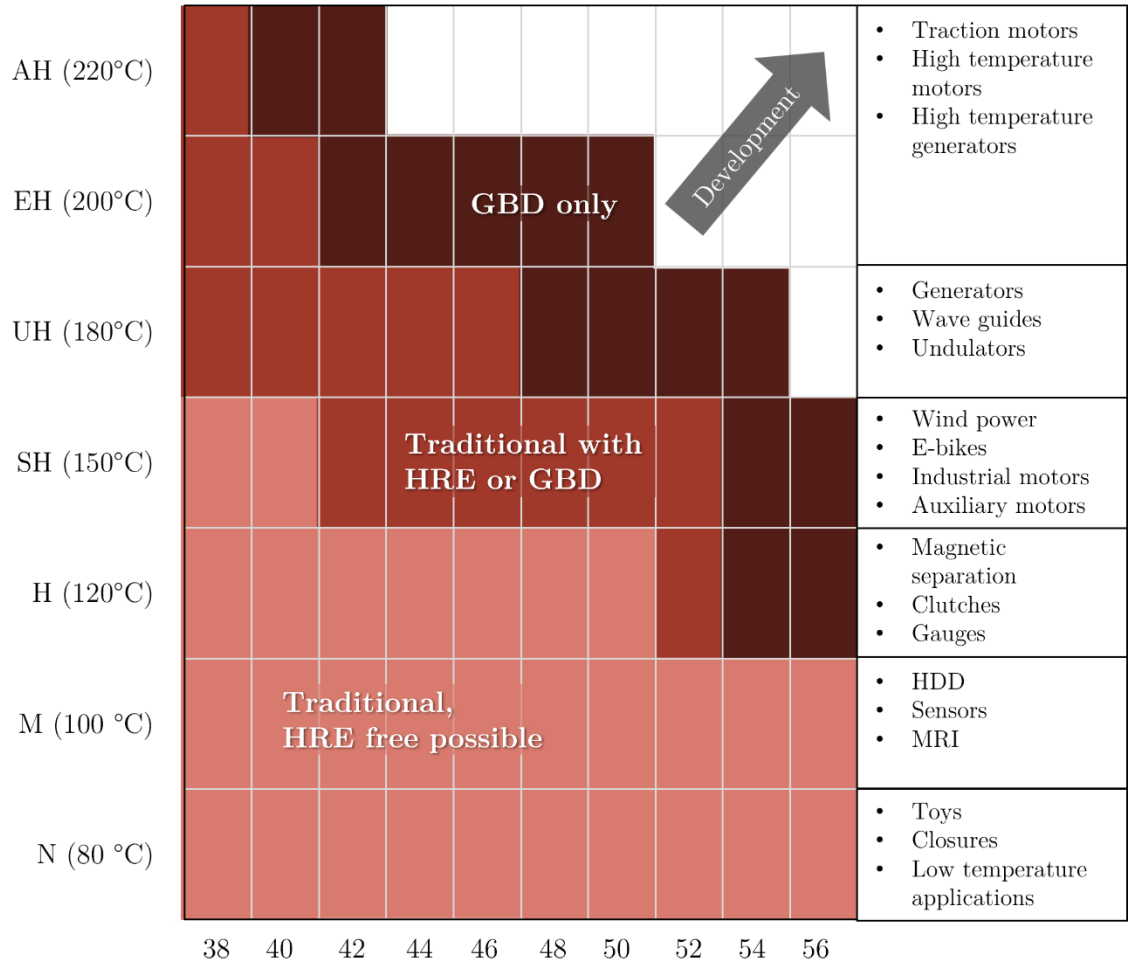


Figure 4.2: Available magnet grades according to own market research, complemented with typical applications according to the US Department of Energy [63]. "HRE" refers to heavy rare earth addition into the alloy, "GBD" refers to reduced heavy rare earth addition by grain boundary diffusion.

4.1.1.4 NdFeB magnets sub-types

As indicated in Figure 4.1, NdFeB magnets can be produced in different ways, leading to drastically different properties. The main types used commercially are sintered and bonded magnets.

Sintered NdFeB is produced by using single-crystal anisotropic NdFeB powder and some extra neodymium to form the intergranular phase. Before sintering, the powder is formed and aligned. This may be performed, e.g., by pressing for simple shapes or by the shaping-debinding-sintering process, where, e.g., metal injection molding or extrusion is used to shape the magnet. The polymer binder used for these processes is removed again before sintering, ensuring the powder compact retains its shape [64]. Sintered NdFeB will be explained in greater detail in 4.1.2. Bonded NdFeB magnets, also called polymer-bonded magnets, are produced by combining melt-spun NdFeB ribbons with a polymer resin,

mostly about 10 wt.% epoxy resin [21, 57]. While these magnets do not exhibit magnetic properties comparable to those of sintered magnets, they are mechanically flexible and can be produced in many shapes while still providing a higher maximum energy product than ferrites [65]. Hot deformed NdFeB magnets use the same kind of melt-spun material, which is essentially nanocrystalline. The material is cold-pressed, then hot-pressed, and finally hot-deformed, all at around 800 °C, to achieve a fine-grained, anisotropic body [66]. As they currently offer lower coercivities than sintered magnets with higher process complexity and cost, they are used mostly for net-shape manufactured, lightly flexible magnets [65]. Spark plasma sintering, or field-assisted sintering of NdFeB, is a modified hot-pressing technique that uses a pulsed direct current and applies pressure to a cold-pressed part, offering short processing times [67]. Various methods to 3D-print both bulk and composite NdFeB are reported, some examples are selective laser melting of NdFeB powder [68], digital light processing with a photosensitive resin and NdFeB powder [69], filament printing with a NdFeB-polymer material [70], and direct-write 3D-printing with an epoxy-based NdFeB ink [71].

4.1.2 Sintered NdFeB permanent magnets

Sintered NdFeB magnets are the central material to be recycled in this work; therefore, they are characterized more closely with regard to their production processes, phases, and magnetic properties. This section should be prefaced by clarifying that "NdFeB" should be seen as a colloquial name as it does not reflect the stoichiometry of the overall material but is rather a simplification of varying elemental compositions both between different NdFeB magnets and between different phases of the material. Furthermore, as the $Nd_2Fe_{14}B$ intermetallic compound can also be formed with other rare earths and since it is standard practice to partially substitute Nd with other rare earth elements like Pr, Dy, Tb or Ce among others, "$R_2Fe_{14}B$" refers to such alternative compounds in the following.

4.1.2.1 Production process

NdFeB material can be sourced from the geosphere via conventional mining and the anthroposphere, also called technosphere via recycling [21, 72]. The steps and effort required to remove the material and prepare it for use vary and should be carefully evaluated. In this chapter, however, the conventional production from the geosphere is the primary focus – recycling, on the other hand, will be addressed in more detail later.

Large deposits of bastnäsite, monazite, or other rare-earth-rich minerals are obtained by surface mining, which are then broken down, sieved, chemically cleaned, and milled in several repeating steps, then dried to achieve a concentrated ore [73]. Ion absorption clays are another source of rare earths. They are especially prevalent in southern China, specifically in the Jiangxi province, and have a considerable environmental footprint due to the nature of their mining [26]. The material is beneficiated, hydrometallurgically separated, purified, reduced to RE-metal, transported, alloyed to the target composition, melt-spun, and pulverized [74]. The typical production process, as presented by Sprecher et al. [33], is summarized in the following Figure 4.3.

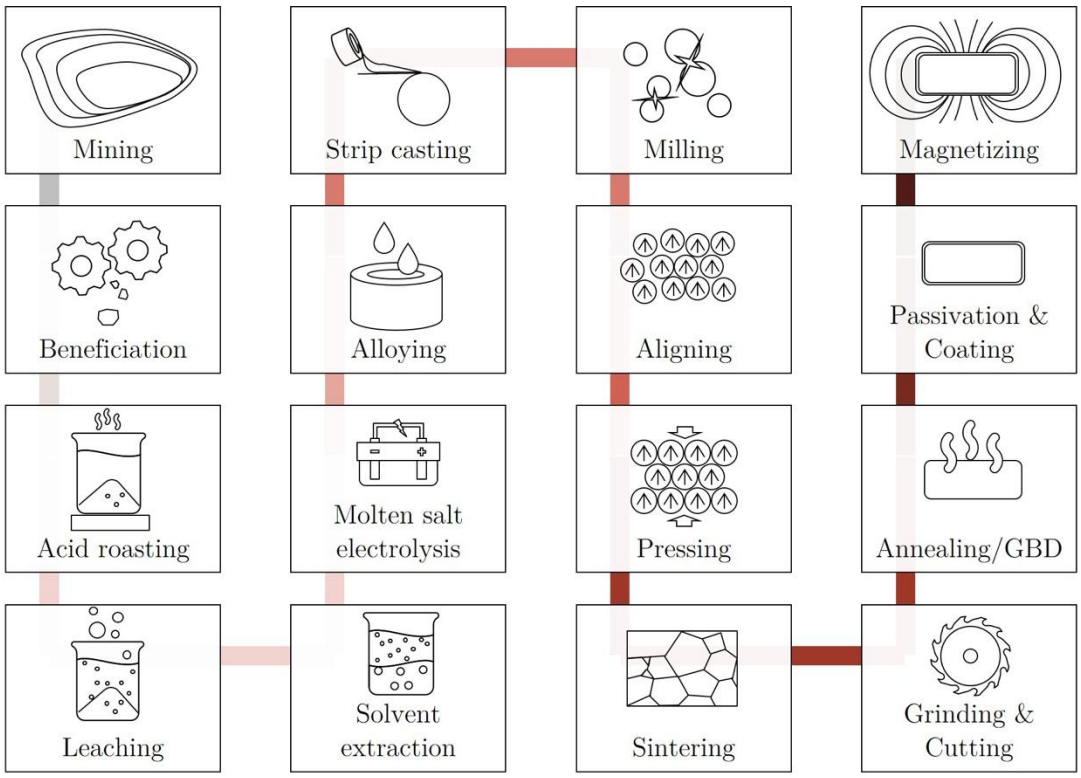


Figure 4.3: Typical steps from mine to magnet as according to Sprecher et al. [33], assuming mining of bastnäsite or monazite ores.

The resulting magnet powder is essentially treated in a very similar way both for virgin magnets and recycled magnets, therefore it will only be summarized shortly here and explained in more detail in context with the recycling process: The powder is prepared for sintering by degassing, milling, further alloying and then aligned and pressed. The compact is then sintered via liquid phase sintering. The resulting magnetic block can be cut and ground to its final proportions, annealed and/or grain boundary diffused to improve the magnetic properties, then passivated and/or coated and finally magnetized.

4.1.2.2 Emergence of magnetic properties in sintered NdFeB

Ferromagnetism as seen in iron generally arises when materials have unpaired electrons, leading to a magnetic moment being formed, and the magnetic moments of the atoms tend to align in parallel in a low-energy state, so even in the absence of an external magnetic field [75]. The distinguishing property is that all magnetic moments spontaneously align and thus are magnetized to saturation within a magnetic domain [76]. Ideally, all magnetic domains within a bulk material are aligned in parallel. Paramagnetism, as seen in Nd, arises when magnetic moments also align with an external field; however, they only align when the field is actively applied [75]. Through exchange coupling, the paramagnetic Nd significantly amplifies the ferromagnetic behavior of Fe.
NdFeB offers exceptional ferromagnetic properties due to its crystal structure and coupling. Iron mainly provides the dominant magnetic moment, while Nd contributes primarily to the magnetocrystalline anisotropy through its 4f electrons and spin-orbit coupling, magnetically “stiffening” the structure to ensure this alignment persists.

To understand the influence of design on recycling-related effects in the recycled material, the emergence of magnetism in NdFeB must be understood, as the material may undergo changes during recycling. Some changes are voluntarily induced, such as adding alloying elements, and some that are not, such as organic contamination from air exposure and glues, or metallic contamination from residues of coatings mixed into the material.

Elementary particle level

The source of magnetic phenomena is found in elementary particles which carry a charge, as magnetism is, simply put, caused by the movement of this charge [75]. This applies classically to electromagnets, but also to solid-state magnets. Each elementary particle carries a particular spin, which is a conserved quantity and one of two angular momentum phenomena in quantum mechanics. Electrons may be assigned with two different, discrete spin quantum numbers: -½ or +½ or also referred to as “down” and “up” spin. Electrons, being fermions, obey a set of behavioral rules, such as Pauli’s exclusion principle. No identical electrons may coexist in a cluster of elemental particles, like an orbital. This means that each electron must at least differ in one of four quantum numbers:

- Principal: energy level of the electron, therefore, the “shell” it occupies
- Azimuth: shape of the orbital and magnitude of the orbital angular momentum
- Magnetic quantum number: direction of movement, where the potential energy of the electron is also characterized, and a description of the orbital cloud
- Spin quantum number: The spin vector of the electron

For better understanding, the meanings of the quantum numbers are visualized in Figure 4.4.

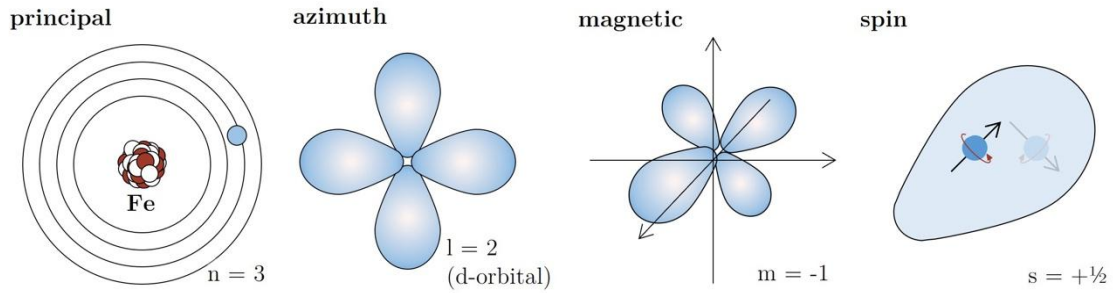


Figure 4.4: Simplified visualization of the meaning of the quantum numbers from principal deciding on which "shell" the orbital is, the azimuth deciding the orbital shape, the magnetic quantum number, the orientation of the orbital, and the spin, which one of the two spaces the electron occupies: up or down? Each electron in an atom has a unique combination of quantum numbers.

Atomic level

Looking at the electron’s surroundings, it also underlies another angular momentum phenomenon: the orbital angular momentum. This momentum is influenced by the occupied position relative to the atomic nucleus and the associated energy level of the electron in the atomic configuration; therefore, it is determined by the principal quantum number. There is an interaction between the orbital angular momentum and the electron's spin, which can be explained by the conservation of momentum, similarly to a gyroscope on a larger scale. Both the spin and the orbital angular momentum have a magnetic momentum, which could be characterized as a vector.

When electrons are distributed, Hund’s rules apply, stating that if there are several orbitals with the same energy level (principal quantum number), they are first occupied with one electron each, all with parallel spin [75]. This is caused by electron exchange

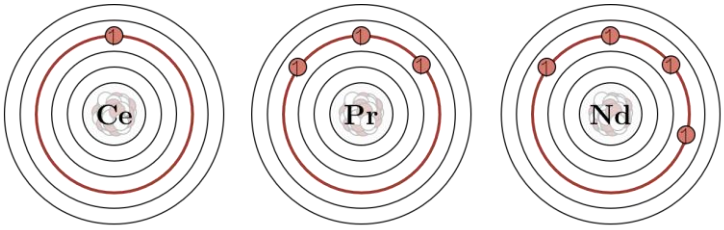
coupling. Exchange coupling is possible because electrons with the same quantum-mechanical properties can be seen as identical and can switch places [75]. Due to the Russell-Saunders coupling, the magnetic moments of the electrons are summed: The vectorial momenta add and subtract to a cumulative magnetic moment of the atom. If all orbitals of one energy level are either filled with none, one, or two electrons, the vectorial addition and therefore the magnetic dipole, is zero.

Therefore, the more uneven the electrons are distributed, the higher the magnetic dipole and hence the magnetic moment μ of the atom, which can be characterized as a vector pointing from south to north with the strength being proportional to the vector's magnitude [75]. Also, even fully occupied orbitals contribute to the magnetic moment to a lesser degree, as their spin moments may be cancelled out, but their orbital angular motion still increases the "magnetic impact" of the atom.

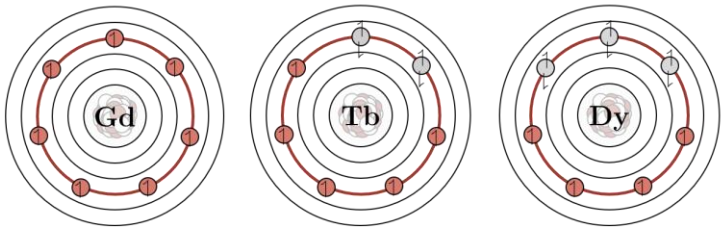
In the case of rare-earth elements like neodymium, the magnetic properties are derived from the 4f electron configuration.

The principal energy level is not the only factor in an orbital's energy level; the orbital type is also essential. That is why, in the case of the rare earths, partially occupying the 5s and 6d shells is more energetically favorable than filling the 4f shell first. In the 6th period of the periodic table, the 4f shell is unoccupied until Cerium, which then has a single electron on the 4f shell. Gd and Eu both have 7 electrons on the 4f shell, so the magnetic dipole caused by the 4f configuration is zero, as all orbitals are occupied once. The more asymmetric, the greater the magnetic dipole due to vectorial addition. The following Figure 4.5 shows the electron configuration of some magnet-relevant 6th period rare earths based on the basic [Xe] configuration. The unpaired or free 4f electrons are marked, signifying their spin-related increase in magnetic moment derived from vectorial addition.

The magnetic dipole is almost the same for Pr, Nd, and Pm (radioactive, synthetic) as well as Tb, Dy, and Ho, which all have 3-5 free electrons. For the latter, heavy rare earths, the higher orbital angular momentum due to a greater number of electrons enhances the total magnetic momentum. These elements, except Pm for practical reasons, are used in $R_2Fe_{14}B$ compounds.



$R_2Fe_{14}B$ properties [Sagawa et al.]			
Anisotropy field H_a at 27 °C [kOe]	29	87	67
Magnetization M_s at 27 °C [μB/formula unit]	24	32	33
Curie temperature T_c [°C]	149	296	313



$R_2Fe_{14}B$ properties [Sagawa et al.]			
Anisotropy field H_a at 27 °C [kOe]	24	220	149
Magnetization M_s at 27 °C [μB/formula unit]	18	14	14
Curie temperature T_c [°C]	386	347	325

Figure 4.5: Electron configurations of selected rare-earth elements with a semi-filled 4f orbital, leading to distinct magnetic behavior. Below each configuration, some key properties of the $R_2Fe_{14}B$ compound are shown, based on Sagawa et al. [77].

Elementary cell level

NdFeB magnets consist of the characteristic hard magnetic φ-phase, $Nd_2Fe_{14}B$, which occurs as the grain phase, and the Nd-rich grain boundary phase, which is added to the stoichiometric $Nd_2Fe_{14}B$ and typically makes up 3 – 10 wt.% of the magnet and is non-magnetic, effectively isolating the grains from each other. The elementary cell structure of the hard magnetic φ-Phase is explained in the following.

In the elementary cell of the $Nd_2Fe_{14}B$ φ-Phase, there are essentially two sublattices present. The 3d transition metal sublattice, in this case with Fe, and the 4f rare earth sublattice, in this case with Nd. B serves to create the particular elementary cell and is assumed not to have an impact on the magnetic properties aside from stabilizing the structure.

The interactions between the 4f and 3d electrons are the mechanisms that give NdFeB its special magnetic properties. The 3d-3d coupling stabilizes the magnetic ordering against thermal agitation and thus is responsible for the basic magnetization. In contrast, the 4f-3d coupling sustains the rare earth sublattice's magnetization and thus the strong 4f anisotropy and to a lesser degree the enhancement of the magnetic field by the paramagnetic properties of neodymium [77].

There is no direct 4f-4f coupling, as the Nd atoms lie too far apart; however, they are indirectly coupled by the 3d lattice, a phenomenon referred to as Ruderman-Kittel-Kasuya-Yoshida coupling [75]. The elementary cell of NdFeB and the structure of its sublattices were first researched and described by Herbst et al. [78] using neutron diffraction, and are shown in a reproduced image in Figure 4.6:

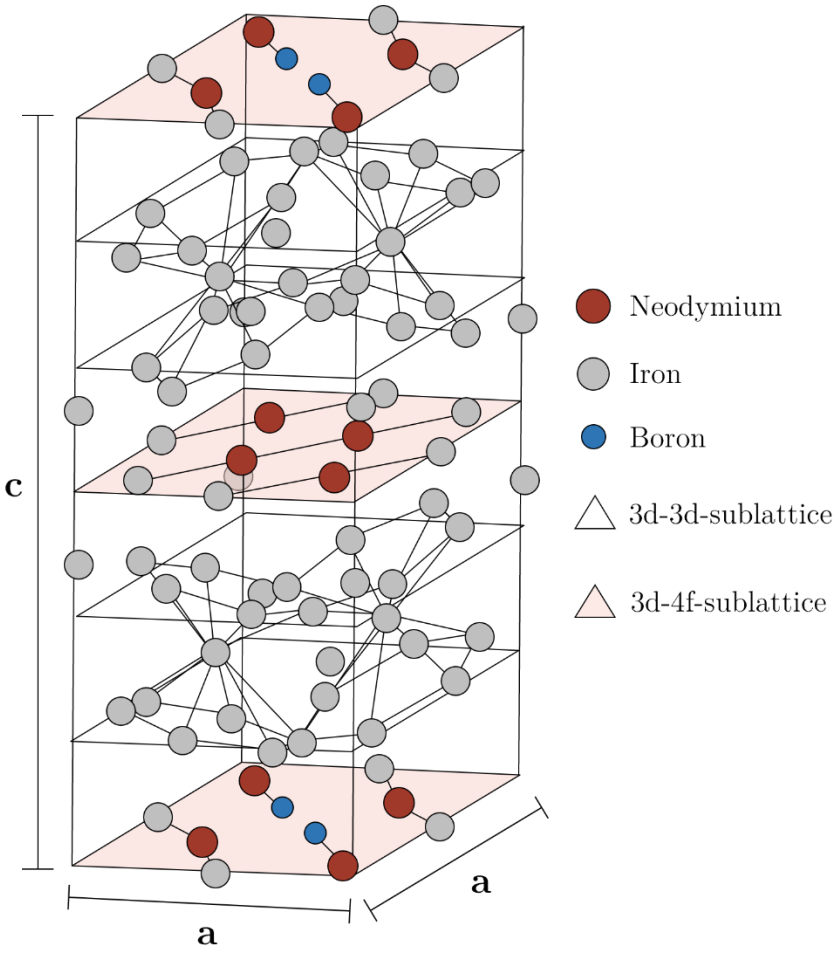


Figure 4.6: Elementary cell of $Nd_2Fe_{14}B$ based on Herbst et al. [78].

The most important characteristic that sets NdFeB apart from other magnet materials is the very high magnetocrystalline anisotropy [75]. Magnetocrystalline anisotropy is one of several sources of magnetic anisotropy; others include shape anisotropy, stress anisotropy, externally induced anisotropy, and exchange anisotropy; however, magnetocrystalline and shape anisotropy play the most significant role in the creation of the overall anisotropy [75, 76]. Magnetic anisotropy means that both the magnetic susceptibility and magnetization vary with direction, with magnetization favored along a certain "easy axis" [75, 76]. The easy axis, the direction in which magnetization can be induced most readily, is the long c-axis, which is exaggerated in the elementary cell model shown in Figure 4.6. The exchange interactions and thus, magnetic coupling in the elementary cell are highly dependent on the distances between the atoms and the atomic environment, which is where substituted and interstitially deposited elements can have dramatic effects which translate into the magnetic properties.

The magnetic anisotropy is an important property, which directly affects the intrinsic properties of the compound, which will be discussed more closely in section 4.1.2.4.

Magnetic domain level

When a magnetic material is magnetized, magnetic domains reorient. At very small grains (0.20 µm – 0.26 µm [75, 77], also called single domain grain size) or high external fields, grains are fully magnetically saturated – all magnetic moments in a grain are aligned parallel. However, in any other state, the magnetic fields in the hard magnetic grain phase are divided into magnetic domains. Magnetic domains or Weiss domains are volumes in which all magnetic moments are aligned parallel [76]. They can be imaged via the Kerr effect, as the localized magnetic field deflects the incoming photons or electrons. In optical microscopy, the Kerr effect is visible with polarization filters; in SEM, it can be simply imaged in secondary electron mode with suitable surface preparation. Imaged magnetic domain structures are shown in Figure 4.7.

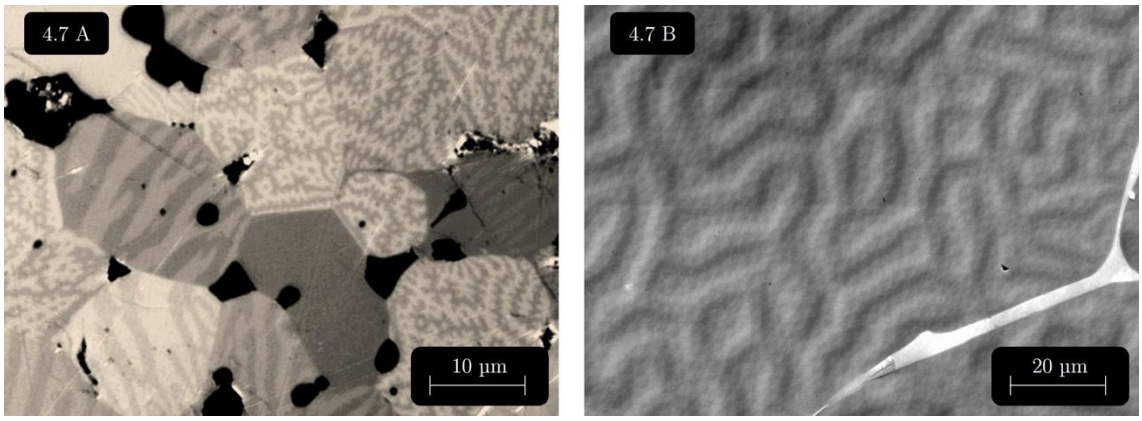


Figure 4.7: Visualized magnetic domains. Deflection into different directions due to domain reversal shows up with different contrast. On the left in 4.7 A, a magnet has been imaged by an Olympus BX41M-LED light microscope; on the right in 4.7 B, a close-up of meandering domains in an abnormally large grain was recorded in a Hitachi FlexSEM 1000II via SE-mode.

Since a large particle being magnetically saturated would lead to large surface charges at its ends, the system has a high magnetostatic energy. This energy is approximately halved when the grain is split into two magnetic domains [79]. The newly emerged, nucleated domains are aligned in reverse to the axis of the original domain [80]. Between the domains, a so-called domain wall is formed, in which the magnetic moment rotates. Depending on the rotation direction, a Bloch or Neél wall forms, which also reduces the magnetostatic energy. The thickness of a domain wall is about 5.2 nm [77], which is equal to the exchange coupling length [81]. As domain walls are generally able to move, domains can grow by the realignment of single atoms or a full moment turn of a domain [82]. New domains may also emerge from domains that are too large to be stable [80]. On the other hand, energy optimization by domain nucleation does not occur in grains below a critical size of 0.26 µm, the single domain grain size [77], and once larger grains run out of excess magnetostatic energy or the neutral grain is overridden by a large magnetic field, no more reverse domains are formed.

Two important mechanisms govern the coercivity, which is simply put, the ability of the permanent magnet to withstand demagnetization, which are nucleation of reverse domains and expansion of reverse domains. The coercivity will be addressed more closely in section 4.1.2.5.

The formation- or nucleation of a reverse domain preferably happens in stray fields, which, for example, can be caused by sharp edges and uneven surfaces at the grain boundaries [77]. These reversed domains also often appear near structural defects or oxidation, lower the anisotropy field and thus, the coercivity [77, 81, 83]. Hence, processing techniques promoting even grain surfaces and grain boundary phases help prevent reverse-domain nucleation [77].

Once a reversed domain is created, domain wall motion occurs, favoring the expansion of the domain that best aligns with the applied external field. The movement of the domain walls is constricted by pinning mechanisms – the moving domain walls need to overcome steep energy gradients near local defects or "perfect" low-energy structures [77, 84]. Until the respective energy level is reached, e.g., by increasing the external field further, the domain wall is stuck.

When an external field unpins the domain walls by exceeding a critical field strength, the sudden movement of the walls generates Barkhausen noise [82, 85]. The Barkhausen effect is both visible as insets on the hysteresis curve as it is audible as a cracking noise during field direction change [82]. Magnetic hardening due to restriction of domain wall movement seems similar to mechanical hardening due to microstructural defects and restriction of sliding planes [86], but these effects do not necessarily have to occur simultaneously [84].

This discrepancy, also referred to as Brown's Paradox [76], is an explanation for why real magnets do not reach theoretically expected coercivities defined by the anisotropy field H_A, which are several times higher than the best measured coercivities. The coercivity, the ability of the magnet to withstand demagnetization, is favored by as little as possible defects to avoid nucleation of reversed domains, but also by the existence of defects to be able to pin and restrict domain wall movement [87]. Domain wall mechanisms are visualized in Figure 4.8 for better understanding.

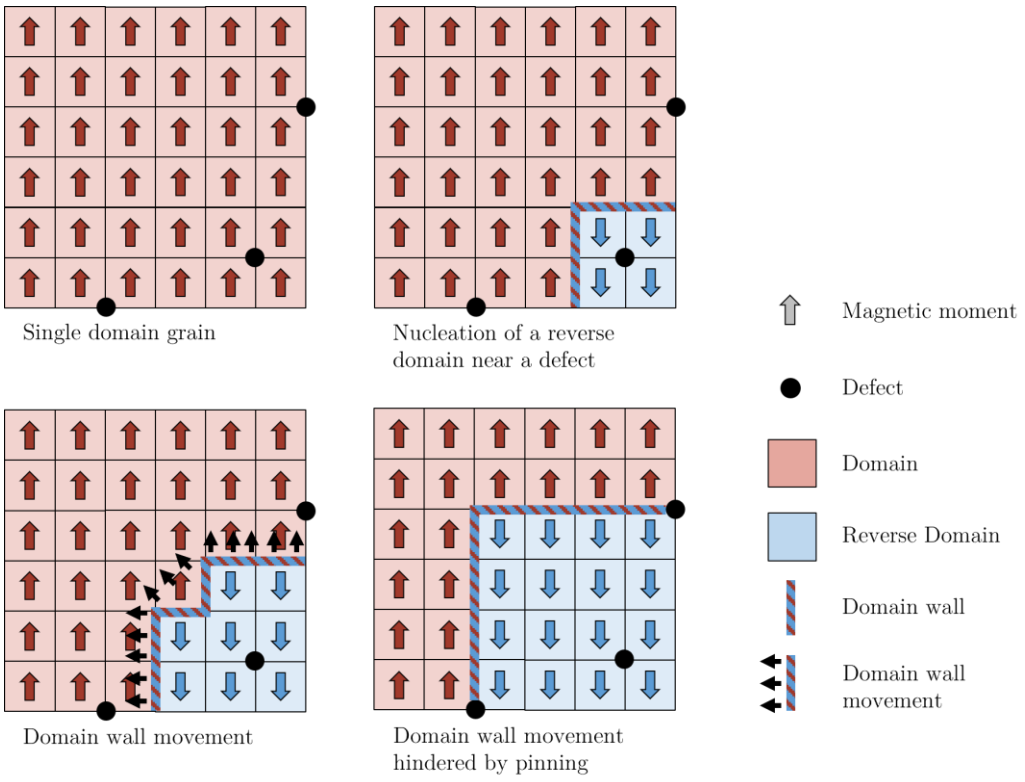


Figure 4.8: Schematic depiction of domain wall movements, nucleation and pinning.

In general, NdFeB is regarded to be a nucleation-controlled system [81], but this has been partly challenged by some research groups [88]. This discussion is also linked to the question, whether the Nd-rich phase is ferromagnetic or not [89]. Ferromagnetic phases in NdFeB permanent magnets act as nucleation sites for reverse domains, especially near edges and corners [87, 90, 91].

The dependence of coercivity on nucleation mechanisms and, therefore, the need for said microstructure is best fulfilled by as small as possible grains in theory [84]. Furthermore, defects and irregular interfaces, for example with the grain boundary phase or oxides, which can act as nucleation sites should be avoided. This is further expanded on in the following.

Microstructural level in sintered NdFeB

To summarize the requirements towards the microstructure that arise due to the mechanisms on domain and domain wall levels, the characteristics of a "good" sintered NdFeB microstructure are quite simple: it requires well-aligned small grains, thinly but thoroughly covered by the Nd-rich grain boundary phase, and few contaminating phases like oxides and carbides present.

Realistically, such a magnet should also be fully dense and have a grain size that offers a good balance between being prone to oxidation and the mentioned domain structure benefits arising from smaller sizes, whereas 3 – 5 µm are described as optimal [92]. Excellent control during magnet production including milling, shaping, aligning, and sintering both in terms of grain size and contamination management is required for producing high quality magnets. The formation of organic contaminating phases, especially in the highly reactive Nd-rich phase, must be suppressed, since, e.g., Neodymium oxide does not distribute well to decouple the grains and thereby hinders demagnetization.

The decoupling of the grains, meaning the propagation of short-range exchange coupling is suppressed between grains, is important to allow for high coercivity, as otherwise, demagnetization by reverse domains can spread quickly. A very thin grain boundary phase of few nanometers, barely exceeding the exchange coupling range of 5.2 nm [83] is theoretically optimal. Any grain boundary thickness above the length of the exchange coupling does not serve the enhancement of the coercivity and thus, lowers the remanence as there is less magnetic mass in the whole magnet.

In Figure 4.9, an example is shown where grains are seen that are decoupled by a minimal layer of the grain boundary phase. Additionally, a core-shell structure from Tb grain boundary diffusion is seen. Such core-shell structures locally modify the magnetic properties. $R_2Fe_{14}B$ compounds with Dy and Tb enhance the local magnetocrystalline anisotropy, leading to an increase in coercivity, but also couple antiferromagnetically with iron, leading to a loss in remanence, which can be described as the magnetic strength. Local application reduces the trade-off between these essential properties and is more resource efficient, showing that combining changes in the composition and elementary cell set-up and processing methods affecting the microstructure is a viable and effective means to optimize permanent magnets. It also highlights that the final magnetic properties depend on the entire chain of decisions from choosing the base alloy to setting each processing parameter.

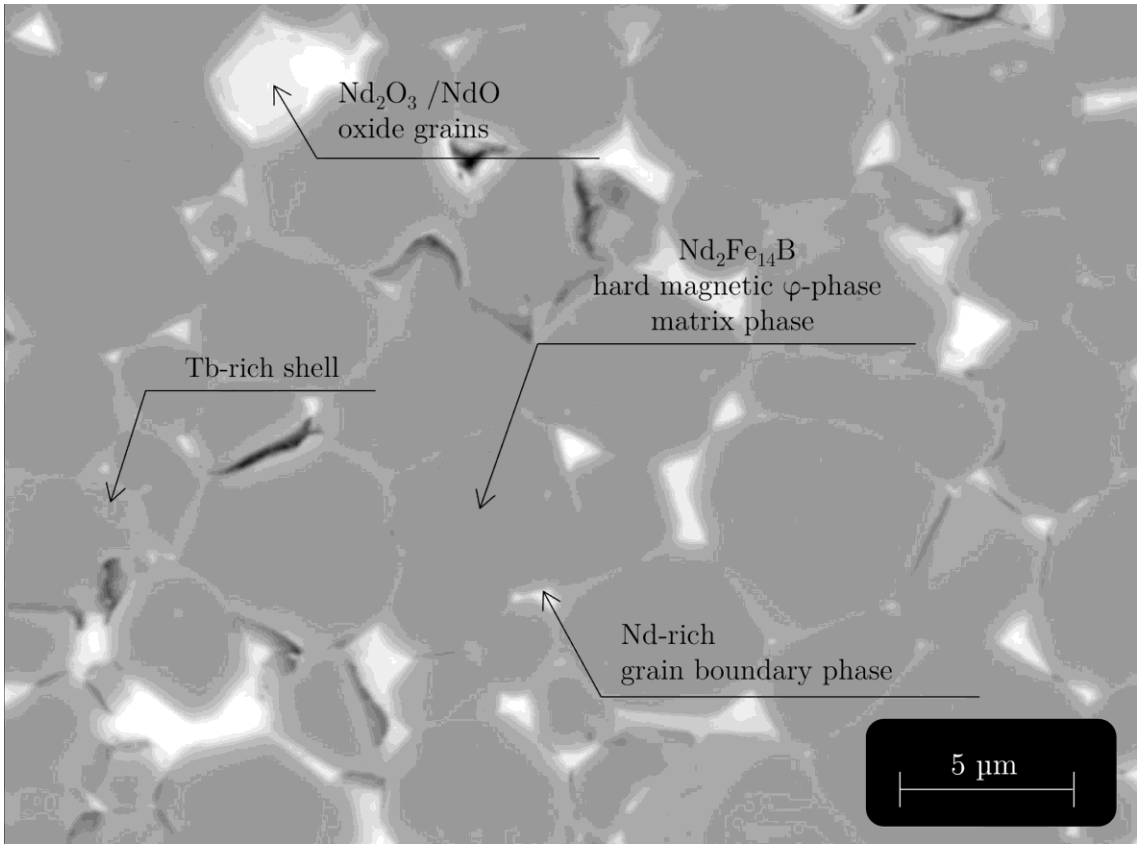


Figure 4.9: Example of a typical magnet microstructure with rather even grain size distribution, a thin, barely visible grain boundary layer, some polishing defects, and a core-shell structure produced by Tb grain boundary diffusion. Recorded by a Hitachi FlexSEM 1000II via BSE-COMP mode.

4.1.2.3 Phases in sintered NdFeB

Due to NdFeB being a ternary system, a number of phases can be formed, some of which are marked in the ternary phase diagram shown in Figure 4.10. A comprehensive overview of possible phases is given by Malfliet et al. [93]. The most important phases in a NdFeB permanent magnet are explained in the following.

The hard magnetic φ-phase, also called matrix phase, is the core of the NdFeB permanent magnets and has the stoichiometry of $Nd_2Fe_{14}B$. This phase is strongly hard magnetic due to coupling between the Fe and Nd sublattices, as described in more detail in section 4.1.2.2, where the tetragonal elementary cell was shown. While boron does not directly contribute to the magnetic structure, it is integral to uphold the elementary cell.

The phase can be formed with other rare earths replacing the Nd, offering different magnetic properties. The properties of selected $R_2Fe_{14}B$ compounds were shown in Figure 4.5 earlier in the document. Generally, compounds with heavy rare earths like $Dy_2Fe_{14}B$ or $Tb_2Fe_{14}B$ offer higher anisotropy fields (doubled with Dy, tripled with Tb), leading to higher coercivities and lower magnetizations (more than halved in both cases), leading to lower remanences. Notable are also $Ce_2Fe_{14}B$, which offers price benefits but a reduction of Curie temperature, remanence, and coercivity, as well $Gd_2Fe_{14}B$, which trades coercivity and remanence for a significantly higher Curie temperature, see Figure 4.5 and Sagawa et al. [77].

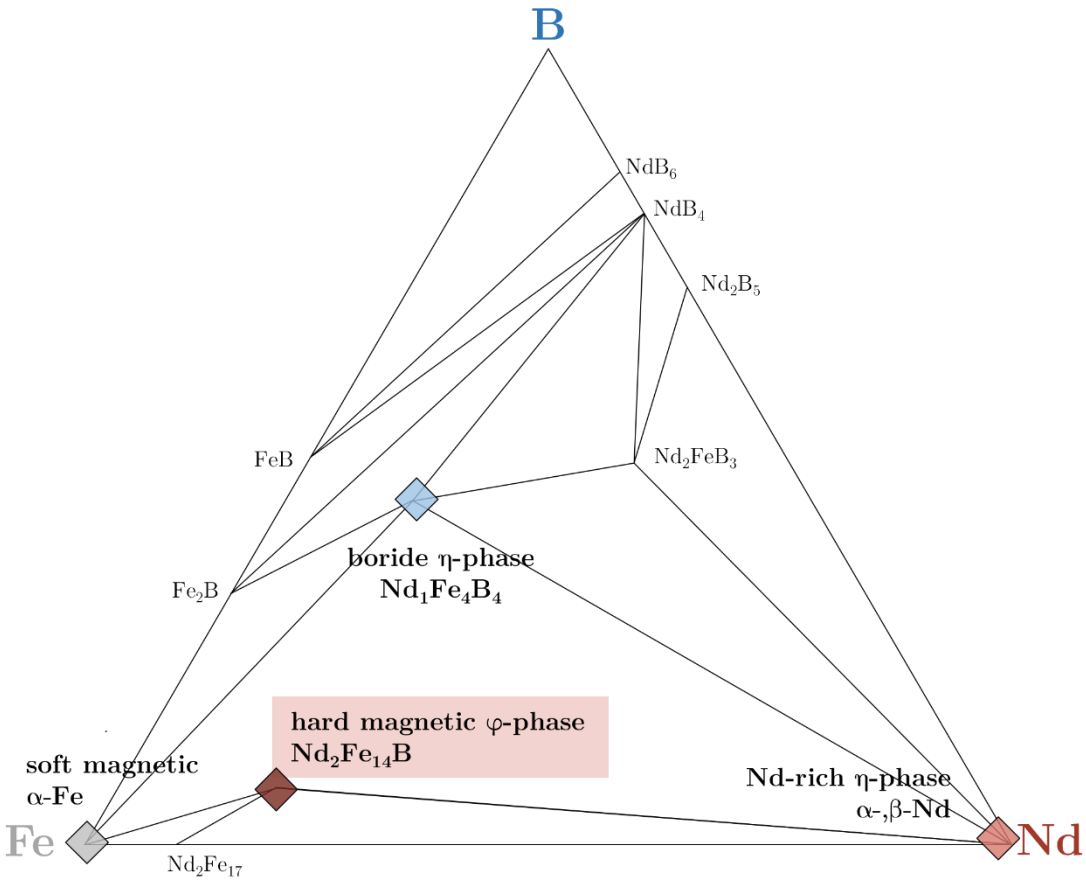


Figure 4.10: Isothermal ternary phase diagram at 873 K based on Fu et al. [94] and Malfliet et al. [93].

Fe can be partially substituted by a number of different elements, namely Co, for which all positions can be substituted as well as V, Cr, Mn, Ni, Cu, Al, and Si, where only some positions can be substituted [77]. Furthermore, B can be substituted by C partially as well [77]. The hard magnetic phase is protected from corrosion by the anodic Nd-rich phase, but can also oxidize at elevated temperatures above 100 °C [95]. When the hard magnetic phase is oxidized, it is disproportionated into Nd oxides and nanocrystalline structures with α-Fe and Fe_3B [96–98]. It can further be mentioned that the $Nd_2Fe_{14}B$ compound can take up to five H atoms, which leads to a small volume expansion [21, 99]. Figure 4.11 shows the hard magnetic phase in conjunction with other relevant phases.

The coercivity of sintered NdFeB magnets is governed by a continuous, thin layer of the Nd-rich phase, which decouples or at least reduces magnetic interactions between grains [77], preventing the propagation of reverse domains. For this, excellent grain boundary wetting is necessary to ensure complete magnetic isolation.

The composition of the Nd-rich phase is not uniform and has been reported by different sources in literature. Sagawa et al. (1987) originally determined the composition to be around 95 at.% of Nd and 3 – 5 at.% Fe and a trace of B, forming a fcc structure, which also aids sintering and is mostly found in the triple points and grain boundaries [77]. However, newer research reflects a more variable composition, where the Nd-rich phase colloquially refers to a number of different phases in the triple points and grain boundaries.

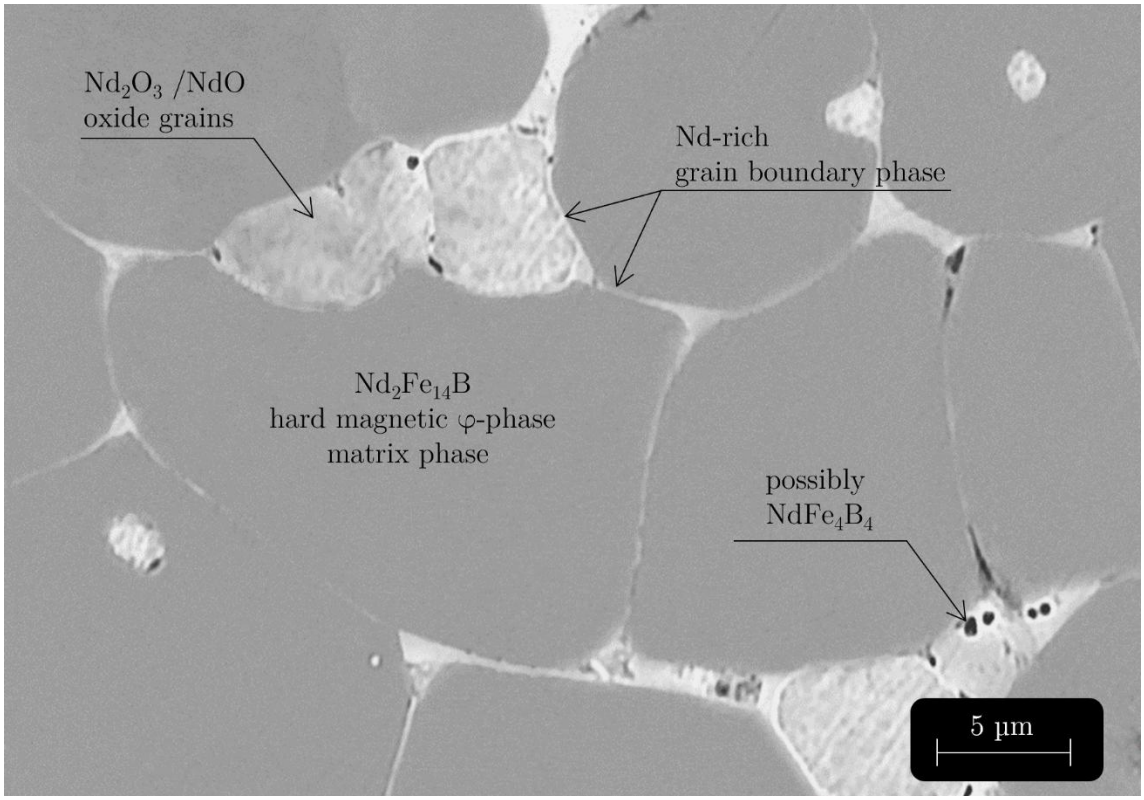


Figure 4.11: Different typical phases observed in NdFeB permanent magnets. Please note that this image does not represent a viable microstructure, but rather as a good visual representation of the phases. Recorded with a Hitachi FlexSEM 1000II via BSE COMP-mode.

Sasaki et al. (2016) compile different studies, whereas several studies [13,20,23,38] found the Nd-rich phase to take dhcp structure, but in the conducted study, the metallic Nd-phase observed showed to have fcc or la3 structure. In the fcc phase, more Cu and Co were found than in the la3 structure [100]. Cu is assumed to be present as NdCu precipitates [100].

Along the c-axis plane, the Nd-rich grain boundary phase is usually found crystalline as described, and contains higher amounts of Nd (~60 %) [100]. At the "ends" of the grain perpendicular to the c-axis, the Nd-rich phase seems to be amorphous, containing lower amounts of Nd (~35 %) [100]. Therefore, exchange coupling and pinning mechanisms are most likely anisotropic, and the structure of the Nd-rich phases parallel to the easy axis (c-axis) are especially critical for coercivity [100, 101].

For successful sintering and development of a high-coercivity microstructure, the Nd rich phase must remain active, therefore non-oxidized, as oxide formation drastically reduces wettability. This effectively reduces the coercivity due to lack of isolation as well as the densification during liquid phase sintering [92, 102, 103].

NdFeB is highly prone to corrosion, as the potential difference between Nd, which has a highly negative reduction potential, and $Nd_2Fe_{14}B$ is high and Nd is highly chemically active [104, 105]. Due to this potential difference, NdFeB forms galvanic corrosion cells, where the Nd-rich phase corrodes primarily, leading to intergranular corrosion [106–108]. The formed Nd_2O_3 and NdO in the grain boundaries of a corroded magnet offer magnetic isolation as well [109], whereas there is no immediate decrease in coercivity caused by corrosion. However, the magnetic properties of a recycled magnet made from the same

material later on can be affected. Nd further forms carbides and hydrides [21, 110]. Carbides affect the Nd-rich phase similarly to oxides, however binding more Nd per C/O atom and to a higher degree, added C/O mass. $NdH_{2.7}$ is formed when Nd is exposed to H atmosphere at room temperature and ambient pressure, causing a high volume expansion [21]. This can be utilized during different processing methods used in magnet manufacturing and recycling.

Secondary phases, like the Nd-rich phase, oxides, carbides, or also borides and α-Fe can influence the coercivity, which can be described by the Kronmüller equation $\mu_0 H_{cJ}=\alpha\mu_0 H_A - N_{eff} J_s$. Here, the anisotropy field H_A is multiplied with the reduction factor α which is based on microstructural defects. Then, the product of the saturation polarization J_s and a variable Neff accounting for local demagnetizing fields is subtracted.

N_{eff} and α are affected not only by processing-induced defects, but also for impurities. The presence of oxides for example can both cause local demagnetizing fields due to serving as a nucleation site (N_{eff}) or by disturbing proper microstructural evolution during sintering (α). Another example of how impurities can greatly affect the magnetic properties is the formation of α-Fe. Under certain circumstances, when the ratios of the elements in the alloy are disproportionate and there is an overrun of Fe, the soft-magnetic α-Fe can be formed, which is a soft magnetic phase that will act as a nucleation site for reverse domains and therefore cause demagnetization and local magnetic inhomogenities [87]. Formation of α-Fe can occur during sintering, the Fe is either replaced by another element which has solubility in the φ-phase or there $Nd_2Fe_{14}B$ compound cannot be formed due to a lack of for example, Nd [77]. When an excessive amount of Nd, which has been dissolved from the φ-phase into the liquid Nd-rich phase, is bound, e.g., by high amounts of unbound carbon being present in the material, α-Fe can be found due to a overrun of Fe [110, 111]. To avoid this, a small overrun of B is added to the alloy, which forms the boride phase instead [112]. Aside from the effects on the coercivity, a reduction of remanence can also be caused by the presence of impurities. While it is generally thought to be governed mostly by the φ-phase, it should also be mentioned that the overall density, degree of magnetic alignment and volume of the φ-phase fraction are important for the overall remanence and can be hindered by impurities. For example, disproportionation of the φ-phase (whereas also soft magnetic α-Fe forms) will reduce the hard-magnetic volume. Further, a lack of active Nd-rich phase will impair densification. Also, it should also be mentioned that misaligned grains are often observed in direct contact with Nd_2O_3 grains at the triple junctions of grain boundaries, as the attached oxide grains may hinder grain rotation during alignment [100].

4.1.2.4 Intrinsic magnetic properties

The explained micromagnetic phenomena lead to the magnetic properties of the bulk. There are various properties which are measurable; some are referred to as intrinsic properties, others as extrinsic properties. A number of properties were already mentioned in the aforementioned explanations and will now be classified and their meaning for technical applications will be explained in more detail. This sub-section serves to connect the dots between magnetic phenomena and magnetic properties observed.

First of all, the intrinsic magnetic properties are decided by the chemical composition of the alloy and the crystal structure [113]. On the other hand, extrinsic magnetic properties are the product of the intrinsic properties and the grain morphology and microstructure [113, 114]. The most important extrinsic properties of a hard magnetic material can be found by measuring the hysteresis curve. A typical hysteresis of hard magnetic materials is shown in Figure 4.12. In the following Figure 4.13, common deviations from the optimal hysteresis shape are shown, as well as their most common causes.

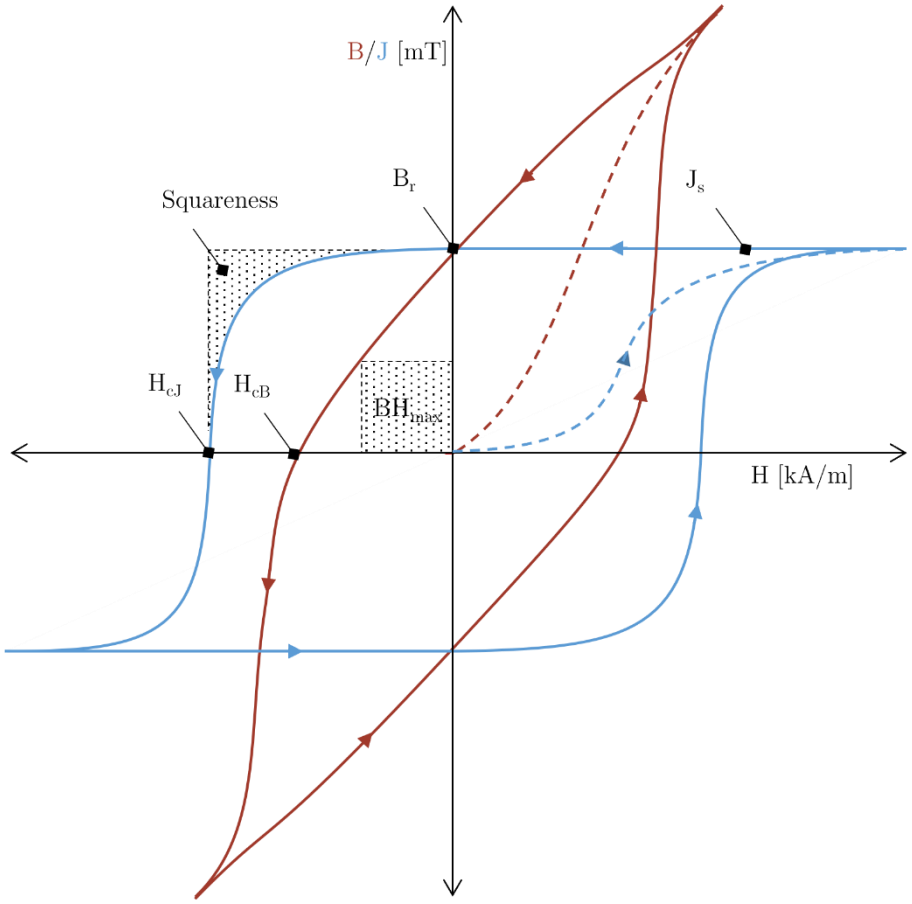


Figure 4.12: Typical hysteresis loop with the most important properties marked.

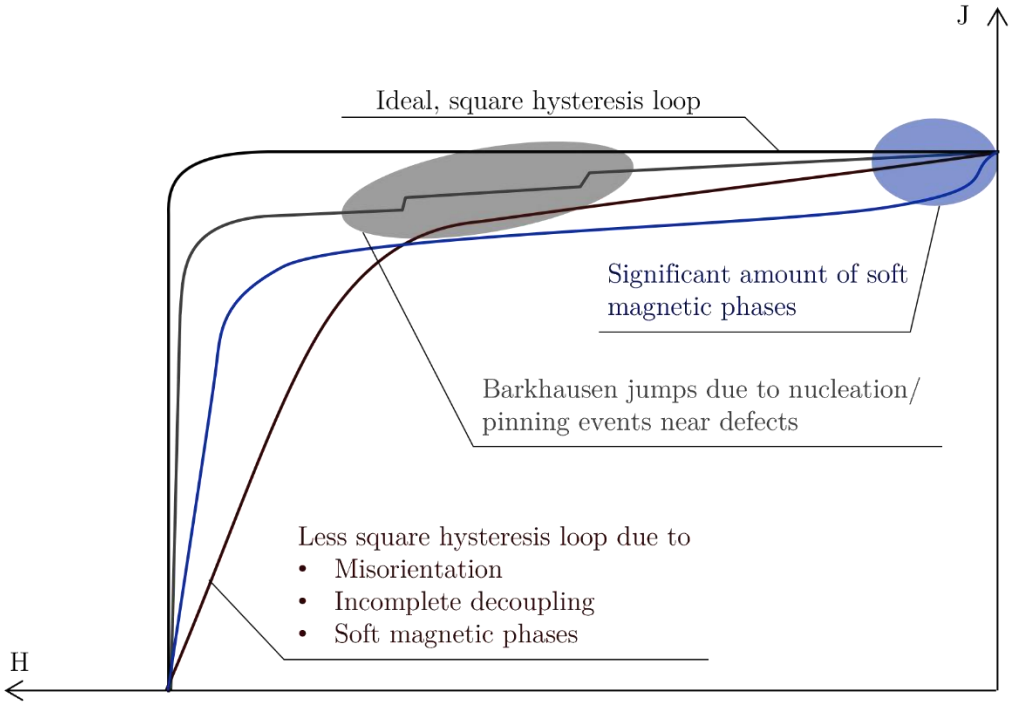


Figure 4.13: Common distortions of the hysteresis loops and some of the most common reasons. Second quadrant of the hysteresis only.

Anisotropy and coercivity

As already mentioned, the anisotropy field H_A and the magnetocrystalline anisotropy K_1, which are both intrinsic properties, have significant effects on the coercivity, which is an extrinsic property. The magnetocrystalline anisotropy characterizes the energy barrier to overcome for a single crystal's magnetization to rotate 90° out of its easy axis. This depends on the crystal structure and chemical composition. A more tangible value to describe this is the anisotropy field H_A, which describes the theoretical magnetic field strength needed to enforce this rotation. In $R_2Fe_{14}B$, the anisotropy field varies depending on which rare earth is in the local compound, as well as on the temperature [77]. It is directly caused by the coupling of the 4f and 3d orbitals, and heavy rare-earth elements like Dy and, especially, Tb provide exceptional magnetocrystalline anisotropy of more than double in case of Dy and triple in case of Tb than that of Nd, see Figure 4.5 [77, 78].

While the anisotropy field is a theoretical property for a single crystal, the coercivity is a measurable property for an entire bulk magnet. The coercivity can be characterized as the ability of the magnetic material to withstand demagnetization. It is governed by the magnetocrystalline anisotropy and the anisotropy field, but also to a higher degree by microstructural properties like the morphology and the defects of each grain and the magnetic decoupling of the grains, see the Kronmüller equation explained in the previous sub-section [114, 115]. Coercivity is defined as the magnetic field strength needed to demagnetize a magnetic substance that has previously been magnetized to saturation [75, 116]. The coercivity is strongly temperature dependent, as the anisotropy field decreases with higher temperature and the application of a reverse field leads to easier demagnetization [117]. There are two very common ways to quantify the coercivity.

For the coercivity H_{cB}, "demagnetized" is defined as there is no remaining magnetic flux density B (given as T), for the coercivity H_{cJ}, demagnetized is defined as there is no remaining magnetic polarization J (given as T). Both are given as a magnetic field strength, thus in A/m or more commonly, kA/m for when the B/J curves reach 0 T. Generally, magnets with a high coercivity are considered magnetically hard, meaning that they show great resistance against demagnetization under the influence of external fields. In practice, H_{cJ} is often the primarily considered value, as with H_{cB}, while no outer field is exhibited when the respective reverse field is applied, there is still remaining polarization within the magnet. This means that if the reverse field is removed, the magnet can still exhibit a magnetic field again. After surpassing H_{cJ}, the magnet is macroscopically and reliably demagnetized.

Theoretical H_{cJ} values, calculated based on the nucleation model, where in large magnetic fields all domain walls are removed, exceed the practically measured values by far, roughly 300 - 500 % for NdFeB [81, 118, 119]. This is due to Brown's paradox explained in prior sub-sections. As noted there, NdFeB appears to be a nucleation-controlled system; however, the nucleation theory cannot explain the measured coercivity well since it relies on a very simplified microstructural model. To date, several methods to approximate coercivity in realistic systems have been established, including micromagnetic simulations and phenomenological models, which have their focus on a more macroscopic model. They are extensively discussed in a review by Zhao et al. [120]. The micromagnetic approaches differentiate between the magnetic reversal mechanisms, see for example Sepehri-Amin et. al [121], Kronmüller & Goll [122] and Schrefl et al. [123]. The phenomenological models neglect those and rather take thermal activation effects into account see e.g., Sagawa et al. [124], which are again neglected by the micromagnetic models. Both are able to predict coercivity quite well, despite taking different parameters into account [120].

Additionally, in a bulk material, the grain boundary phase also plays a major role in governing coercivity as the Nd-rich phase significantly influences coercivity, like first majorly reported by Hiraga et al. [125]. Kou et al. [126] further report that they found

single grains to not exhibit coercivity in the absence of a grain boundary phase. A widely accepted theory is that magnetic isolation and therefore decoupling of neighboring grains via a non-ferromagnetic intergranular grain boundary phase avoids exchange interactions, and thus propagation of demagnetization to neighboring grains [127, 128]. Contrary to this, other researchers suggest that the Nd-rich grain boundary phase is actually ferromagnetic and must therefore act as a pinning site rather than a decoupling barrier [88]. Generally, grain boundaries as pinning sites are an accepted concept [127].

Saturation magnetization and remanence

The high saturation magnetization M_s observed in NdFeB is owed to the large magnetic moment of the material, and is an intrinsic property based on the composition and crystal structure.

The highest M_s values can be achieved by $Nd_2Fe_{14}B$ at 33 μ_B per functional unit (f.u.), closely followed by $Pr_2Fe_{14}B$ with 32 μ_B/f.u., as shown in Figure 4.5. If the $R_2Fe_{14}B$ compound is formed with other rare earths, a significant loss in saturation magnetization can be seen. For example, a fully Ce-substituted compound will only exhibit 24 μ_B/f.u., a fully Dy- or Tb- substituted one will merely amount to 14 μ_B/f.u. each, which is less than half of what is achieved with "regular" $Nd_2Fe_{14}B$. This leads to remanence and coercivity being in a trade-off relationship to a certain degree [129].

The saturation magnetization is the basis for the optimal remanence that can be reached in a perfect magnet, however, as addressed before, the density, the volume fraction of the hard magnetic phase and the alignment play a role in forming the measurable remanence of the bulk. The remanence is thus an extrinsic property.

The remaining magnetization of a material after the removal of an external field is characterized as the remanence M_r [116]. The magnetization is equal to the amount of the magnetic moment per volume [116]. M_r is rarely used in engineering, as the more designative property is B_r, the residual magnetic flux density given in Tesla (T; 1 T= 1 Vs/m^2), which is also the determined property in B-H-analyzers. When the remanence is described in the following, it is referred to the technically relevant B_r usually given as T or mT.

Other relevant magnetic properties in technical applications

Aside from the coercivity and the remanence, other properties derived from the hysteresis loop are commonly used to characterize the suitability of permanent magnets for technical applications. These are the maximum energy product and the squareness, which can both be calculated from the hysteresis loop and can be classified as "compound" extrinsic properties.

One of the most designative properties arises from the combined magnetic properties with regard to both the strength of the magnet (the remanence) and the magnet's resistance to demagnetization (the coercivity) is the maximum energy product. The maximum energy product of the magnet which is given in kJ/m^3 and aligns with what magnet volume is required to achieve a certain performance [130]. Looking at a hysteresis, BH_{max} is usually shown as the square with the largest possible area fitting under the B-graph on one quadrant. The theoretical maximum of the maximum energy product is 512 kJ/m^3, which would correspond to a magnet grade of N65 [39, 114].

Another aspect is the so-called squareness, which characterizes how "square" the hysteresis loop is, meaning how long the polarization J remains stable when a reverse field is applied. This is especially important for electric machine applications, where the permanent magnets are constantly exposed to reverse fields. There are different mathematical models to characterize the squareness [131, 132, 133].

Other relevant magnetic properties in magnet recycling

The Curie temperature T_c is an intrinsic property, which describes at which temperature the magnet is fully demagnetized. The Curie temperature again varies depending on the chemical composition of the $R_2Fe_{14}B$ phase and typically does not have technical relevance during the use phase of an electric machine, however, in recycling, it is important to demagnetize at least at T_c.

It should be mentioned that the reverse fields and the temperature under which they are applied interact strongly, influencing the hysteresis shape. Tendentially, a higher Curie temperature improves the thermal stability of the NdFeB magnets, as the properties of the magnet remain stable and/or sufficient over a wider temperature range. However, the temperature dependance of H_{cJ} is also related to H_A.

4.2 Recycling of NdFeB Permanent Magnets via HPMS and Sintering

Hydrogen Processing of Magnetic Scrap (HPMS) is a direct recycling process, where NdFeB is exposed to hydrogen at room temperature, where it embrittles and disintegrates [50, 133]. The produced powder is prepared, shaped, aligned and re-sintered. Although short-loop recycling via HPMS and resintering is very straightforward, especially compared to long-loop processes, as illustrated in Figure 4.14, it requires significant pre-processing. The magnets have to be located, demagnetized, extracted, and separation of different types of contaminants like coatings and adhesives has to be done before or after HPMS processing. These procedures are described in this chapter.

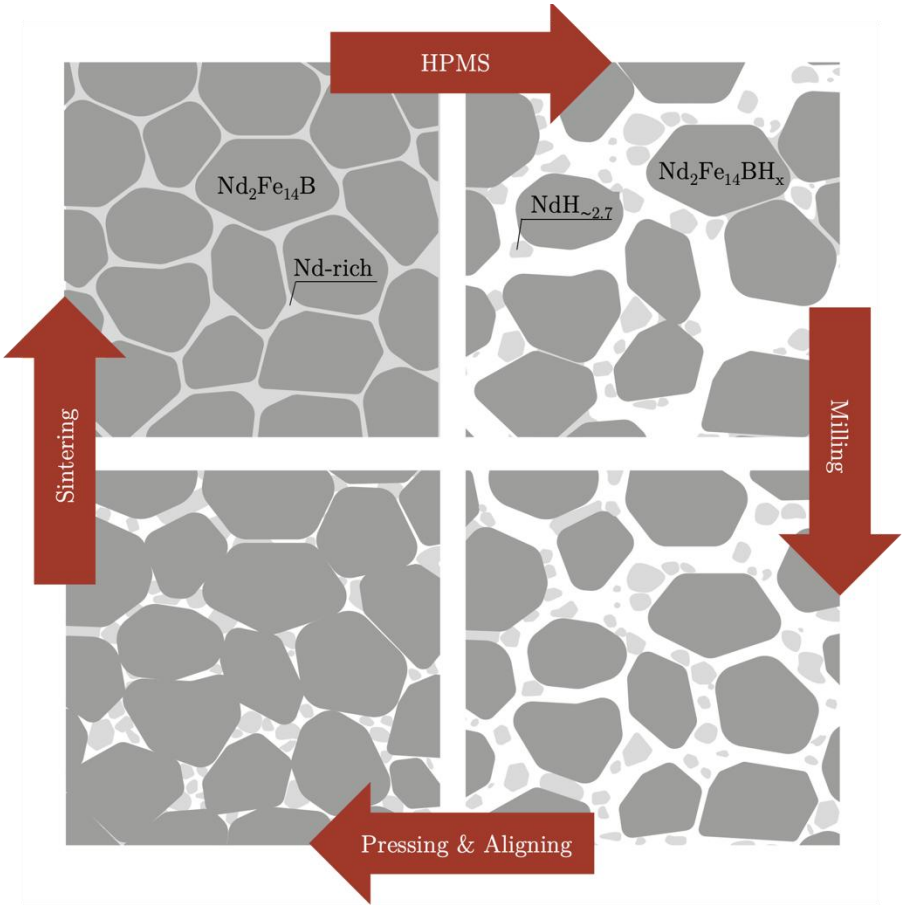


Figure 4.14: Short-loop recycling via HPMS and resintering.

4.2.1 Pre-processing

Pre-processing is hereby defined to comprise all necessary steps from finding and disassembling to preparing the magnets for HPMS. Generally speaking, while the pre-processing is a part of the recycling process of NdFeB, it is usually only partially carried out by designated NdFeB recyclers [11]. Post-consumer waste, meaning materials that are obtained after the end of life, for example from outdated or broken products, can vary widely in terms of material quality losses and contamination and may be difficult to trace. In comparison, production scraps, for example including magnets or mounted rotors or

motors that do not pass the quality checks, are typically easy to identify and are often supplied in bulk with the same materials and designs. Post-consumer waste is highly mixed and sometimes entirely unmarked [134]. Depending on the used recycling processes and whether there is a one-fits-all process or rather different separate ones, the motors may be sorted into specific treatment categories or even models or producers, the latter serving the purpose of returning the motors for a closed-loop scenario.
Dealing with consumer and industrial waste is regulated by different directives. The End of Life Vehicles (ELV) Directive of the European Union requires the last owner to ensure that the end of life vehicles are delivered to an authorized treatment facility [135]. There, available data about the present vehicle model is gathered electronically. After the removal of service fluids such as fuels, oils, etc., valuable reusable and recyclable materials are dismantled [134]. At this point, the electric motors can be collected as post-consumer waste. For Waste Electrical and Electronic Equipment (WEEE), the responsibility for ensuring that the products are treated in authorized facilities lies with the producers, while distributors must offer collection points for consumers [136]. Here again, the authorized facility must remove fluids and dangerous components and ensure separation of materials for recycling [136].

In the following sub-sections, the preprocessing from accessing the end of life (EOL) material, demounting and disassembling the NdFeB-containing components, and the preparation of the magnets for recycling are explained.

4.2.1.1 Access to electric machine waste streams

First of all, a distinction should be made between recyclable material from production scrap, also called pre-consumer waste, and from scrap derived from EOL scrap, also called post-consumer waste.

Production scrap tends to have the advantage that the information accompanying it can easily pass from the producing company to the recycling company, and that the material has not yet undergone a lifetime of use and corrosion. Pre-consumer scrap can make up around 25 % of the produced material [137]. However, in production, highly contaminated scraps can also be produced, such as grinding sludges. As a result, up to 30 % of the initial rare-earth material can already be lost before magnets are integrated into components. In this work, the focus is on the recycling of finished permanent magnets integrated into electric machines, regardless of whether they are pre- or post-consumer scrap.

The availability of recyclable EOL REE streams was predicted in 2013 by Binnemans et al. [21] to increase for magnet production scrap and automotive sources, remain stable for mobile phones, personal computers, and consumer electronics, while decreasing for hard disk drives. Reviewing this prediction in 2025 shows that most aspects are correct, however hard disk drives are being applied in large scales at data centers while being functionally extinct in personal computers as expected.

In Figure 4.15, some selected applications where NdFeB are integrated into electric machines are shown.

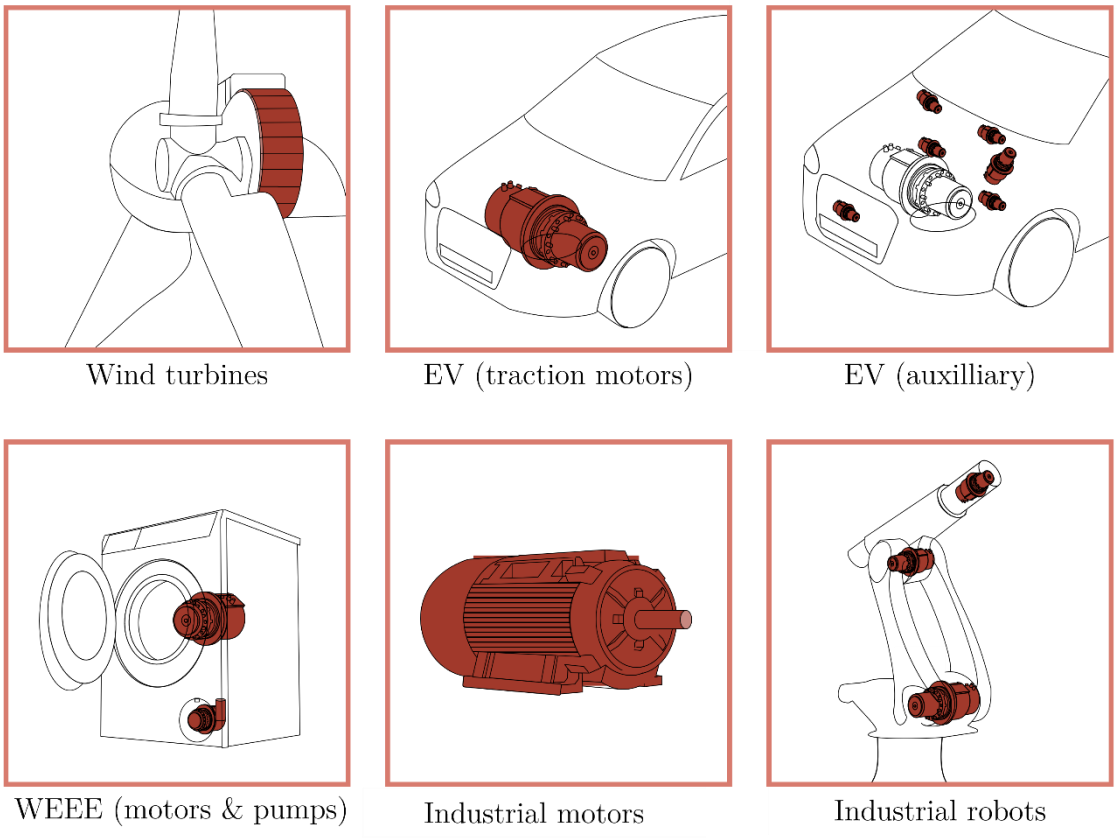


Figure 4.15: Some possible application categories of electric machines using NdFeB permanent magnets.

According to Busch et al. [34], around 80 % of Nd is deemed recyclable. The limitations arise from the efficiency of the recycling process, what is deemed economically viable, as well as the collection efficiency [34]. Whether a scrap source is valuable or not is decided by two main factors: firstly, the dissipation of the material coupled with the complexity, which dictates the efforts required for acquiring a certain amount of material [138] and secondly, the integration and protection of the magnets, which is decisive for removing them from the final sub-component and separating them from any contaminants [139].

On the other hand, while hard disk drives (HDDs) are essentially extinct in personal computer applications, they are widely used in server farms. As scrap sources with rather uniform anatomy, large numbers in few places, and a short lifespan, they are highly attractive [140]. For this source, recycling can already be highly automated – for electric motors, which are a much more diverse scrap source, such systems still need to be developed. Habibzahdeh et al. [141] predicted that the recyclability research regarding NdFeB will no longer need to focus on HDDs, but rather switch to the automotive and energy sectors.

Different predictions of return flows are offered in literature, for example, by Reimer et al. [142, 143]. The possible sources for NdFeB from electric machines are discussed and compared in the following subchapters.

Wind turbines
Wind turbines use large electric machines, generators, which can be built with or without NdFeB permanent magnets. Due to the commonly high HRE content and the sheer amount of large magnets (250 to 600 kg of NdFeB per installed MW [39]), which can be recycled, wind turbines are a highly attractive scrap source as the yield warrants basically any effort [39]. Even manual disassembly of the individual magnets can be economically viable, as a single magnet can offer a similar amount of permanent magnet material as a whole traction motor in an electric vehicle. The lifetime of wind turbines is estimated around 25 years [144]. While traditional drivetrain wind turbines do not contain NdFeB, direct-drive and hybrid systems, which are preferred for offshore applications due to their low maintenance, usually do [143].

Electric vehicles
It can be assumed that 90-95 % of current electric vehicles use electric motors with permanent magnets [145]. In 2023, Heim et al. [146] reported that electric vehicles are now the largest contributor to worldwide REE demand. This is by far earlier than predictions from 2012 – 2013, which assumed that only by 2050, electric vehicles would be the major REE end user [15, 147].

Electric motors are used in all non-fossil fueled drivetrains, including battery electric vehicles and many of these use NdFeB to maximize the efficiency [148]. Generally, battery electric vehicles usually use 1 to 4 traction electric machines with 1 kg to 3 kg of NdFeB magnets, with hybrid electric vehicle (HEV) motors being on the lower and battery electric vehicle (BEV) motors on the higher end [39, 149]. Larger models like E-SUVs usually use two or more traction motors [16]. Common traction motor types are primarily internal permanent magnet synchronous machines (IPMSM), as well as permanent magnet assisted synchronous reluctance machines (PMSynRM), induction motors and externally excited synchronous machines, like induction machines (IM) [16, 150]. Hybrid electric vehicles, including different types like plug-in hybrid, mild hybrid and full hybrid usually only use one additional traction electric machine in conjunction with a combustion engine, either in parallel or in series, on average holding 1.25 kg of NdFeB [39, 143, 151, 152]. Aside from the traction motor, power steering also oftentimes uses permanent magnet driven electric motors, and other smaller motors, pumps, actuators, sensors, and even audio modules may use NdFeB magnets, even though most small motors either use induction or other magnets to reduce costs. Overall, Bitzer et al. [153] state, that an electric vehicle has around 30 different applications for NdFeB. However, internal conventional combustion engine vehicles (ICEV) also use a number of auxiliary motors which can be equipped with NdFeB. Different sources assume between 175 g to 250 g of NdFeB on average to be built into auxiliary motors of a vehicle [143, 154].

The average lifetime of an electric vehicle is reported to range 13 - 14 years [43, 145]. For planning the recycling of NdFeB from EV, data on the vehicle sales in the last 10 - 20 years can be valuable, as for example compiled for 2015 by a study of Elwert et al. [155], where data on motor type is also provided. A major problem of planning recycling of NdFeB from electric vehicles is the large number of “vehicles of unknown whereabouts”, which are vehicles which are de-registered, but without a certificate of destruction, usually also without information if the vehicle has been treated or if it has been exported [156].

In terms of extractability of EOL traction motor magnets, Mueller et al. [72] classify them as moderately available but with low approachability in 2017 due to the motor designs not being recycling friendly. However, the magnets tend to have a high heavy rare earth content, therefore being an especially valuable source [21]. The most common type of motor used in electric vehicles are synchronous motors with internal permanent magnets due to the high power density, efficiency and sturdy fixation of the magnets which makes

them fit for high rotational speeds [146]. Compared to for example, industrial motors, traction motors operate under harsher conditions due to shock, vibration, higher operating temperatures, long service life and environmental influences [16]. Therefore, the protection of the magnets is of high importance, which can be achieved by very tight housings, which may be harder to demount, and coatings used. NdFeB in traction motors are almost all epoxy coated and the used adhesives for fixation are normally also epoxy based [145, 153]. The reason for this uniform choice of coating materials are the mentioned high requirements towards corrosion protection, but also the high rotational speeds in traction motors which are commonly running at roughly 20,000 rpm, which leads to high eddy currents in the magnets which heats them up if they are not properly electrically isolated. Furthermore, the use of segmented magnets, where a magnet is assembled from several pieces which are usually glued together to reduce the volume in which eddy currents can be generated is very common in traction applications due to the same reason.

WEEE

WEEE include household appliances like refrigerators, dishwashers or washing machines, small appliances like microwave ovens and vacuum cleaners, as well as IT equipment, entertainment electronics, power tools, sports equipment and toys, some medical devices and temperature regulating equipment like heat pumps and air conditioning. Sometimes, they are divided into white goods, which are utility driven appliances and brown goods, which are meant for entertainment and communication [157]. White goods with NdFeB-containing motors are reported to hold anywhere from 50 g to 325 g NdFeB and last around 7 to 15 years [145].

For WEEE, the main recycling steps are usually a manual pre-sorting process followed by partial disassembly, shredding and sifting [24, 115]. Full disassembly is likely too costly due to small amounts and low quality of the built-in magnets [24], however shredding has been reported to lead to loss of the magnetic material [39]. Extending over the scope of NdFeB, Alatalo et al. [12] report that recycled iron from shredded material usually has a bad quality due to the introduced copper content, which is present in most motors.

Personal computers, loudspeakers, electric power tools, electric personal care- and cooking tools as well as other small home electronics are commonly not disassembled, as the yield is too low for the required effort [39]. This is aggravated by the trend of miniaturization of components [138]. Therefore, this sources as one with a high number of individual products, could be potentially tapped by employing a recycling procedure which does not rely on dismantling.

Light means of transport

Light means of transport with electric motors also fall under WEEE regarding the regulations for disposal. Examples for this category are e-bikes, e-scooters and e-motorbikes. These typically contain 60 g – 350 g of NdFeB and a typical lifetime of 6 years according to Rizos et al. [145]. E-scooters as emerging transport options within cities, often available by rental today can be assumed to all contain NdFeB [145]. According to Couto et al. [158], all magnets in e-scooters are found to be Zn coated, a finding that our own investigations support. Furthermore, the use of low-grade NdFeB, oftentimes with high Ce substitution or high variations in composition within one motor appears common.

Industrial motors, pumps and robots

Industrial motors can be an attractive source of recyclable NdFeB material depending on the ease of disassembly, the amount of permanent magnet material and the worth of the alloy. Generally, a good transmission of information between the producer, user and recycler can be assumed if all actors cooperate. Take back schemes and closed-loop

recycling also seem very viable for such applications. It is generally understood that industrial motors and industrial pumps tend to operate connected to a power source without dramatic weight and size constraints, which is why many designs do not require permanent magnets. The usage rate was therefore reported to be rather low and the NdFeB mass being used is estimated between 50 and 325 g with an average product lifetime of around 20 years [145]. An exception are industrial robots. Here, according to Rizos et al. [145], 560 to 3360 g of NdFeB are used in almost all industrial robots with an average lifetime of 12 years. High HRE contents can be found in some applications, especially when the motors are used in high temperature environments [36].

4.2.1.2 Identifying and sorting

The first challenge of recycling sintered NdFeB is finding out, whether a product contains permanent magnets, whether they are sintered NdFeB and where they are mounted. As other magnet types, like for example ferrites and bonded NdFeB are routinely used in electric machines as well and require different recycling methods than NdFeB, sorting is essential.

How much of a challenge this is, can be illustrated by reporting on efforts conducted during the INSPIRES Project:

In the INSPIRES Project, 440 WEEE items, corresponding to 6000 – 10,000 kg of WEEE waste were sorted and dismantled by the Slovene waste management company SUROVINA. The items contained WEEE products like power tools, kitchen tools, headphones and loudspeakers as well as washing machines, ovens, dishwashers and e-scooters among others. A total of 144 WEE items were found to contain NdFeB with a total mass of 9.62 kg of NdFeB. However, the distribution of contained magnet weights is far from even: The distribution graph shown in Figure 4.16 demonstrates that the majority of the NdFeB containing products only hold very small amounts which are likely not worth dismantling for. This is especially relevant for large white wares like dishwashers, ovens, washing machines and dryers, where oftentimes only a very small door magnet was present. In most cases, the large white wares held Ferrite-powered motors.



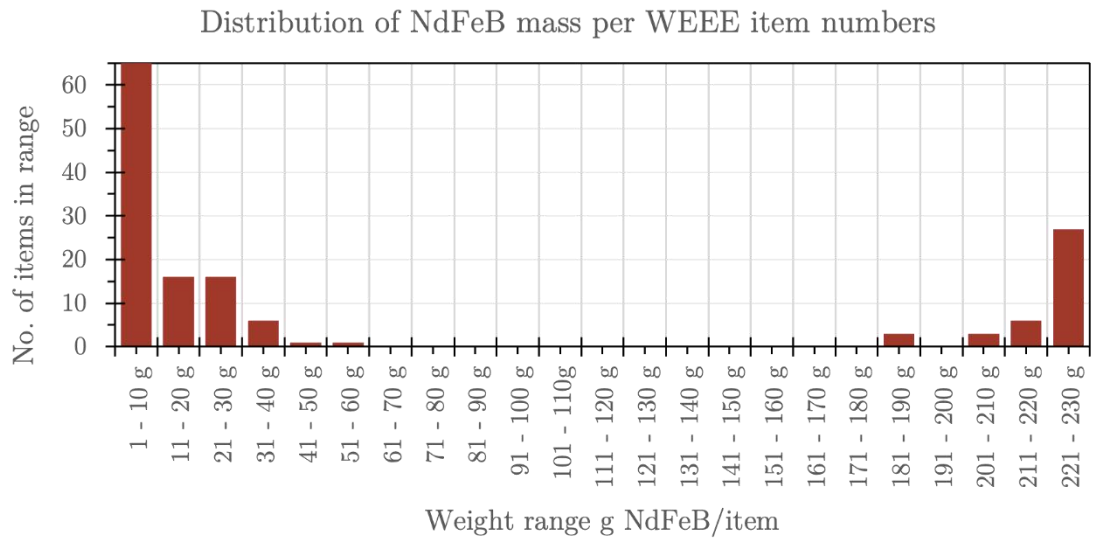


Figure 4.16: Disassembly results from the INSPIRES project, only showing the items with NdFeB enclosed in the graph below.

There are different solutions to alleviate the effort: Firstly, labelling is a powerful tool to identify where certain materials are present. Labelling is employed for various components and materials already, with leading examples being plastic resin codes, the seedling logo for compostable waste, and the WEEE label, indicating that the product cannot go into municipal waste or the battery symbol indicating also contained dangerous elements.

This demonstrates that implementing labels to promote the recycling of critical raw materials like the REE in NdFeB is a viable regulatory tool. In fact, in 2024, the EU launched the Critical Raw Materials Act (CRMA), where Article 28 introduced the requirement to label several types of products that typically contain permanent magnets. The presented dissertation work was integrated into the work for establishing the Implementing Act for Art. 28, as the working group for the creation of a technical specification as a basis for the Implementing Act was led by the author. This is more closely elaborated on in section 6.3.

Another option to improve recyclability is using detection systems, however, this rather limited for magnets found deep within larger product due to the highly localized magnetic field. However, some detection systems, like one developed within the REESilience project

[140] or a method to demagnetize and then monitor remaining magnetic fields to sort can distinguish different magnet types [159].

What should not be underestimated is also the experience and pattern recognition of professionals working in the waste management field, who, with growing interest in recycling NdFeB will likely be able to find built-in magnets much quicker in the future. However, with rapid development and diversification of electronic equipment and other WEEE, it can be assumed that it still poses a challenge in the future and leads to the waste management chain to slow down.

Therefore, it can be concluded that labelling is likely the most viable option for all types of products, while for small products detection systems may be viable.

4.2.1.3 Disassembly

Demounting or disassembly is characterized by the separation of joint components. It is generally a relatively time-consuming step which is also thought to be "non value added" [160]. Soh et al. [13] state, that it is very important to either have instructions or that the disassembly is very intuitive and that tools are accessible and easy to identify. Disassembly can be total or selective for especially desirable or dangerous components [161].

Disassembly is a main focus of research both in design for recycling, design for remanufacturing, as well as other methods under the umbrella of design for sustainability and is thus methodically researched by a significant number of groups. Chang et al. [160] summarize how disassembly can be planned, reviewing different studies and presenting methods to do so. This includes for example planning by using petri nets, graphs, geometry-based planning, dynamic-based planning, CAD-based planning and intelligent methods for planning disassembly [160]. Some simplified examples of these methods are shown in Figure 4.17. Another subject of research is the calculation of the disassembly time as a marker for the efficiency, which can either be based on the product and present connectors or on basic motions, as reviewed by Vanegas et al. [162]. When demounting, the type of joint is determinative for the effort required, as a single joint can require between one and a high number of individual operations to reverse it. This includes operations which are not directly conducted on the product, like searching a tool, testing it and taking another size of the tool to test it again. For example, to quantify the time required for different joint, a study tested the time required to separate different joints as well as the reversibility and damage done to the components [163]: Male/Female joints, socket joints and snap fits had a relatively short disassembly time of an average of 10 s. Screws and threaded fasteners take 20 – 100 s, joining by adhesive, soldering or welding showed excessive, hard to generalize disassembly times [163].

As literature is very diversified and well available here, and many papers are already summarized by Chang et al. [160] and Vanegas et al. [162], the results and ideas are not reproduced in detail in this work – instead, the specific demounting steps will be addressed in the following sub-sections.

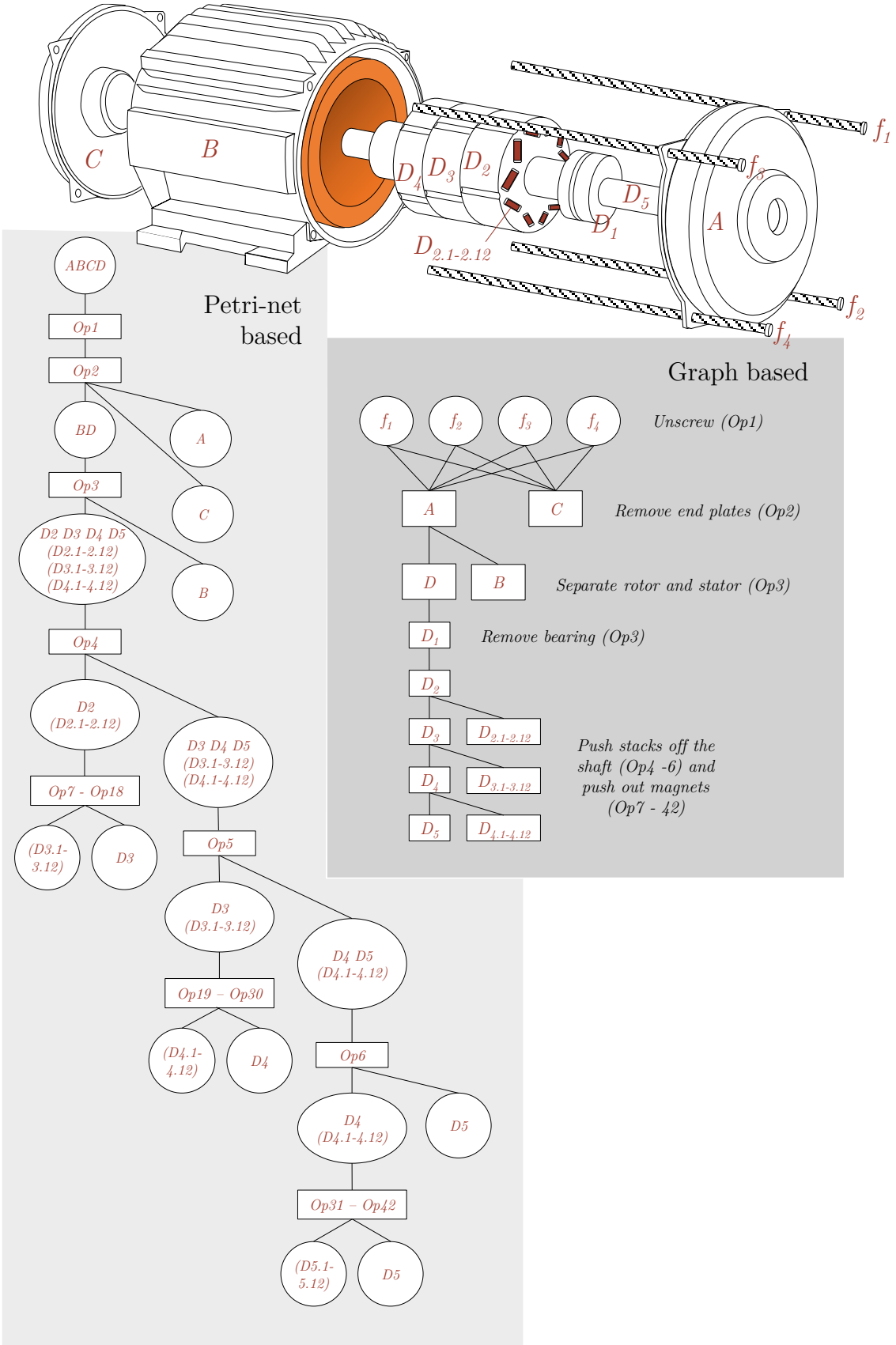


Figure 4.17: Depiction of two different types of disassembly planning.

General disassembly operations

Demounting has a range of benefits compared to shredding or other "shortened" procedures like in situ treatment of the magnets without removing them first, but also some disadvantages. Positive aspects are that close to ideal separation can be enabled to achieve better quality material while loss of material can be minimized [146]. Furthermore, demounting allows for reuse and remanufacturing aside from recycling [146]. On the other hand, the time consumption, especially in terms of manual labor and required precision instruments to disassemble can be too expensive to be economical at all.

The disassembly can be manual, semi-automated or automated. Research regarding the automation of the disassembly and by human-robot cooperative is conducted by a research group at KU Leuven, where the most important criteria for robotic disassembly were identified as connector conformity, component graspability, task manipulability and task complexity [164]. This means, the connectors should be demountable non-destructively, non-adhesive and ideally require standard tools only [164]. The components should not be too heavy and accessible to the robot as well as offering enough space to grasp it. Lastly, per operation, only one tool should be required at a time [164].

Depending on whether components are disassembled by robots or humans, the allowable weights and the requirements for the graspability may vary, as human hands are much more dexterous but more limited in terms of the component mass that can be safely removed. Nevertheless, the criteria by Pulikottil et al. [164] are sensible for both robotic and human disassembly operations. It is further advisable to simplify the parts, try to use as much standardized parts as possible and to generally reduce the variation of components.

A significant advantage of manual or robot-aided disassembly is the high flexibility with regard to the variety of devices and their condition. Fully automatized disassembly without flexible robots would only be able to deal with a very narrow range of products. With commercially available tools, expert personnel or a human-robot cooperative can dismantle the old equipment with a high degree of sorting purity, with the main disadvantage being the high expenditure of time and personnel.

While they may make further separation challenging, with destructive methods instead of disassembly, there is greater format flexibility, which can function independently of existing connections and geometries. For example, the motor housing can be broken hydraulically to remove the rotor and stator. In the next step, the assemblies can be separated into two parts by hydraulic splitting by using a pulling device to separate out the copper wire [165]. Another method effectively saws a whole motor in half, pulling out the stator windings [166], in conjunction with this, it may be possible to remove the magnets from the rotor as well, for example by the methods "demag to demount" or an in situ treatment. Both of these methods are explained in detail in sections 5.2 and 5.3.

For connecting two parts with each other, permanent or non-permanent fixings can be used. They perform differently in terms of reversibility, meaning that they can or cannot be opened without damage as well as disassembly time.

Different methods for joining are systematized and explained in DIN 8580 [167] and DIN 8593 [168]. The groups of joints and whether they can be disassembled destructively or potentially non-destructively is shown in Figure 4.18.

Male/Female joints and socket joints showed good reversibility, while disassembling snap fits often lead to damage on the joint components [163]. Threaded fasteners and screws themselves sometimes showed damage, but did not damage the components [163]. The reversibility of permanent joints was, as expected, poor, due to requiring destructive disassembly [163].

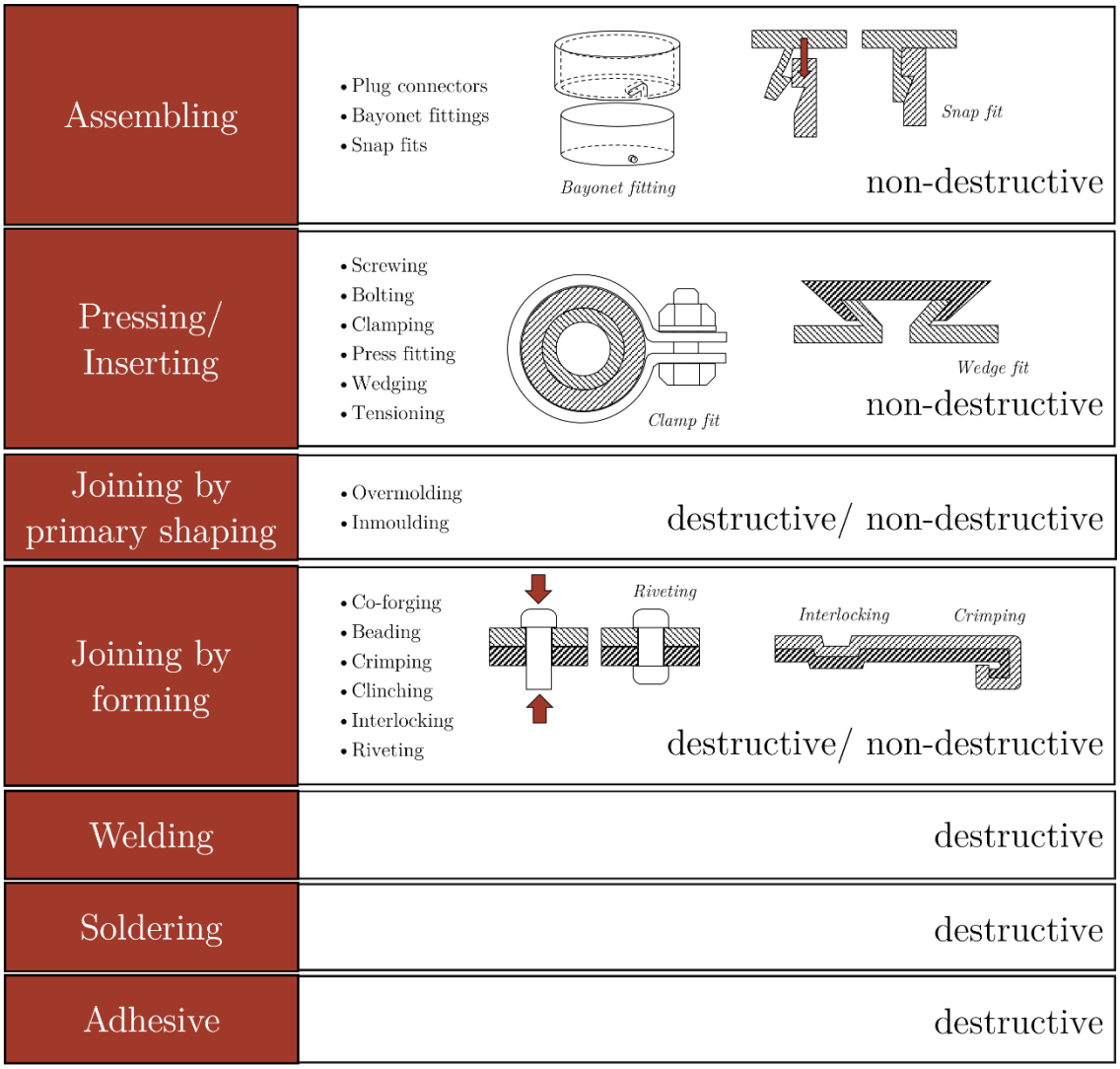


Figure 4.18: Examples for typical connectors, their categories and whether they can be disassembled destructively or non-destructively according to DIN 8580 [167] and DIN 8593 [168].

Many influencing factors, such as raw material prices, labor costs, logistics and energy prices, affect the economic efficiency of the partly manual disassembly. Since full automation does not seem promising due to the complexity and variability of designs, partial automation can reduce costs and increase productivity and flexibility [169]. One challenge is the continuous supply of larger quantities of identical or similar electric motors in order to create product-specific disassembly processes [165]. This is basically only given in a closed loop scenario of a high quantity product. This would result in reduced time and manpower requirements, thus increasing cost-effectiveness. At the same time, flexibility with regard to product variants, product states and plant utilization decreases. In addition, higher investments and qualified employees are required.

Separating the motor from the periphery
Motors are built into different kinds of products: in WEEE waste or for small, auxiliary motors in vehicles, this step can be quite straightforward. In more complex assemblies like the drivetrain of a vehicle, it can be more challenging. Generally, the motor must be disconnected from any power source to ensure dismantler safety. Then, accessibility of the motor must be achieved. Oftentimes this requires removing other components first. Removing electric traction motors from EV can be especially challenging due to their position not being as standardized as for combustion engines, as well as the high weight and prior safety preparations [170]. However, rather "crude" methods can be employed, like grabbing the motor block with a clamshell excavator, lifting it and "shaking" the rest of the vehicle off.

Separation of rotor and stator
Separating the rotor and stator requires to open the housing first. Some housings use non-permanent fixation like screws, whereas others rely on welding, adhesive and other permanent fixations methods. This step may take place either at the dismantler or already at the recycler – the dismantler could be interested to remove other valuable materials like copper and the housing material, oftentimes aluminum first, before forwarding the rotor to the recycler.

The challenges of this step are the vast variety of models, magnetization of the magnets and usually also the lack of knowledge regarding the obtainable materials inside the motor. Several studies have elaborated on the disassembly of electric motors, for example regarding the unscrewing operation [171], the forces required for disassembly of different components [172], or the automatic detection and tool choice [173]. An example of how automatic detection, tool choice and unscrewing could work is shown in a design is shown in Figure 4.19. After the housing is opened and components which could hinder rotor removal are removed, (e.g. bearings on the shaft outside the housing), the rotor can be pulled out. Liang and Pham [172] report that it is possible to use an optimized disassembly path based on magnetic field calculations to avoid damaging the rotor and stator.

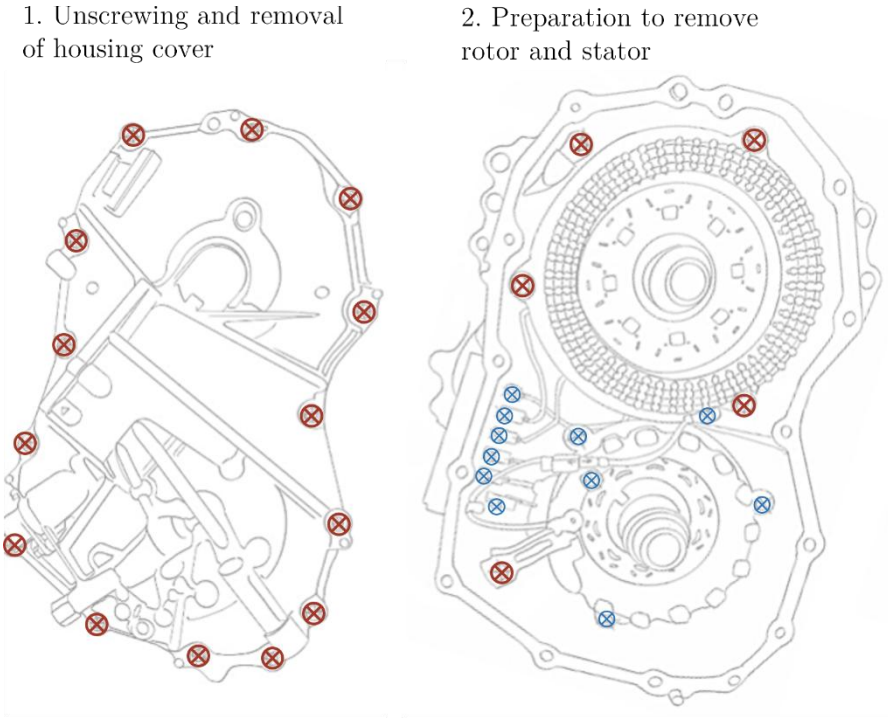


Figure 4.19: Example of a housing design that allows automatized unscrewing. All screws can be visually detected and removed from the top. Only two types of screw heads are used. The housing cover can be lifted by grasping of suction, the stators and rotors can be removed by grasping. The drawing is based on photographs provided by KU Leuven.

Disassembly and magnet removal from the rotor

For the rotor that mostly consists of electric steel sheets, a steel shaft, (coated) magnets, lacquer and adhesive, different treatments are possible.

WEEE are often sorted, partially dismantled, shredded and then sifted [115]. The rotor could be demagnetized and then shredded to separate the magnets and the steel parts. But the separation of steel and NdFeB is quite challenging and the exposure of finely ground NdFeB to oxygen, residual adhesives, etc., may lead to detrimental effects on the quality of the recycled material, as it was shown for the shredding of magnets from hard disk drives in a study [33].

A common approach is completely removing the magnets from the rotor. For surface mounted permanent magnets, the removal of the magnets is usually quite simple. Klier et al. [174] present methods to remove SPM by shearing them off and IPM by pressing them out both using specialized tools.

For this, demagnetization is required beforehand and, in some cases, the removal of the shaft is as well, since the rotors can be skewed and the stacks need to be separated from the shaft in order to allow the magnets to be pushed out.

After that, the magnets can be decoated and further treated. The advantage is that contamination can be minimized by offering the opportunity of different tailored coating removal processes and processing reactors can be kept small, but the effort required can be quite high. Though, the efforts can be mitigated by suitable designs.

As this process is very problematic in terms of time consumption, possibilities for automation and format flexibility, in this work, different methods have been considered and researched to facilitate demounting of the magnets from the rotor. These methods are further explained in section 5.

Alternative: shredding
When NdFeB should be recycled without any demounting, shredding is an alternative. Small E-waste would most likely be shredded instead of demounting the magnets [39].

However, shredding has some major drawbacks like a higher risk of contamination with most NdFeB ending up in slag due to being magnetic and being sorted into ferrous scraps [24, 39, 43]. However, Sprecher et al. [33] raise an important point: despite the drawbacks and the worse environmental footprint compared to demounting and recycling, shredding and recycling still far outperforms primary production ecologically.

The shredding procedure itself is usually accompanied with different pre-treatment and sorting steps. First of all, scrap is sorted by an eddy current separator, then it must be demagnetized and shredded [175]. The material is separated into three fractions: non-conductive, conductive or conductive and magnetizable [175]. When remagnetizing the last fraction, and demagnetizing again, materials can be sorted by Curie temperature, potentially filtering out SmCo or even high-grade NdFeB magnets with an enhanced Curie temperature from lower grade ones [175, 176].

4.2.1.4 Demagnetization

Especially sintered NdFeB are challenging and dangerous to remove from rotors due to their strong magnetic field without demagnetization. Shredding is also a risky operation when dealing with magnets, as shredders may be clogged by magnetized material. Due to this, demagnetization is usually employed in some step of the recycling process.

The most straightforward method to demagnetize is heating the magnet to a temperature above the Curie temperature, which is around 312 °C for basic NdFeB, but can be higher with the addition of certain alloying elements [117, 177]. Thermal demagnetization can be conducted in air, vacuum or protective atmosphere at varying temperatures. Convection ovens, IR radiation or inductive heating have been reported to be used [155, 178]. At elevated temperatures and oxygen-rich atmospheres, magnets can oxidize, forming an external oxidized layer and an internally oxidized zone when exposed for a longer time [98]. During thermal demagnetization, adhesives and coatings can be disintegrated as well, potentially creating hydrocarbon fumes [21] and even Zn fumes from Zn coatings. Without proper deduction and waste gas treatment if required, health hazards can arise [178]. Furthermore, equipment can become damaged, especially when the demagnetization is done under vacuum. Thermal demagnetization can also serve another purpose, like for sorting magnets by Curie temperature [159, 176] or for removing adhesives and organic coatings [176, 179]. However, choosing suitable parameters like a sufficient temperature and atmosphere are required to achieve this [176, 178, 180].

Methods aside from thermal demagnetization, are for example degaussing with a solenoid [181] anhysteretic deperming which involves applying an alternating field with decreasing amplitude, or a combined method using induction to demagnetize with a field and heat the magnets at the same time [181–183]. However, demagnetization in a field requires utmost care in terms of designing a fitting demagnetization tool and oftentimes fails to eradicate residual magnetization completely [183]. For this, computational methods can be used to make the process more efficient [184]. Short-circuiting of rotor and stator can also be used to demagnetize, however, it appears that this method only offers partial demagnetization [185].

It should be noted, that if the magnets do not need to be demounted and handled, a dedicated demagnetization process could potentially be omitted, as HPMS processed powder is demagnetized [39]. Elwert et al. [155] also claim that even for demounting of magnets, demagnetization may not be necessary – however for reasons of safety and contamination mitigation, it should still be recommended.

4.2.1.5 Separation by cleaning and coating removal

Separation of contaminants from bulk magnets can be done by several methods. A simple method can be the removal of the coating, oxidized surfaces and adhesive residues from the bulk magnet by sandblasting or grinding it.

Aside from mechanical methods, solvolysis is a possible route for organic materials like epoxy coatings and adhesives [186] and anodic coatings like Zn [158] with common, well available solvents and acids. For Ni-based coatings, a solvometallurgical method with elementary Br is reported [187].

Finally, also thermal removal of organic materials and Zn is well possible as mentioned in the last section on demagnetization.

4.2.2 HPMS processing

The HPMS process has been developed and patented by the University of Birmingham [39, 44, 133]. It involves exposing a NdFeB bulk magnet to hydrogen atmosphere at room temperature and atmospheric pressure [39]. Different studies varied the properties, showing that a higher temperature accelerated the reaction, but reduced hydrogen solubility and that lower pressure lead to finer particles, with slower processing [141, 188–191].

The φ-phase is able to absorb up to five hydrogen atoms per formula unit, or 88 hydrogen atoms per unit cell [99].

HPMS is performed at between room temperature to 400 °C, where the solubility of H is rather high, not to be confused with hydrogen decrepitation disproportionation and recombination (HDDR) processing above 650 °C [192]. During HPMS, Nd reacts with H to form different types of hydrides, mainly NdH_2 and NdH_3 [189]. On average, $NdH_{\sim 2.7}$ is formed in an exothermic reaction [39]. To achieve maximum hydrogen uptake, the temperature should be limited to below 100 °C [188], whereas for large amounts of highly reactive material, it may be necessary to cool the reactor.

The relative change in volume of the Nd-rich grain boundary phase is higher (given as 16 % [189]) than for the φ-phase (given as approximately 5 % [39, 189] or 1 - 2 % [193] in different works, related to the hydrogen uptake of the matrix), leading to high strain and decrepitation, as the formed Nd-hydride is very brittle, leading to a friable powder [39]. Further, due to the distortion of the φ-phase caused by the interstitial absorption of H, the resulting powder is essential demagnetized [39]. Here, the anisotropy field is greatly reduced due to the anisotropy contribution of the Nd sublattice being highly affected by its local atomic environment and distances [194, 195]. During the process it is important to leave enough space to allow this necessary volume expansion and avoid jamming and clogging of the arrangement with the resulting, coarse powder [51]. Some magnets during hydrogen processing are shown in Figure 4.20. The process was also illustrated in Figure 4.14.

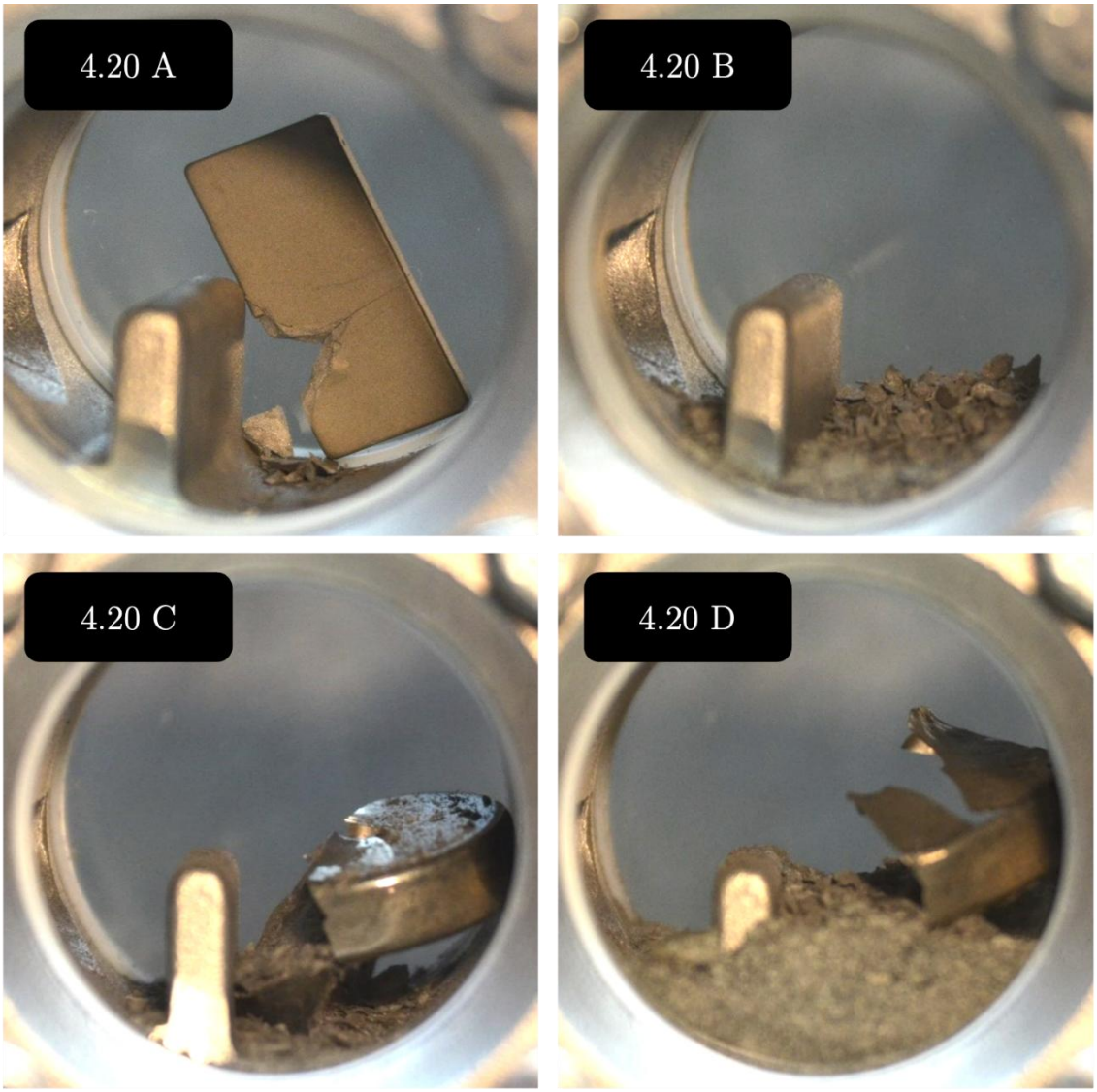


Figure 4.20: Magnets during the HPMS process. Panel A shows a passivated magnet just after the start of the process, B to the end of it. The same is shown in C and D for a Ni-Cu-Ni coated magnet. It is visible that some coatings come out of the process basically in one piece, while others blend in with the powder and are challenging to remove.

4.2.3 Powder processing and resintering

4.2.3.1 Degassing

Nd hydrides are rather brittle and stable, while degassed material, meaning the hydrogen is removed, is again reactive [196]. On the other hand, the hydrogenated φ-phase is also brittle, which may lead to transgranular cracks during milling [141] and less magnetically anisotropic due to the distortion of the crystal lattice like explained in before [194, 195, 197]. Thus, alignment of non-degassed material is impaired. The timing of the degassing should therefore be chosen carefully.

The φ-phase starts to degas below approximately 200 °C, then NdH_3 is degassed into the more stable NdH_2 until 400 °C and finally at around 550 - 650 °C, NdH_2 also decomposes [191]. Practically, higher temperatures between 700 °C and 1050 °C under vacuum have been reported for degassing procedures as well [198–200]. The degassing temperature should be chosen according to what is sought to be achieved: For pressing and sintering, degassing the φ-phase at 600 °C for 2 h [201] is enough to restore its magnetic anisotropy to ensure proper alignment, as the remaining H will degas during the sintering treatment. For use of the powder in bonded magnets, there is no additional high-temperature processing step which is why full degassing is required [198]. Generally, the degassing temperature must be high enough to achieve the required H removal, however it must be mentioned that degassing temperatures should be kept low, as a study reported that degassing temperatures beyond 700 °C lead to a decrease in magnetic properties [191], which can likely be attributed to oxidation.

4.2.3.2 Milling

Most commonly, for milling NdFeB, a jet mill is used. For this, compressed N is introduced tangentially into a chamber through a nozzle. This creates a vortex on which the NdFeB particles are carried, colliding with each other and fracturing into smaller particles. Particle separation is governed by a balance between centrifugal force and drag force. The centrifugal force, stronger for heavier particles, presses them outwards so they keep colliding until the particle size is reduced. Finer particles are more influenced by drag and thus, are pulled inwards and are carried to the outlet. When classifiers are used, the classifier wheel creates a centrifugal field which only small enough particles can pass. This way, typically single crystalline particles are achieved [7]. As an optimal particle size, 3 µm is often cited. Smaller grain sizes in the finished magnet are preferable, however, too fine particles can cause oxidation and an uneven distribution of the grain boundary phase [103]. More recent research has addressed refining the jet milling process, for example by using He or H instead of N to achieve smaller particle sizes down to single domain grain sizes [202–204].

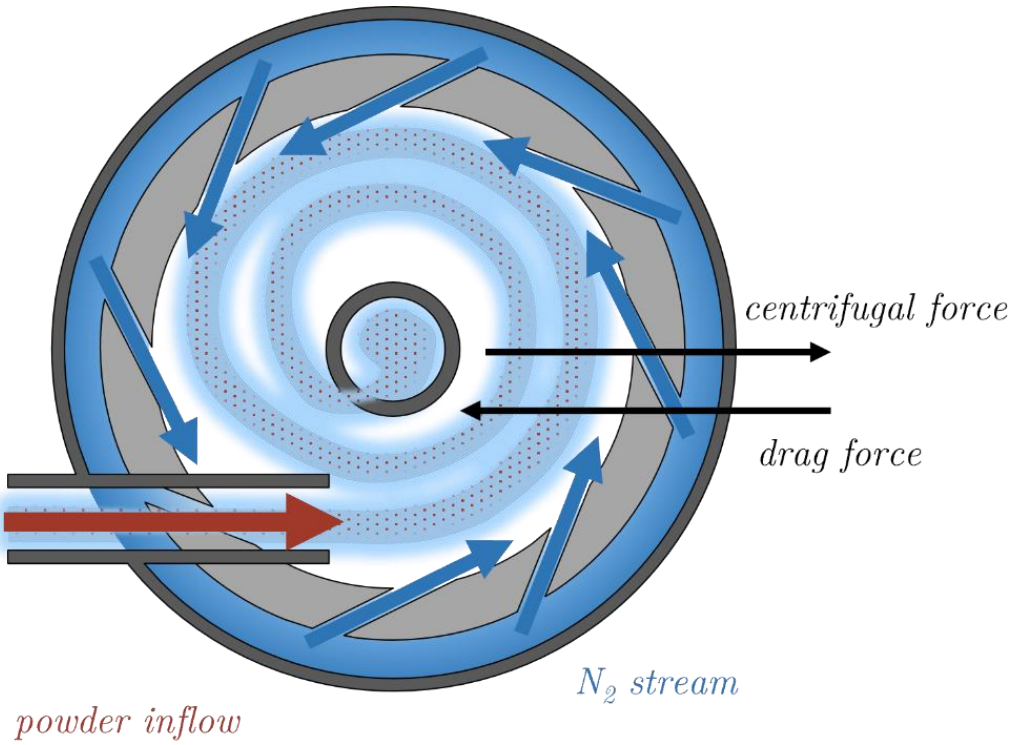


Figure 4.21: Function of a jet mill: particles are fed into a circular gas stream and collide to mill. Large particles are pushed to the sides due to centrifugal force, small particles to the middle due to drag and can exit the mill.

In lab scale operations, other milling methods can be applied, such as high energy ball milling. In conjunction with milling, other powder preparation steps can be employed. NdH_x can be added to compensate for oxidized, inactive Nd before sintering [196], which will degas during the heating and then liquify to wet the grain boundaries. If no active Nd is added, the magnetic properties will deteriorate with each recycling process [196].

After milling, the powder can be used for producing a new magnet or it can be further processed, for example, for separation from contaminants from the powder.

4.2.3.3 Separation of contaminants from powder

In some cases, however, it is necessary to separate contaminants, namely coating residues from Epoxy, Zn, Ni or Ni-Cu-Ni or some less common coatings, adhesives residues and oxides from the powder.

Sieving or sifting

The powder can be sieved to remove contaminants which are coarser or finer than the powder. The size of both the magnet powder and the contaminant can be modified by milling, for example, which may affect both differently and potentially separate the contaminant from the powder. Employing sieving together with milling is more closely described in section 5.2. Sifting, as used in jet milling relies on particles being light enough and getting enough buoyancy force in an upwards airstream due to enough surface per weight. Sieving and sifting are good candidates for contaminants that can be mechanically separated from the φ-phase and that have different milling characteristics than hydrided or non-hydrided NdFeB, for example by being significantly less brittle or more ductile. This especially applies to Ni and Ni-Cu-Ni coatings.

Chemical separation
Using a leaching procedure, some contaminants may be removed from the powder, especially if they have a lower standard electrode potential than the φ-Phase. This applies especially to anodic coatings, like Zn. A method suitable to remove both the oxidized Nd-rich phase and potentially also anodic coatings is leaching the powder in an acid, like investigated by Schieren et al. [205].

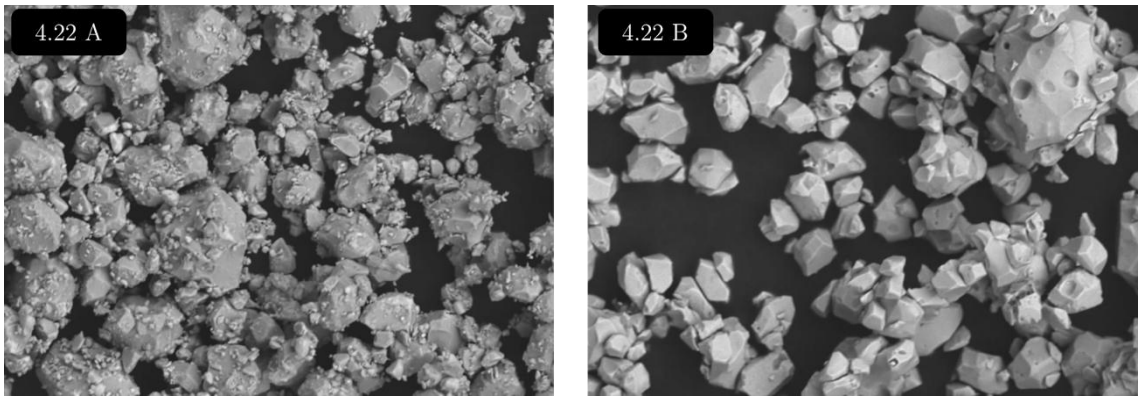


Figure 4.22: Ball-milled NdFeB powder before (A) and after (B) chemical treatment.

Physical separation techniques
Physical separation techniques rely on differences between two or more particles in terms of their magnetic properties (e.g., for magnetic separation), conductivity (e.g., for electrostatic separation), particle shape (e.g., for sedimentation separation), or density of the materials. To separate Epoxy and other polymeric substances, as well as non-magnetic metal residues from the magnet powder, potential options could be magnetic separation, electrostatic separation or separation by density appear promising. However, it is required that the polymer and the magnetic material are separate particles to behave significantly different [206]. Furthermore, for loose polymer parts, gravity separation using viscous liquid, for example tetrabromoethane, or electrostatic separation via corona charging is possible [206, 207]. A very common reported set-up, like shown by Helland et al. [208] is to combine a magnetic drum and an eddy current separator, which is shown in Figure 4.23.

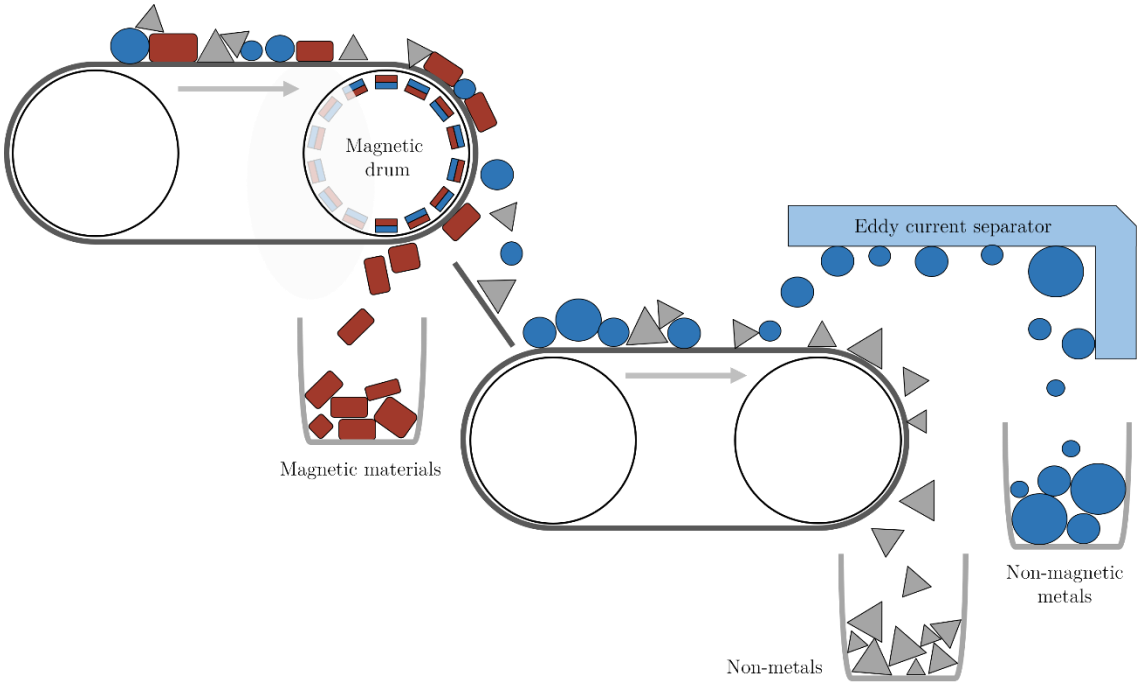


Figure 4.23: Separating unit for shredded material using eddy current and magnetic separation. Based on Helland et al. [208].

In the magnetic material fraction, also called ferrous fraction, different materials can end up: steel parts and permanent magnets like NdFeB, SmCo, AlNiCo and hard ferrites [209]. Ihne et al. [209] compared their densities, Curie temperatures, relative permeability and electrical resistivity and found that the Curie temperature of NdFeB which is significantly lower than for the other magnetic materials can be utilized to separate NdFeB from the other permanent magnet materials. This is a simple method that has also been applied in the INSPIRES project earlier and was reported by Baberg et al. [175] as well.

4.2.4 Pressing and resintering

The processes for producing recycled sintered magnets are inherently the same as for virgin magnets, which is why the processes are only shortly explained here.

4.2.4.1 Pressing and aligning

To produce a bulk magnet from powder, the powder must be pressed and aligned. This can be done in sequence, usually with the alignment coming first, or in parallel. Typical press types are isostatic presses and uniaxial presses [7, 210]. Rubber isostatic pressing is suitable for different magnet shapes and achieving homogenous density with simple means, which makes the method a viable option for laboratory scale processing [211]. Other methods usually produce blocks or prismatic shapes and have limited shape flexibility.

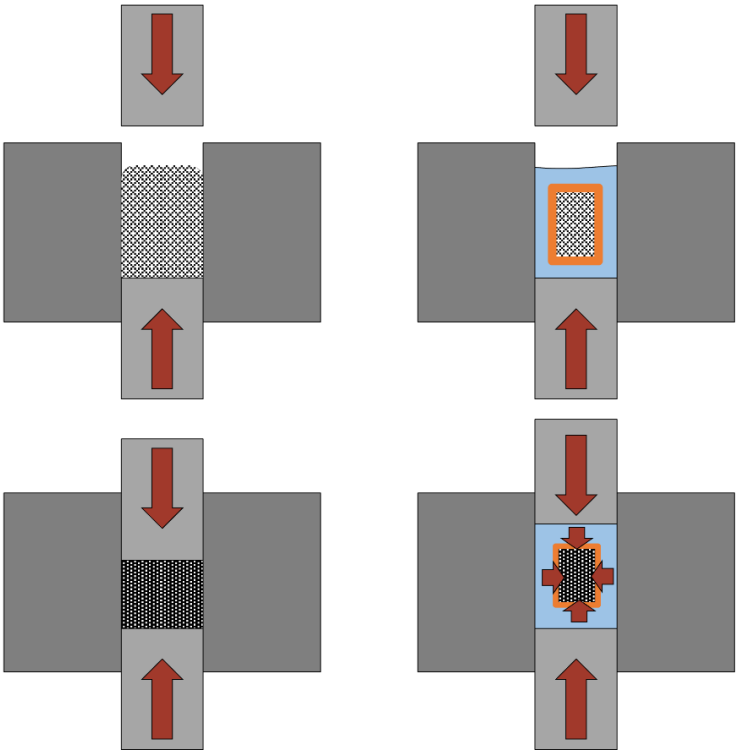


Figure 4.24: Uniaxial pressing principle (left) and isostatic pressing principle (right).

4.2.4.2 Alternative shaping and aligning methods

A mentionable method to produce parts for sintering is the shaping-debinding-sintering method, using either metal injection molding or powder extrusion molding to produce a green part, which consists of powder in a shapeable polymer matrix, which is then removed in a debinding process before sintering [64, 212, 213]. In metal injection molding, multipole magnets can be produced [214], while in powder extrusion molding, only recently a tool has been developed to magnetize green parts in line during continuous extrusion [215].

4.2.4.3 Sintering

Liquid phase sintering is usually done under vacuum, sometimes also protective gas atmosphere [7]. Ideally, virtually no grain growth occurs and 98 % density is achieved [7]. However, in reality, this is highly challenging to achieve.
For this, the compacts are heated in a vacuum. NdFeB is an eutectic system, where above eutectic temperature (655 °C for a ternary NdFeB alloy), the intergranular Nd-rich phase goes into liquid state [92]. This eutectic melting occurs locally at the grain boundaries and interfaces with the $Nd_2Fe_{14}B$ phase. According to Okazaki et al. [216], the grain boundary phases make up around 6.6 vol.% at 40 °C. Due to the liquid phase forming, capillary forces on the solid particles cause a densification burst, which Taylor et al. [92] describe as the first stage of liquid phase sintering, called "rearrangement" [92, 112, 217].

The active Nd forms the liquid phase and partially dissolves Nd from the φ-phase grains [92, 111]. The Nd is then re-precipitated onto the grains. Smaller grains can be fully dissolved and re-precipitated on larger ones, leading to grain growth [92]. This step, referred to as the "solution reprecipitation," is dominated by dissolution and diffusion and occurs at roughly 900 °C [92]. The last step usually is continued grain growth and densification, which is governed by solid-state diffusion. However, the microstructural evolution is governed by both grain coarsening and grain-shape accommodation, which are coupled and driven by the reduction of interfacial energy. Grain coarsening, meaning mass transfer from smaller grains to larger grains, and grain shape accommodation, meaning local morphological adjustments occur simultaneously and are mediated by diffusion or dissolution-reprecipitation mechanisms. The magnets reach full density at 1000 – 1150 °C [92, 218]. The process is shown in Figure 4.25.

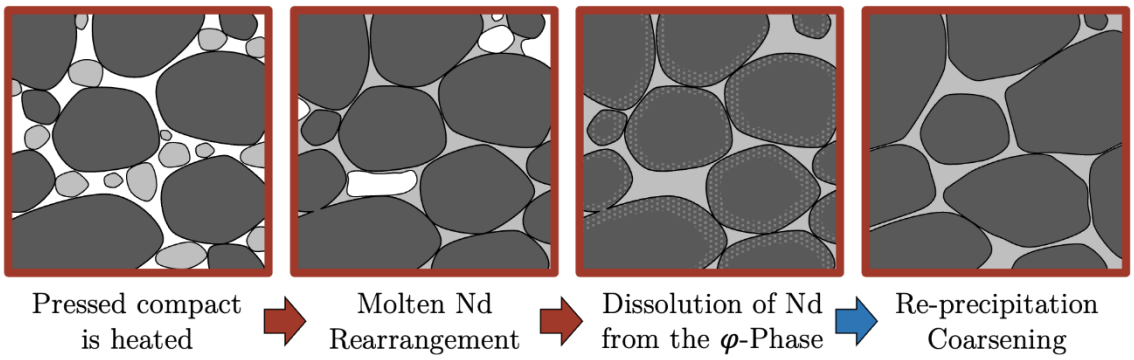


Figure 4.25: Liquid phase sintering stages.

4.2.4.4 Post-sinter processing

The magnets are subsequently shaped, often annealed or grain-boundary-diffused, and coated. Post-sinter annealing is performed to increase the magnet's coercivity by smoothing grain boundaries, mitigating surface defects, sharp corners and edges while also ensuring even wetting with the Nd-rich phase [89, 219]. Both irregularities on the φ-phase grains as well as a lack of isolation or a merely discrete, spotty distribution of the Nd-rich phase, or contact with oxides diminish the coercivity. Such areas can act as strong nucleation sites and should be avoided by proper heat treatment [77]. Annealing parameters must be chosen carefully, as cracks formed at low annealing temperatures can induce even stronger local stray fields [89]. A possible treatment in conjunction with annealing is grain boundary diffusion. Grain boundary diffusion is not to be confused with the less common grain

boundary modification, where the entire grain boundary phase of the original magnet is replaced by another alloy [24]. Typical grain boundary diffusion uses a low-HRE base magnet to allow targeted deposition of HRE via diffusion along grain boundaries [24]. For this, HRE is applied to the magnet's exterior, and the magnet is heated for a prolonged period [220, 221]. During the process, liquid films form along grain boundaries, allowing the applied HRE to migrate through the grain boundaries and deposit as an asymmetric, distinct shell-like structure in the outer region of the φ -phase grain [220]. HRE like Dy and Tb enhance the local magnetocrystalline anisotropy, leading to an increase in coercivity, but also couple antiferromagnetically with iron, leading to a loss in remanence. The related mechanisms were explained more closely in section 4.1.2.2.

When grain boundary diffusion is employed for NdFeB magnets using HRE, significantly lower amounts of HRE can be used to achieve the same coercivity improvement, thereby also resulting in lower remanence loss. For example, a study found that the optimal Dy addition via GBD led to an 80 % increase in coercivity and a 6 % decrease in remanence [24]. However, the improvement in coercivity by GBD is limited to approximately 2 T when initially HRE-free magnets are used as a starting material, as the diffusion depth increases, the HRE content and thickness of the HRE-rich shell decrease [220]. Grain boundary diffusion, whether performed with HRE or other materials such as Nd-Cu, where grain boundary morphology and decoupling are supported, generally targets enhancing the coercivity [87].

4.2.5 Other methods

In Figure 4.26 the possible recycling and reuse paths are summarized. The recycling processes have different advantages and disadvantages, which Binnemans et al. [21] summarize: reuse is likely the most economical option, which can be entirely waste-free, but is only viable in specific cases [21]. It has to be addressed that reuse, along with repair and remanufacturing, should be prioritized before recycling as according to the sustainability hierarchy [222], which is also the most simple, and obvious approach to reintroduce magnets or magnet containing components into another lifecycle [21]. Component reuse is especially interesting, as around half of all failures of electric machines are attributed to the bearings breaking down, which could be replaced if the motor is otherwise still proven to perform well [208, 223]. It is expected that reuse allows the lowest energy consumption and process complexity [224]. The degradation of magnetic properties over the lifetime of a product is in most cases negligible [34]. It has been shown that thermal demagnetization is also viable for most coatings, with some needing to be re-done after the process while other coated magnets can be immediately reused [225]. Not recycling but rather reusing the magnets after possibly refreshing the coating furthermore avoids issues caused by oxidation which arises during resintering. Busch et al. [34] assume an optimistic rate of 95 % of magnets which could be reusable, if good design for reuse is applied, this includes the reuse of full magnet containing components. The requirements are similar to design for recycling, however the focus is more on designing for ease and intuitiveness of disassembly and the reversibility of joints [163], which also benefits repair and remanufacturing [11]. Reuse is not a solution for all scraps – components and magnet alloys that are outdated are likely better put into recycling [11], however, before recycling, the option of reuse should be investigated.

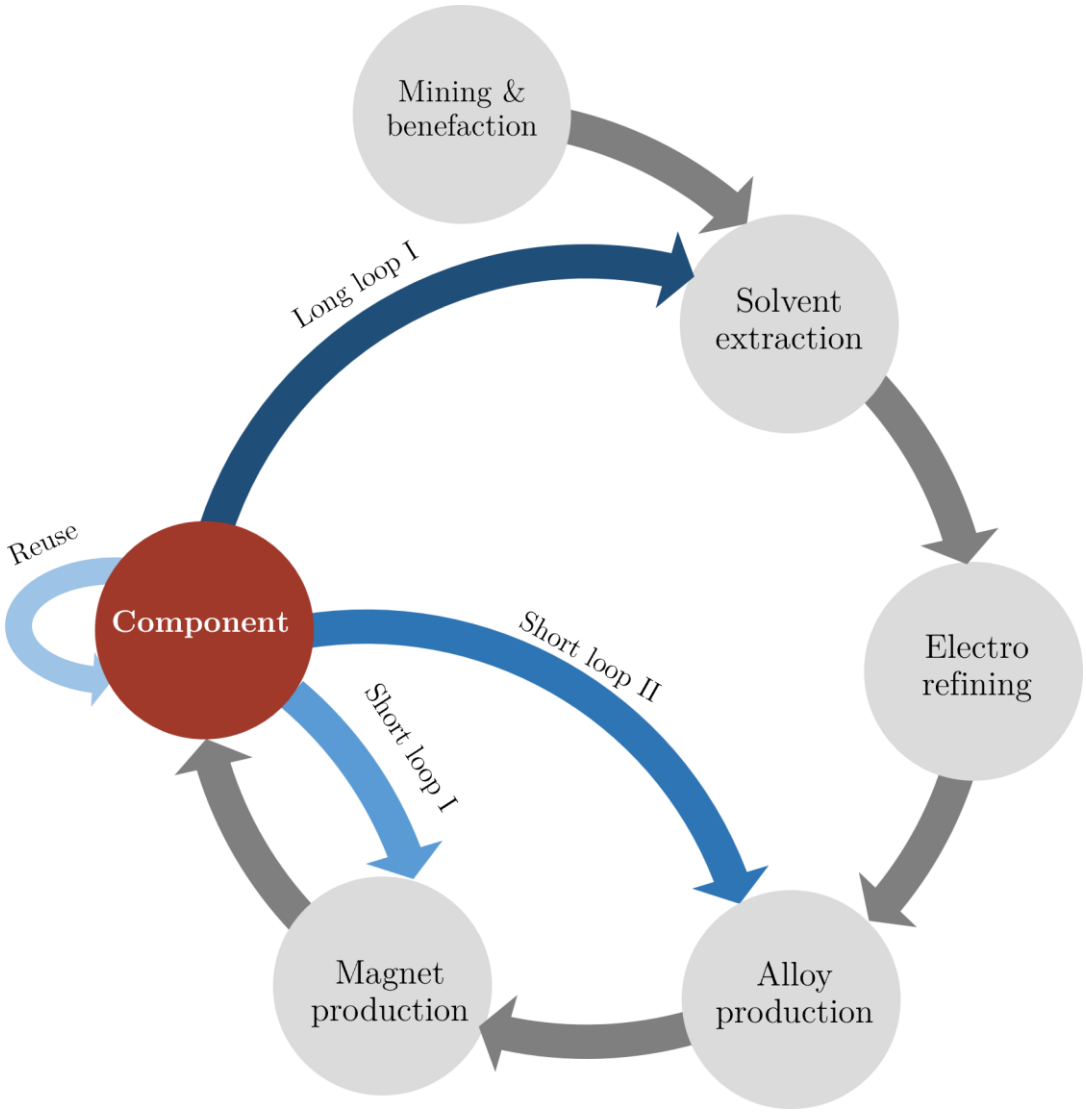


Figure 4.26: Integration of reuse, short loop I (HPMS only), short loop II (HPMS + strip casting) and long loop (hydrometallurgy, solvometallurgical, pyrometallurgy) can be integrated into a circular model. The decision of which path is most suitable should be made based on the condition of the component and integrated magnets. It may be viable to also divide intermediate recycling products into different paths.

Short-loop processing by HPMS is the next-sustainable option which among the mature, widely applied technologies offers less environmental impact [46, 226] and effort required, but trades these advantages for problems arising from contaminated, oxidized scraps [21]. Hydrometallurgical long-loop recycling, on the other hand, has effectively no relevant restrictions regarding the input material, however, many tailored steps are required which leads to large amounts of wastewater, consumption of chemicals and extensive chemical analysis [21]. Pyrometallurgical recycling offers the same non-selectivity regarding the input material with less processing steps, however the energy input required is vast and a large amount of solid waste can be generated [21].

Hydrometallurgical leaching uses different types of solvents like organic acids, inorganic acids or deep eutectic solvents, either carrying out complete or selective leaching [43]. Selective leaching is most common when recycling NdFeB and commonly, a pyrometallurgical pretreatment of some kind is conducted to improve selectivity [43].

Through leaching, the targeted elements and sometimes other elements are extracted, which also have to be separated.

Hydrometallurgical separation techniques are for example precipitation, solvent extraction, ionic liquid extraction, membrane technologies or combinations of these [43]. The use of such techniques is to extract a certain element from a mixed solution. For example, Onoda and Nakamura [227] report, that over 99 % of Nd can be extracted from a Fe-Nd solution using phosphoric acid and ascorbic acid.

While separating REEs from other elements is very viable, separating REEs from each other is a significant challenge due to their high chemical similarity. There are specialized solvent extraction methods using organophosphorus acid extractants for which the REEs show varying selectiveness [43]. Another long loop method commonly applied after hydrometallurgical treatment is electrolysis to metallize the REE oxides and separate them from each other using molten fluorides [43].

Solvometallurgy is very similar to hydrometallurgy as a process, but uses non-aqueous extractants which only have a low or no water content [228].

As mentioned, a combination of hydrometallurgical methods and pyrometallurgical methods is common. A typical first step for pyrometallurgical REE recycling is roasting, where the material is oxidized, chlorinated or sulphated at high temperatures to bring the REE into a processible, uniform chemical form [43]. REE can also be separated from iron by molten state extraction. Examples for molten state extraction methods are liquid metal extraction, which involves forming a MgREE alloy and then purifying by removing the Mg by vacuum distillation, and molten salt extraction where REE are dissolved and reacted with fluorides, followed by hydrometer extraction [43, 229].

Recently developed methods for bioleaching using microorganisms like *Acidithiobacillus* or *Leptospirillum ferrooxidans* offer more resource efficient, selective recycling at the cost of significantly longer processing times [230, 231].

The different recycling processes can furthermore feed into each other to some degree. Also, material deemed to be too contaminated for proper HPMS processing can be fed into long-loop processes to regain the material [21].

4.3 Electric Machines

"Electric machines" is a collective term for any type of machine that transforms electric energy into mechanical energy or vice versa. Classically, this includes electric motors, generators, and combinations of both.

Rotational electric machines are most common, and they are applied for inducing movement, for propelling movable media like liquids and gases in the form of pumps and ventilators – or in terms of generators, to do the opposite.

Consequently, they are built into a large variety of products. It is estimated that (electric) motors are responsible for around 40 % of the world's electricity consumption while contributing around 13 % of global carbon emissions [223]. On the other hand, most energy production systems use generators – this includes renewable energy systems like wind turbines and hydroelectric power stations as well as fossil fuel energy generation and nuclear power, which all rely on turbines.

Electric machines which hold NdFeB permanent magnets usually are either constrained in terms of mass and volume or by efficiency. High efficiency is especially important in battery-driven systems, but also in energy generation among others.

In this chapter, electric machines are systematized and explained from a perspective which is strongly focused on the recycling of NdFeB and thus, the rotor component.

4.3.1 Function and types of electric machines

In an electric motor, electric energy is used to generate targeted electromagnetic forces which in turn, generate rotating or linear movement. If, in turn, electric energy is generated through movement of electromagnetic forces, the electric machine is referred to as a generator, or more precisely a dynamo when direct current (DC) is produced or an alternator, when alternating current (AC) is produced. Most electric machines, while being specialized and optimized for one option only, can function as a motor or generator; however, the same flexibility is usually not given between the electrical systems available, AC or DC.

In AC machines, the stator generates a rotating magnetic field by applying single phase or three phase AC. The field of the rotor moves due to the rotation. If both rotate at the same speed, the machine is considered synchronous, if the rotor field is faster or slower, it is considered asynchronous.

For DC machines, there is no division between these types. DC machines use alternating voltages and currents and a commutator, so technically, the fields do not rotate in this case.

Brushes are used to transfer electricity to the rotor in some designs, specifically the brush is energized by the stator and transfers the energy to the rotor's slip ring. Not all electric machines use brushes, induction is also an option for electricity transfer – alternatively a rotor's magnetic field can be generated by permanent magnets which do not require energy supply.

Different types of electric machines and whether they use permanent magnet rotors are shown in Figure 4.27.

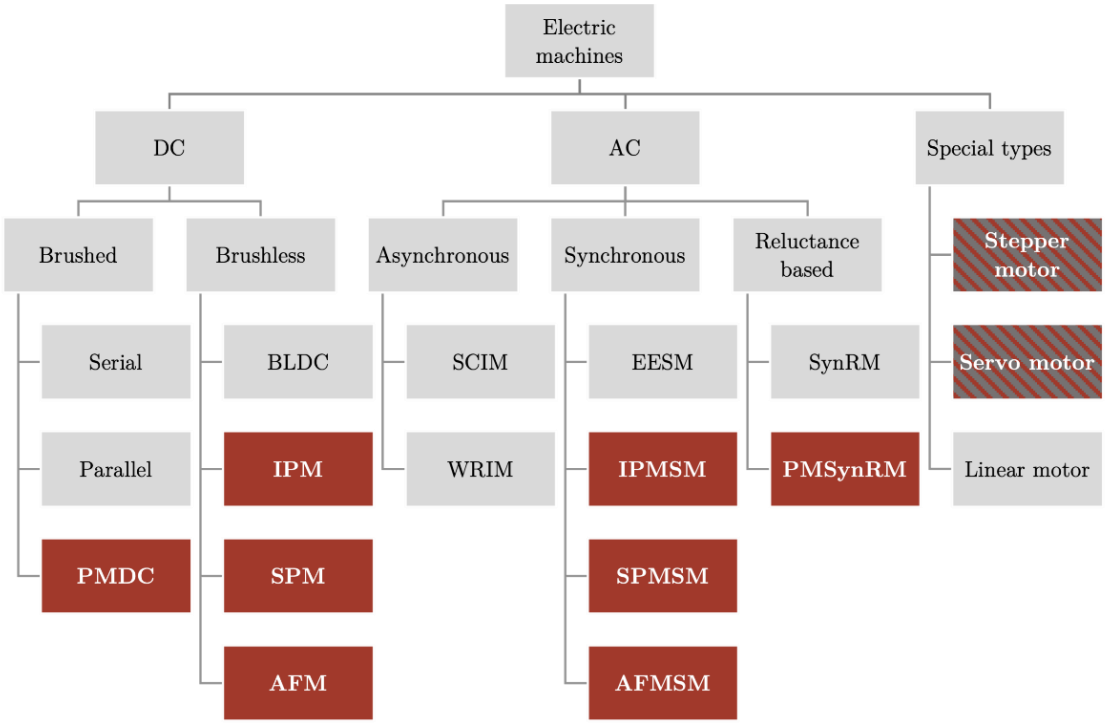


Figure 4.27: Types of electric machines, the ones that contain permanent magnets are marked in red.[1]

Overall, electric machines come in vastly different sizes, ranging from miniature motors used in entertainment and personal care electronics, for example, to generators of wind turbines. Due to this, the performance characteristics can be very varied. A selection of important characteristics is shown in Table 4.1.

One requirement that is purposefully missing in this list is the recyclability, which will be addressed in detail in the following section 4.4. At this point, it is important to note that none of the requirements in Table 4.1 should be neglected in favor of the recyclability.

Depending on the application of the electric machine, the weight of the requirements can differ, for example for electric traction motors, Zeraoulia et al. [152] name efficiency, reliability, cost, driver expectations, vehicle constraints and energy source as the main requirements.

[1] To translate the acronyms without having to go back to the glossary, here a few explanations: AC refers to "alternating current", AFM to „axial flux machine", BL refers to „brushless", DC refers to „direct current", EESM for „externally excited synchronous machine" IPM refers to „internal permanent magnet", PM to „permanent magnet", RM refers to „reluctance motor", SCIM to „squirrel cage induction motor", SM to „synchronous motor", SPM to „surface permanent magnet", Syn to „synchronous", WRIM to „wound rotor induction motor".

Table 4.1: General motor requirements. Non-exhaustive list, which does not include specific requirements for different applications.

Requirement	Meaning
Efficiency	• Output power/ Input power. • Determines energy conversion and losses.
Power density	• Output power per unit mass or volume. • Reflects compactness.
Torque-Speed Characteristic	• Relationship between torque and rotational speed. • Determines operating behavior.
Speed range, control capacity and dynamic response	• Determines operational speeds and opportunities to control it. • Speed and precision of reactions to control inputs and load changes.
Thermal performance	• Possibilities to deal with and dissipate heat. • Determines operating limits.
Noise and vibrational performance	• Includes factors like e.g., torque ripple and cogging torque or noise induced by rotor imbalance. • Influences comfort, mechanical integrity, function and compliance with acoustic standards.
Robustness, reliability, monitoring and repairability	• Determines lifetime and servicing. • Can also include predictive maintenance.

4.3.2 Permanent magnet electric machines

As shown in the previous section, a number of different electric machine designs use permanent magnets. The permanent magnets are usually applied in the rotor to generate a magnetic field without relying on an electric supply. Rarely, magnets are also or exclusively mounted in the stator. However, the geometrical relationship of the rotor and stator can vary as well as the integration of the magnets in the rotor.

Permanent magnet machines offer a wide speed range, enhanced efficiency and high reliability compared to other options [232, 233]. When comparing permanent magnet synchronous motors (PMSM) to permanent magnet-free options, they offer close to 30 % more torque and power density than a brushless direct current motor (BLDC), and close to 90 % more than an synchronous reluctance motor (SynRM) and 200 % more than an induction motor (IM, includes e.g. SCIM or WRIM) under the same operating conditions [234]. In traction applications, using permanent magnet machines like PM BLDC or PMSM (including PMSynRM) is most common [152, 234–236].

Surface mounted permanent magnets (SPM) are attached to the rotor surface and sometimes covered with a bandage or a fixation feature. Internal permanent magnets (IPM) are affixed in pockets within the rotor stack material. The rotor can furthermore be placed inside the stator (inner rotor, IR), which is most common, or around the stator (outer rotor, OR). Alternatively, the designs are not limited to radial flux arrangements like mentioned before, as axial flux machines (AFM) are also possible. In the following, the electric machine types, or rather their rotors, are classified and explained based on the arrangement of rotor, stator and permanent magnets. A visual overview is shown in Figure 4.28.

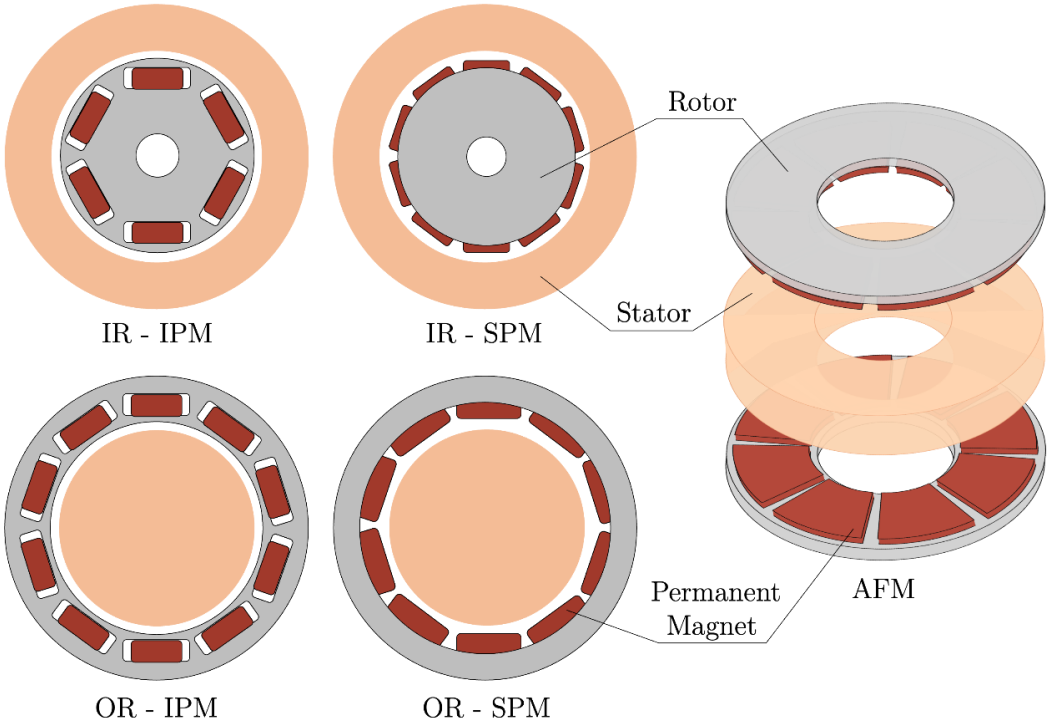


Figure 4.28: Permanent magnet rotors classified by the relative position of rotor and stator and the mounting of the magnets. There are different arrangements for AFM, just one example is shown.

Generally, a rotor with permanent magnets usually consists of one or multiple stacks of electric steel sheets, which can be stacked by different joining methods like interlocking, backlack, dotted glue, lasering, or other means [237, 238].

The stacks are then mounted on a solid or hollow shaft, which can be done in parallel or with a skew. Skewing is a prevalent method to suppress torque ripple and ensure the motor is running smoothly with minimal noise [16]. Anything between a no skew and a multi-level skew is possible, also using different skew angles either in a line or in a V-shape. However, the skew angle should be optimized between the reduction of torque ripple and conserving the output torque and back electromotive force [239].

4.3.2.1 Internal permanent magnet rotors

IPM are the most common motor topology for traction motors due to the high power density and efficiency offered by this type of rotor [146]. Generally, when the main requirements are high power density, wide speed range, and efficiency, IPM are most capable of meeting the requirements [240].

In IPMs, the magnets are placed in magnet pockets and either held by force, adhesive fixation, in-molding, or mechanical features [153, 241, 242]. The layout of the magnets can greatly vary, as they can be placed at an angle. It is furthermore common to layer the magnets tangentially or in a “fir tree” shape. Some examples of magnet layouts are shown in Figure 4.29. The magnet pockets are usually extended into a so-called flux barrier, which serves to direct the flow of the magnetic field. For example, in traction applications, it appears that all rotors use flux barriers [16]. Some especially large flux barriers can be seen in PMSynRM to increase the salience and the reluctance moment [243, 244].

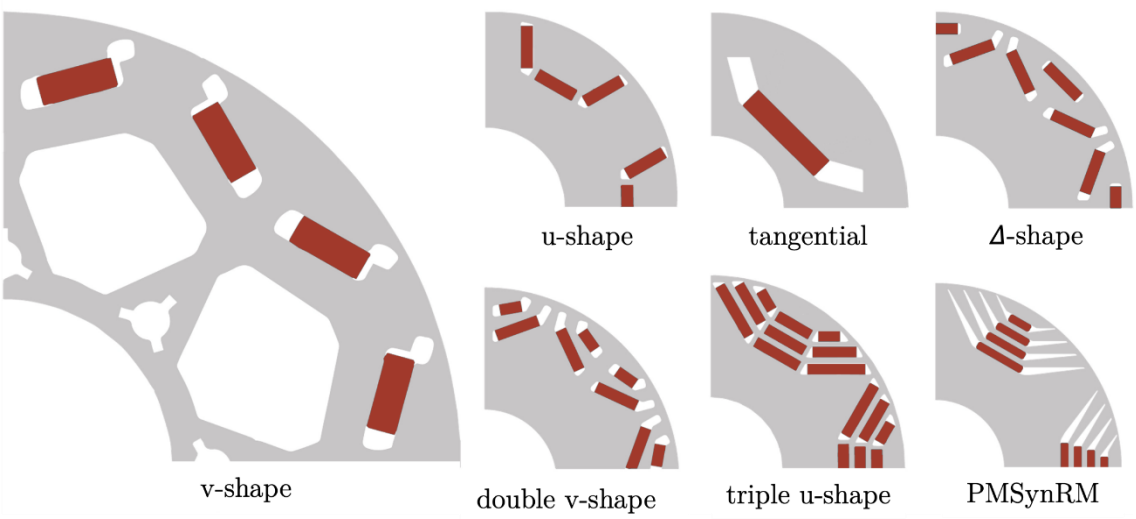


Figure 4.29: Typical IPM rotor topologies.

4.3.2.2 Surface-mounted permanent magnet rotors

Industrial motors, in comparison to traction motors, are usually exposed to significantly less harsh environmental conditions, shocks, vibration, operating temperature cycles, and service life [16]. Furthermore, less extreme rotational speeds are common, allowing a less robust integration of the magnets.

The reduced requirements regarding the robustness of the machine allow a higher freedom of design, which opens the process for more considerations regarding the recyclability of the machine. For instance, a rotor type which can be demounted easily can be used – like a surface mounted permanent magnet synchronous motor (SPMSM).

SPMSM are very commonly used in industrial motors as well as smaller motor designs and show a variety of possible topologies, for example SPMSM with an inner rotor or an outer rotor, which offer different benefits.

In SPM, the magnets are affixed to the rotor surface, either just using an adhesive or also a bandage. Bandages can be organic, like fiber-reinforced tape or metallic, like a shrink bandage made from aluminum. In inset rotors, the magnets are held mechanically, therefore they could be classified as something between IPM and SPM [245]. Magnet fixation is especially simple if the magnets are not exposed to centrifugal forces which point away from their glued-on side. This is especially true in outer rotor SPM (OR-SPM) or in AFM, where the magnets are axially placed on a rotor disk.

4.3.3 NdFeB in electric machines

NdFeB are a highly popular choice of permanent magnet in electric motors due to having the highest energy density. This allows manufacturing motors with the highest power to volume and mass ratio and very high efficiency. However, a specific downside of NdFeB should be addressed as well:

The main issue of the application of NdFeB faces in the application of electric machines is the high sensitivity of the magnetic properties to temperature. Compared to other magnet types used on a commercial scale, NdFeB has the lowest Curie temperature. An example of temperature-related decrease in magnetic properties is shown in the following Figure 4.30:

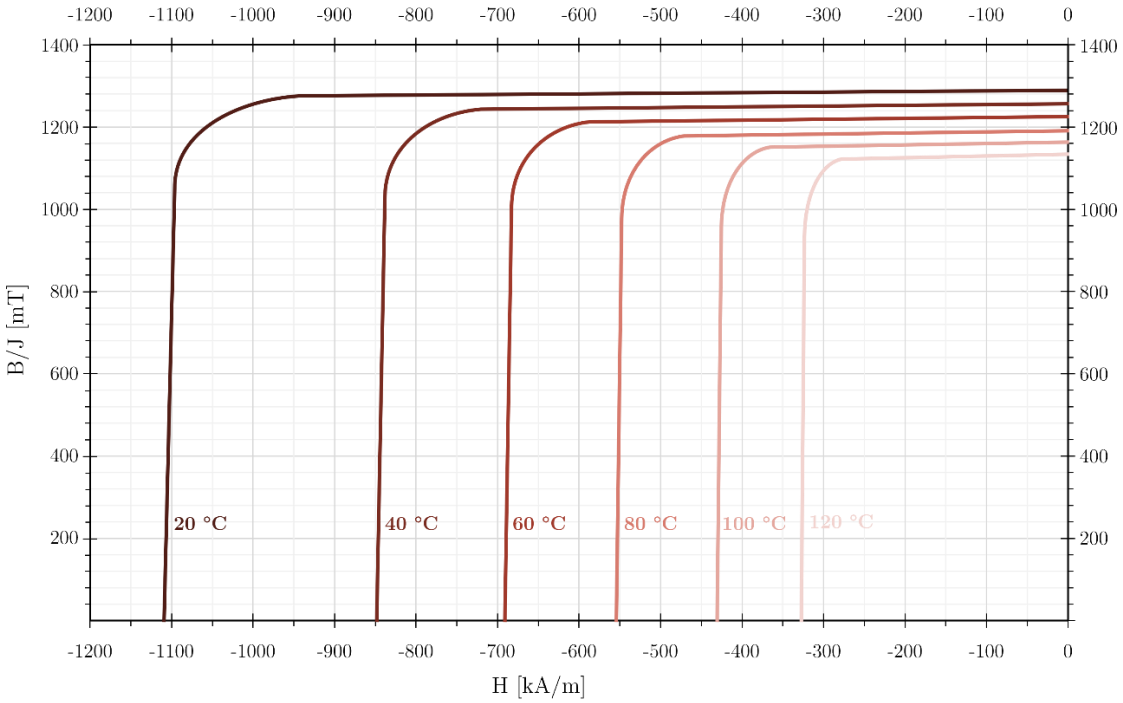


Figure 4.30: Decrease of H_{cJ} and B_r linked to the usage temperature.

Eddy currents, s. Figure 4.31, are circulating electrical currents which can be induced in a conductive bulk material like an NdFeB magnet due to Faraday's law of induction. In simplified terms, they generate an electromotive force opposing the original magnetic field within the material, leading to energy losses due to Joule heating [246]. The overall losses by eddy currents can be suppressed by blocking the current paths and limiting the volume in which the currents can be generated [246]. The reverse electromotive force decreases proportionally with decreasing the loop area, which corresponds to the size of the conductive volume [246].

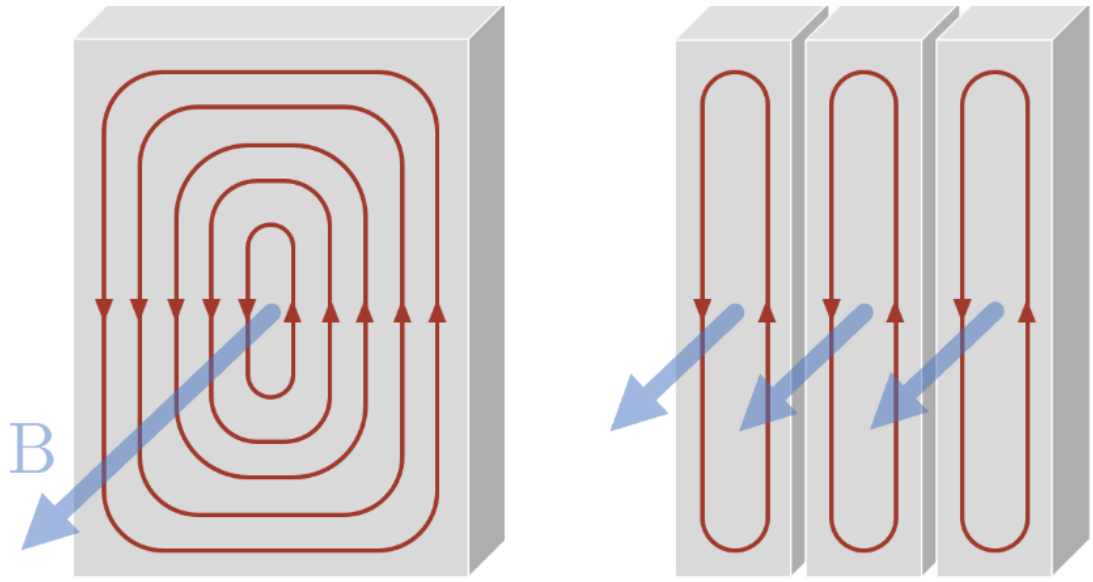


Figure 4.31: Eddy current generation related to the conductive volume. Left is an unsegmented magnet, right a segmented magnet with isolative material like adhesive dividing the magnet bulk.

Eddy current losses within the magnet represent a high amount of the total losses in high speed machines [247, 248]. Eddy currents increase with the square of the frequency, whereas high speed machines like traction motors are especially affected [16]. The induced heating of the magnets further causes demagnetization [247], meaning that machines that run at higher speeds of around 20,000 rpm will require sufficient temperature stability or suitable means to reduce eddy currents. Common solutions to this are using magnets with a high heavy rare earth content and segmenting the magnets. However, a trend to reduce lamination thickness of the rotor can also be observed, which reduces the eddy current generation as well [16].

4.4 Design for Recycling

4.4.1 Circular economy and the sustainability hierarchy

The circular economy can be defined as an industrial economy that is restorative by intention and design [223]. Sustainable development is defined as that it fulfills the need of the current generation without impairing the capability of future generations to fulfill their own needs [249].

A primary characteristic of circular development is to view scrap material as a valuable resource instead of waste [134]. This perspective has largely shaped research, infrastructural development, culture, and even linguistics within the society:

For example, in German, the words "Wertstoffsammlung" (collection of recyclables, literally translated as "valuable material collection") or "Wertstoffhof" (scrap yard, literally translated as "valuable material yard") are highly established in colloquial parlance. This indicates that end of life materials have gained widespread recognition as valuable raw materials.

The principles of circular economy recognize the value of raw materials and the associated environmental impact of primary production, aiming to keep these materials, especially finite and highly processed ones, in a loop for as long as possible.

It is important to recognize that recycling is not a primary objective, but rather a last resort before landfilling and the ultimate loss of the material. The efficient use of raw materials, the design of products with a long lifetime, under the avoidance of waste, and emissions are to be prioritized before recycling [250]. This is illustrated in the waste hierarchy depicted in Figure 4.32.

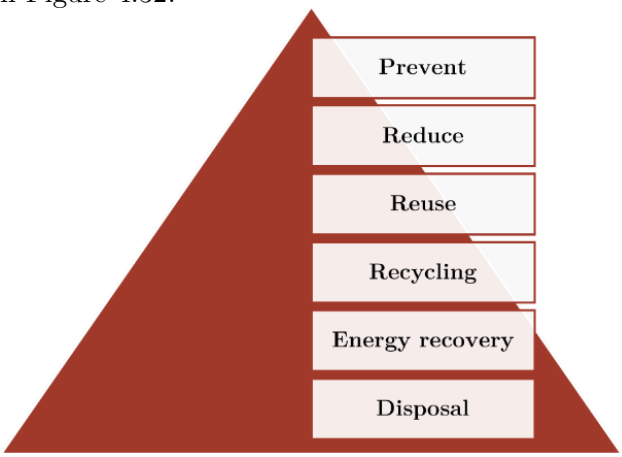


Figure 4.32: Waste hierarchy, based on [251]. Preferential treatment of end of life products and managing retention of raw materials and products within the technosphere.

However, that does not diminish the need for recyclable products at all, especially since even the most durable products will eventually no longer meet the current requirements of consumers or may be replaced by more efficient and eco-friendly alternatives at some point.

Accordingly, the goal of longevity must be considered in a differentiated manner at the product level, and it must be evaluated how sensible reuse applications are.

Design for a circular economy, therefore, aims to design products that promote a more sustainable consumption pattern, with low emissions during production, and enhanced recyclability.

While this work is essentially focused on design for recycling, any measures towards better recyclability must also be considered from the perspective of overall sustainability and eco-design. An overview about eco-design requirements is given in Figure 4.33.

Reducing designs to their essentials and minimizing waste by creating durable products should be the first step. For design for recycling, this means that measures that improve recyclability but diminish the product's durability, causing it to fail before its useful life cycle length, are not sustainable and should be avoided.

Furthermore, the addition of extra features and production steps should be avoided if no other features can be reduced or replaced with more suitable ones.

Production waste should be minimized by optimizing material usage. Parts that serve no essential function should also be reduced, as they may complicate recyclability.



Figure 4.33: Eco-design requirements as according to the Ecodesign for Sustainable Products Regulation Article 5 [252].

Remanufacturing, repurposing, repairing, and replacing of products should be prioritized before recycling as well [223, 251]. This may refer to the product, where certain components are replaced or to component recycling. Functioning, reusable parts are removed from a used product, refurbished, and assembled into a new product. For motors, this could mean that the rotor is reintegrated into a new motor or that the magnets are removed, cleaned, de-coated, and recoated, and then integrated into a new design. This process includes quality assurance and possible improvements or modifications to the components [23].

Non-destructive disassembly is crucial for recycling at the product or component level and depends heavily on the design, particularly in terms of manufacturing methods [160, 253].

Accessibility of parts to replace or repair them usually goes hand in hand with good disassembly and, thus, recyclability [13]. For remanufacturing and repurposing, knowledge of the product and its wear and aging, as well as excellent protection of critical parts and materials such as permanent magnets, is key. For example, a digital twin is a modern tool used to predict the properties of components at their end of life [254].

Another important factor is the variability of designs, as more standardized parts mean more accessible disassembly [11]. For repurposing and remanufacturing, modular and platform-based designs can be helpful solutions.

A minimum requirement for any circularity-friendly product is a suitable supply of information about the product that allows recyclers to know what to expect from the end of life product in terms of materials and hazards, as well as how to process and test it. This is especially true for the last measure to extend the usefulness of a material: recycling.

4.4.2 General design for recycling approaches

The final step in the circular economy is the recovery of materials by "classical" recycling. For this purpose, the product is broken down into its individual parts and consequently treated at the material stage. Here, the least effortful and the least environmentally straining method that produces material of sufficient quality should be preferred. At the current state of the art and industrial development, for NdFeB, this would be short loop recycling via hydrogen decrepitation [46, 226]. However, other methods may emerge as even more resource-efficient in the future, as already discussed in section 4.2.5.

To obtain sufficiently pure material, it is of utmost importance to be able to separate components, coatings, and adhesives that would decrease the quality of the most desired materials or to avoid these altogether.

Some Components and materials that are not returned to the production process can be recovered via energy recovery, such as incineration. Energy recovery and landfilling of waste must be minimized, as these processes are not based on the idea of recycling-friendly product design and do not advance the efficient use of resources associated with the circular economy.

There are different definitions of design for recycling, many being specific to a product or component, like packaging. Therefore, design for recycling should be defined as it is used in the scope of this work.

To design for recycling, the goal is to facilitate the leap from one lifecycle into the next: It is not limited to product design, as process design is just as important [11], nor is it limited to the operation of recycling itself.

The importance of process design exceeds that of product design in case of NdFeB in electric machines as of now, as electric machines are highly technologically developed and highly optimized, while NdFeB recycling is still under development. The leeway for changes in machine designs may be high in theory but would potentially sabotage the fine-tuned developments of motor designers and lower the performance, efficiency, or durability of the electric machines with larger changes. Therefore, a compromise is sought by approaching the problem from both ends and focusing on the processability and compatibility of machine design features with different treatment techniques.

Another factor considered in the design for recycling approach applied here is the influence of the overall infrastructure, exceeding the recycling process itself. Therefore, policy design, societal norms and education also play a role. This leads to the following definition:



"Design for recycling comprises all methods of altering the to-be-recycled products, recycling processes, and recycling infrastructure, taking into account the adjacent technoeconomic and social environment to ensure high recycling rates and high-quality material."

However, most available literature usually has both a product-centric approach and is also only focused on the product design. This, however, is also very useful for the case of the recyclability of NdFeB from electric machines. Therefore, general rules, as shown in Table 4.2 and approaches for (product) design for recycling are addressed in the following.

The THEMA matrix, as mentioned in Table 4.2, was developed by Castro [255] and shows what materials must be separated, should be separated or should be kept together. It includes materials like cast and wrought Al, alloys of Cu, Pb, Mg, Pt and Zn, different types of steel as well as different polymer classes, natural materials, glass and ceramics [255]. It describes how minor additions of each element will affect the recycling streams of other material when they are accidentally added to them [255].

In terms of disassembly, which is important for most design for sustainability approaches, considerations of disassembly force, material handling, tool requirements and accessibility, as already partly addressed in Table 4.2, are addressed by Desai and Mital [161], who developed a scoreboard for disassemblability. The scoreboard gives different points for the ease of disassembly including component size and weight, position and types of fasteners and the type and force required for disassembly, culminating in an evaluation of the ergonomy of the disassembly [161]. Soh et al. [13] add the graspability of parts to this consideration.

Table 4.2: General design for recycling recommendations.

Category	Rule/Recommendation	References
Joints	• Choose joints with a short disassembly time. • Choose joints with good reversibility and low damage. • Use standard joints which can be disassembled with common tools. • Avoid adhesives.	[11, 163]
Materials	• Limit the number of materials. • Use common, standard materials. • Choose recyclable materials. • Choose materials that do not age significantly, can be protected or restored well. • Avoid composite materials. • Avoid adhesive components like stickers, labels, tape. • Combine materials in a way that they can be separated and avoid putting such materials together, which are incompatible according to the THEMA matrix. • Avoid surface treatments. • Insulation material should be easily separable from metals. • Avoid hazardous materials and ensure that if they are necessary, they can be found easily and removed safely.	[10–12, 224, 255]
Components	• Use standard parts. • Avoid dissipation of valuable materials over different components and within components, if possible. • Use as few components as possible.	[138, 253, 256]
General	• Available recycling technologies must be minded. • Overall economics of scale must be minded as they decide whether recycling is economically viable. • Sociological factors should be considered as the success of the recycling depends on the behavior of actors in the recycling chain. • Consider the demand of secondary materials when choosing which materials to prioritize for recycling.	[138]

It has to be noted, that disassembly is regarded as a non-value-added process, which can take excessive amounts of manual labor if a design is not suited for disassembly. This implies that clear incentives must be given – both to design for disassembly and to actually disassemble products at the end of life. Such incentives can be of economic nature, due to scarcity of materials or by legal means. Ideally, a combination of these. Disassembly planning and time calculation has already been addressed in section 4.2.1 where the pre-processing of NdFeB components is discussed.

Regarding the process of implementing design for recycling into product design, systematic approaches like those shown in Figure 4.34, based on Chen et al. [19] can be applied. The approach by Chen et al. is an especially realistic method which factors in cost and benefits both in terms of their benefit and detriment to society and environment, but also to actual costs and economic viability. Here, the design is analyzed regarding the fulfillment of the functions, the material recyclability and the compatibility of materials and joints. The design is then iteratively refined until it is theoretically recyclable, followed

by a cost-benefit analysis. When the results of the cost-benefit analysis are also acceptable, the design can be finalized.

The costs of recycling are also investigated by Desai and Mital [161], who summarize that there are disassembly costs, cleaning costs and recycling costs which comprise costs for material processing, manufacturing the recycled materials, packaging costs and assembly costs. What also comes into play for the overall cost calculation of recycled material at this point is how the recyclable material is procured: Is it returned via a closed loop or collection scheme, or do manufacturers buy the scraps?

Villalba et al. [257] describe the devaluation by dividing the cost for scrap materials by the cost for primary materials. In case of a high devaluation, higher recycling costs are more likely to be economically viable.

It is clear that while design for recycling should not impair the functionalities of the product, it must also be economical to be applied properly [19, 161]. The success of design for recycling measures can be potentially calculated, for example as suggested by Zeng and Li [258] based on entropy, complexity, quantity, and quality of the materials.

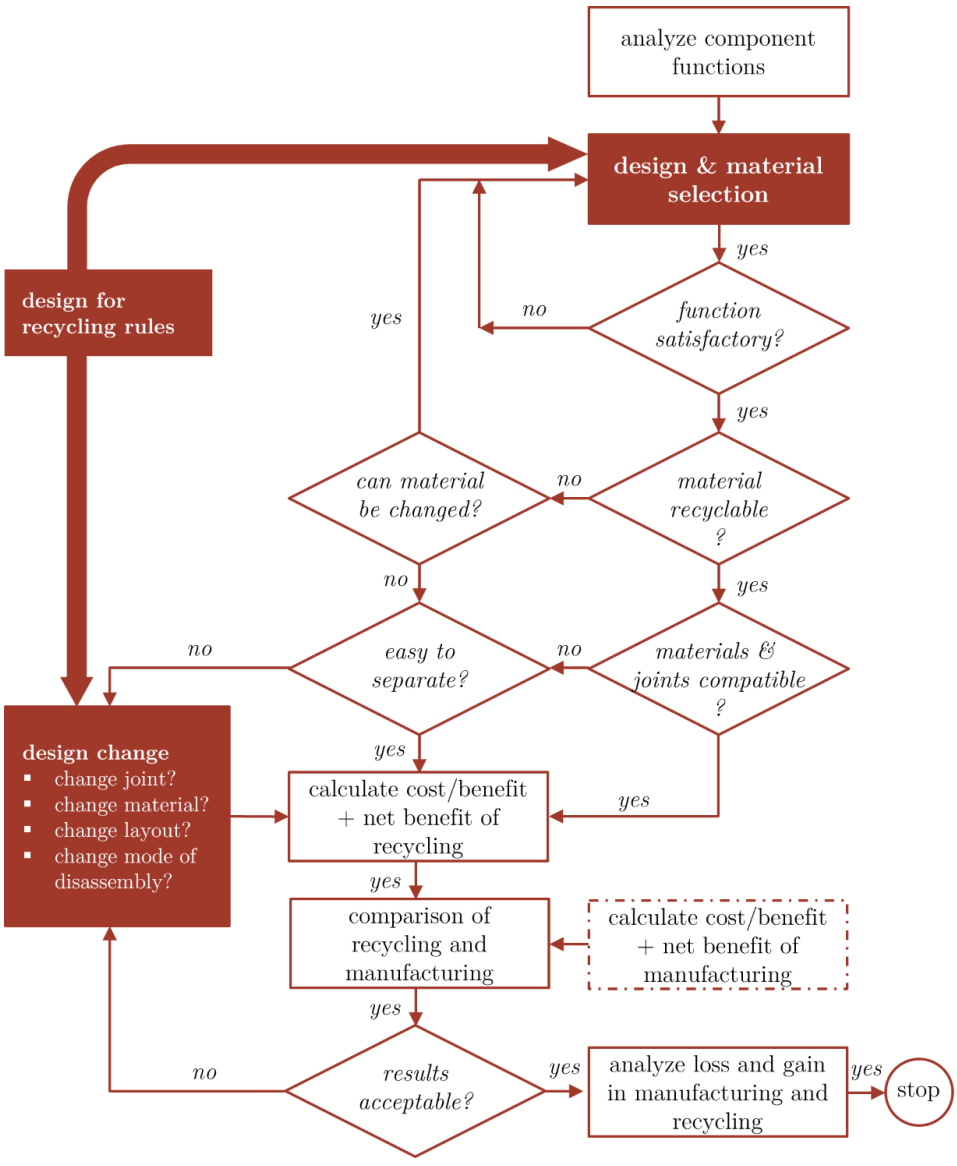


Figure 4.34: Process to implement design for recycling into product design, minding the costs and benefits of doing so. Reproduced and adapted from Chen et al. [19].

4.4.3 Design for recycling considerations regarding electric machines and permanent magnets

When looking into the design for recycling of NdFeB from electric machines specifically, it is important to consider what sets it apart from other products to direct the research effort to what is necessary to explore instead of replicating what is already known.

The first factor is clearly the extremely strong ferromagnetism of NdFeB, which makes disassembly and shredding in a magnetized state challenging and dangerous.

The second factor is the sensitivity of the material towards contamination: Nd is highly reactive, and the magnetic coupling system is easily disturbed by materials which impair the anisotropy of the elementary cell. The de-coupling between the hard magnetic grains is essential and relies on the Nd being liquid during sintering, whereas any organic binding of Nd is having negative effects on the densification and coercivity development of the material.

Third, while this is not an inherent problem of recycling NdFeB-containing electric machines, this work adopts a largely material-centric approach to recyclability, which is why NdFeB, as a material, is central to the topic.

Lastly, an electric machine is a system where several materials that are highly incompatible in recycling are very closely combined and durably connected.

The following compiles the current literature on this specific problem to identify knowledge gaps.

4.4.3.1 D4R in machines and electronic products

When recycling electric machines, it is essential to fully separate copper and steel, both of which are primary materials in most electric machines. The acceptable copper content in steel is below 0.02 % [166]. Windings tend to be made from Cu or Al [12]. Alatalo et al. [12] recommend using a claw pole machine with a stator made from soft magnetic composites (SMCs) and Al windings for better recyclability, as SMC cores can be easily crushed and the Al content is far less critical than Cu in steel [12]. The silicon steel used in electric steel sheets is not well-suited for recycling, as silicon is a contaminant in the production of ferromagnetic steel and can only be added in small amounts to steel recycling batches [224]. Avoiding permanent magnets can be achieved by using induction motors, for example [232, 240].

4.4.3.2 Research and inventions regarding D4R to recycle permanent magnets from electric machines: Product design

Generally, the technical requirements towards electric machines are often very narrow, both in performance and durability, as well as in size and cost. This especially applies to traction drives, where the motor is most often designed to fit all the other mechanical equipment, narrowing the window of possible design features even more and rendering actual design for recycling to be very challenging to implement [12]. However, there are some approaches to improve the design for recycling for NdFeB-containing electric machines. In order to find available methods to improve design for recycling, aside from literature research, a systematic patent research has been conducted. Documents within the classes H02K1/27, H02K1/28 and H02K1/30 were combined with the terms “permanent magnet” and “rotor” and additionally the non-mandatory terms “hold”, “recover”, “disassembly”/”disassemble”, “recycle”/”recycling”, and “demount”. In total, 2164 documents were screened, and 208 of those documents were analyzed in detail. While it is unclear for many patents whether the motivation was to improve recyclability or not, it is worthwhile mentioning all options that may positively contribute to recyclability. The

most interesting solutions are compiled shortly in the following, including patents and patent applications as well as published research.

A number of design for recycling approaches rely on concepts which are entirely diverting from "regular" electric machine design, where sintered magnets are mounted in or on an electric steel sheet stack. For example, some authors propose designs which stray away from the typical rotor manufacturing and propose using non-traditional materials like soft magnetic composites instead of steel sheet stacks, like Lundmark and Alatalo [259], Henaux et al. [260], Zhang [261] and Garcia Gonzalez [262].

The main advantage of soft magnetic composites for recycling is that they can be crushed easily [260]. Another concept going down an uncommon, but nonetheless interesting path is the "IP magnet" which is named after the protectivity class IP68 [183]. This concept essentially avoids glue, coating and any shaping process for the production of the magnet by inserting magnet powder into a stainless steel sleeve, pressed and then hermetically sealed by welding the end caps [183]. Later on, the powder can be extracted by thermal demagnetization and crushing after opening the sleeve and releasing the powder [183].

Aside from using different materials than usual, a number of methods also propose alternative rotor topologies.

Some examples are shown in Figure 4.35, where concepts, which were published as patent applications or patents, are summarized. Please note that this list has no intention of completeness, but rather compiles the most interesting concepts to be applied within the most common rotor topologies. The use of entirely different types of motors like axial flux machines, which are inherently more recycling friendly, are excluded from this analysis. Instead, the focus is entirely on improving radial flux permanent magnet machines like they are most commonly used.

One strategy is to use segmented rotor designs to promote recyclability [263–266]. While not all segmented designs help to disassemble the magnet, they usually allow that the rotor is broken down into smaller parts without deforming the magnet pockets, potentially allowing a higher fill of the hydrogen reactors. The concept by Brandau and Siebald [264] uses a 2-piece steel sheet stack: One, inner part on which the magnets are mounted and an outer part which holds them in place similar to a sleeve in SPM, potentially largely facilitating the demounting (s. 4.35 O) [264].

Another approach is to replace "regular" magnet fixation methods like adhesive fixation, or more rarely, using a press fit by smarter features. As several authors [49, 183, 208] state, polymers can impair recyclability, especially the widely used epoxy resins are mentioned as problematic, as they contaminate the powder [49] and are not recyclable themselves [208]. Due to this, it is proposed to replace them with more recycling friendly variants [49, 208]. An approach to do so was published by Podmiljšak et al. [267]. Makihara et al. [268] propose a new resin composition for magnet fixation which is meant to be disassembled easily by dissolving.

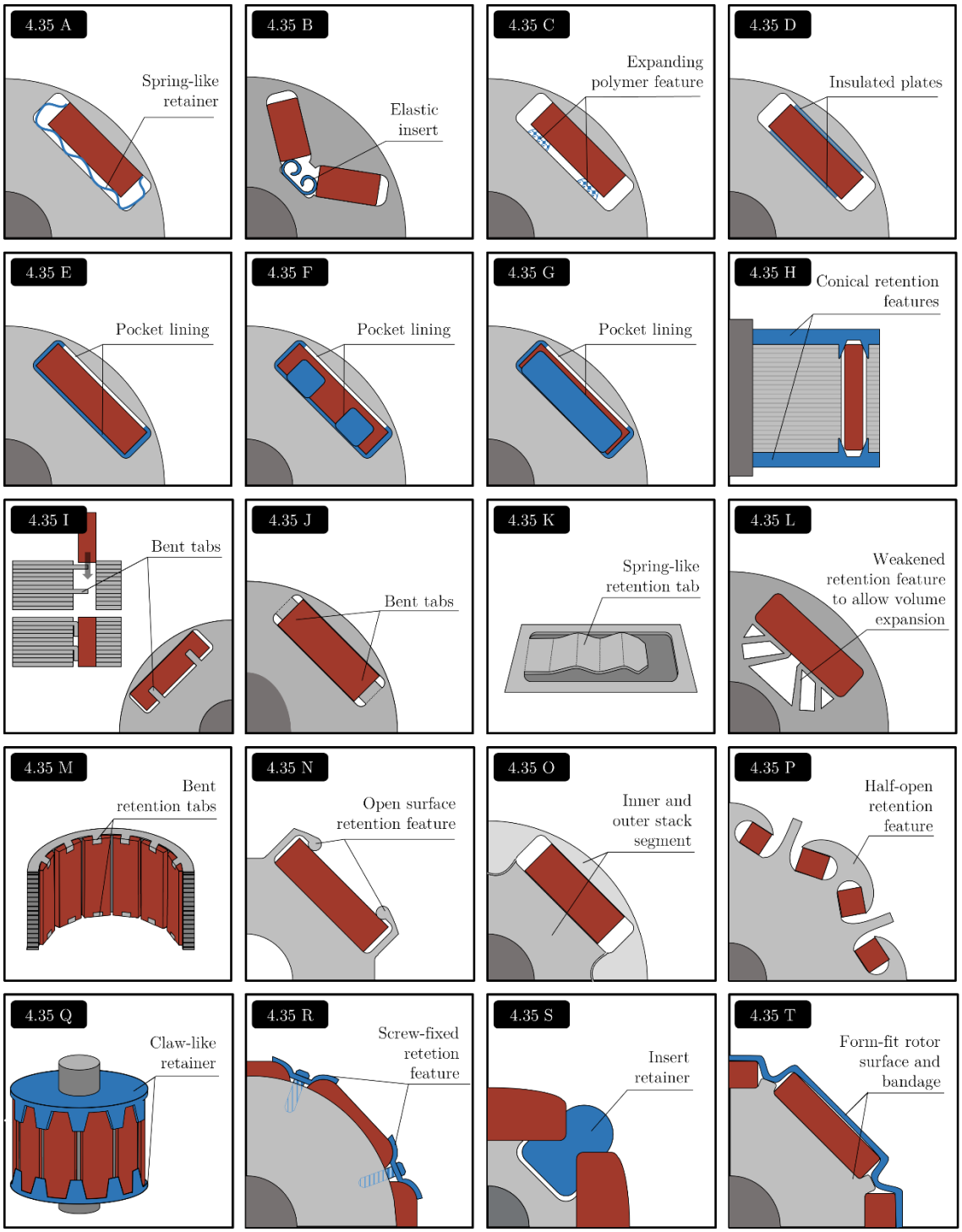


Figure 4.35: Different features found in patent research which may be useful to improve magnet recyclability. Not all features were developed in a targeted way to promote recyclability, however, they have the potential to improve magnet removal and hydrogen processing in situ, depending on the arrangement. The panels are referred to in the following text, where the inventions are described and the corresponding sources are mentioned. Panel A is based on [269], B on [270], C on [271], D on [272] E to G on [273], H on [274], I on [241], J on [275], K on [276] L on [277], M on [278], N on [279], O on [264], P on [280], Q on [281], R on [282], S on [283], and T on [284].

Regarding the fixation of magnets, different patents were found, providing mechanical fixation by innovative features which may facilitate demounting of NdFeB magnets:

For example, concepts where permanent magnets are held on the surface of a rotor using a mechanical feature, which may avoid using adhesive and simplify demounting [282, 285, 286]. The concept of Nehring et al. [282] is highly compatible with HPMS if a passivated magnet and no adhesive is used, as the retaining feature does not fully cover the magnet and is furthermore suited for non-destructive disassembly due to using screws (s. 4.35 R). A similar concept using form fitted features instead of screwed on plates is shown by Li [287].

For outer rotor surface permanent magnet machines, where magnet mounting is less demanding, a very simple but useful concept is proposed by Zhang et al. [278], where tabs are folded to hold the magnets without requiring adhesive (s. 4.35 M). A similar concept is used by Foucault [281] for surface mounted magnets, however with a sturdier claw-like feature (s. 4.35 Q).

Monzie and Straub [284] propose a method which holds the magnets between the stack and the sleeve, where both surfaces adjacent to the magnet are molded, whereas the use of adhesive is unlikely required due to the given form fit (s. 4.35 T).

Tucker [279] uses a prong-like cutout of the stacks to hold the magnets while also aiding assembly – the concept thus benefits both assembly, disassembly and suitability for direct HPMS treatment without demounting (s. 4.35 N).

Similar concepts exist for fixing permanent magnets within a magnet pocket, like leaving a small tab of material within the magnet pocket in some steel sheets which then fold over and hold the magnets in place, as presented by Kim [241] (s. 4.35 I). This feature is a brilliant way to create an easy to mount and demount press fit which also offers some empty space which can be used for expansion during HPMS [241]. It has been replicated by several other patent applications in a slightly modified way. Takashi [275] also uses bent tabs to fix the magnets, but from the sides (s. 4.35 J).

Several authors use spring-like features, holding the magnets in place (s. 4.35 A) [269, 288–290]. A generic spring-like feature, most akin to the versions by Paul and Stein [269] is shown in 4.35 A. Yue et al. [270] use a modified version, where the spring-like feature is in the flux barriers and one feature holds two magnets in place at the same time (s. 4.35 B). Jin et al. [276] combine the approach of using a foldable tab and a spring-like retainer by bending the tab into a spring-like structure (s. 4.35 K).

Kim [274] shows an invention which features end plates which enclose a singular permanent magnet axially and radially, due to limiting movement with a conical profile (s. 4.35 H).

Ushida et al. [271] use a spacer-like feature made from expanding polymers to hold the magnet in place and pressing it towards the air gap (s. 4.35 C). While this may still lead to contamination of the powder with polymeric residue, it may offer the option of volume expansion during HPMS as the magnet cavity is not fully filled with adhesive [271].

Hattori and Noda [272] manage to avoid having to coat magnets with an isolative coating by inserting them together with coated retention plates (s. 4.35 D), which is also a very recycling-friendly approach. Similar to featured mentioned prior, Gorton et al. [273] propose using a "pocket lining", which encloses the magnet from three to five sides with a metallic feature, with the coverage either being full or partial (s. 4.35 E-G).

Mochida and Imamori [280] propose a design where embedded magnets are not fully enclosed but rather just held back by an umbrella-shaped feature of the stack (s. 4.35 P), which would be beneficial for HPMS treatment as significant expansion space is given and the powder can easily fall out. Guardiola and Lasebnick [283] use a similar segment-like feature, which is inserted to fix the magnets in place like a puzzle piece (s. 4.35 S). An option to allow more expansion space is also offered by Moule and Smith [277], where the

expanding magnet during HPMS can be allowed by breaking a structure behind the magnet (s. 4.35 L).

In a more general sense, Hahn et al. [291], who studied disassembly of NdFeB containing motors, propose some measures which align with the general design for recycling approaches presented in the last subsection: standardization of joints and components, component modularity and labelling for identification, as well as choosing recyclable materials. They also mention that good protectivity of the design and proper cooling can be beneficial [291].

4.4.3.3 Research and inventions regarding D4R to recycle permanent magnets from electric machines: process design and compatibility

In terms of process design, available literature is limited. As it was mentioned before, enough space for volume expansion is required [51], not only due to jamming of the particles in the arrangement but also since the material simply does not fall apart without enough space. Jönsson et al. [49] research the effects of the rotor layout and alloy design and find that both a lack of expansion space and higher Dy additions significantly slow down the HPMS reaction. They furthermore recommend to adapt the HPMS process so the material is moved during processing [49]. Overall HPMS processing parameters are furthermore well-established, however, clean, demounted magnets without contaminants are assumed.

Few studies consider the demounting and preprocessing of NdFeB. The most comprehensive, very recent study is by Hahn et al. [291] who propose a methodical approach to disassembly consisting of a pre-treatment to clean and open the housing, visual inspection and sorting for shredding and dismantling, disassembling down to the rotor, demagnetizing the rotor and then removing the magnets. This can be seen as a sensible approach, considering safety and making use of the possibility to only selectively disassembly. A patent by Ortiz Liebanas [292] presents an automatized method to disassemble rotors with a combination of mechanical excitation and thermal treatment.

The major knowledge gaps are therefore identified as the methods required to separate the magnets, what contaminant levels are tolerable and the viability of “common” motor features for recycling.

To approach filling these gaps, several investigations have been launched, which are presented in the following chapter 5.

Chapter 5

Practical Investigations

In this chapter, several conducted studies are integrated. In the first section 5.1, the overall methodology is explained in some additional detail to the provided publications, in the following sections 5.2 to 5.4, the published studies are presented. In section 5.5, some minor investigations are briefly introduced and in section 5.6, the overall results are summarized before leading over to chapter 6 on the implementation of the results of the investigation.

5.1 Methodology

The used methodology comprises a laboratory-scale setup for processing electric machines and NdFeB magnets as well as analytical methods to determine the success of the methods. The parameters for processing are mentioned in the sections 5.2 to 5.4.

5.1.1 Laboratory scale processing methods

For disassembly, a common array of tools, including screwdrivers, angle grinders, hammers, pliers, x-acto knifes, tweezers, different manual bearing pullers, and a heavy wrench are sufficient for basic tasks. Ideally, the tools are non-magnetic. Using protective gear is highly recommended.

For heatless demagnetization and magnetization, an impulse magnetizer can be used, where samples can be inserted into a solenoid coil and pulsed with a defined current and magnetization direction.

For thermal demagnetization, a simple muffle furnace heating to 400 °C is sufficient, however, when any organic materials like polymer coatings and adhesives are involved, proper deduction and waste gas treatment are necessary. Additionally, for larger parts, a heat gun is useful. Furthermore, an induction furnace combined with a graphite crucible was tested for demagnetization, which is a very quick option but must be sufficiently controlled to avoid extreme temperatures.

For decoating and cleaning, primarily sandblasting with a corundum medium is used along with an ultrasonic bath with isopropanol.

The magnets are processed via HPMS using a custom hydrogen reactor inside a protective atmosphere (Ar) glovebox, as depicted in Figure 5.1 at room temperature and varying pressures. The same set-up is also used to process neodymium hydride.

For degassing, sintering and annealing, a tube furnace is used in conjunction with an Inconel 625 furnace tube or a glass furnace tube, where the material is inserted on an aluminum oxide or yttrium oxide ceramic crucible covered in iron or tantalum foil to avoid reaction of the NdFeB material with the crucible. The tube is filled with material inside the glovebox and the resulting Ar-atmosphere. The Ar and degassed H are continuously evacuated at high vacuum, using a turbo pump.

Milling is conducted with a high energy ball mill or by an external service provider for jet milling.

To press and sinter, powder is pre-compacted in a silicone mold in the glovebox and then covered, wrapped and vacuum sealed. The vacuum bag is again wrapped. The powder in the mold is aligned within the solenoid coil and then pressed in a manually operated, laboratory scale isostatic press. The aligned and pressed compact is unpacked in the glovebox and introduced into the furnace tube, which is sealed before connecting it to the vacuum pump and tube furnace.

The resulting sintered magnets can be annealed, potentially also applying grain boundary diffusion and then used for further tests.

The arrangements and methods are depicted in Figure 5.1

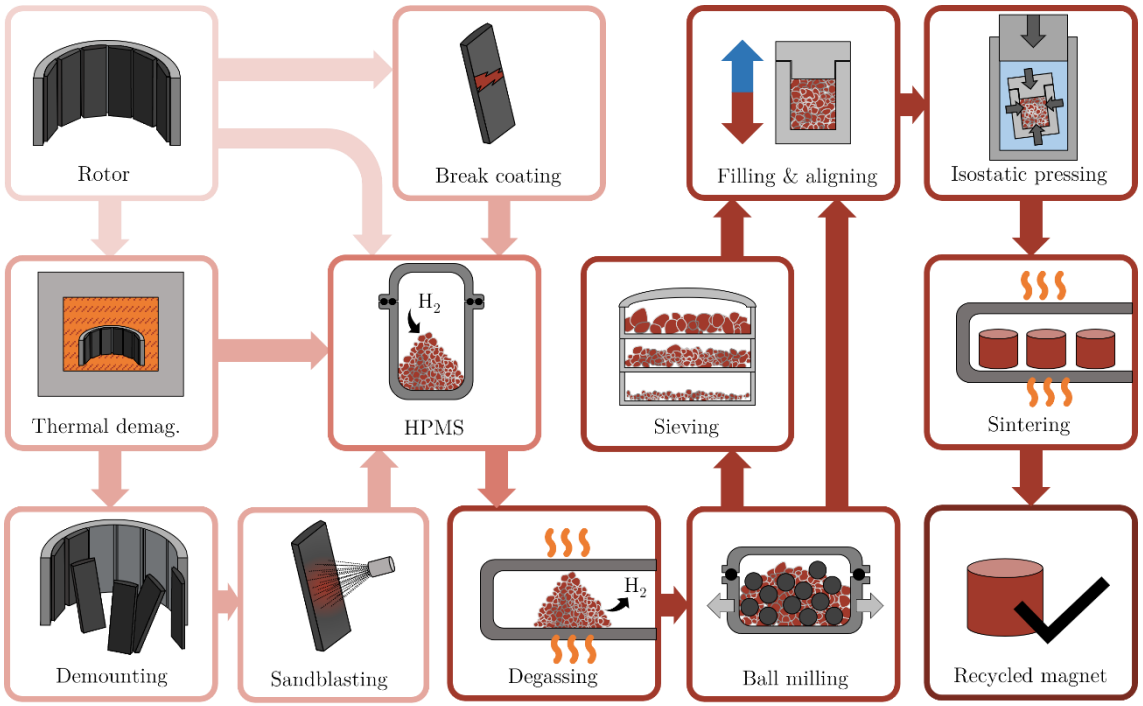


Figure 5.1: Experimental powder production and magnet recycling.

5.1.2 Analytical methods

Microstructural and superficial surface analysis is conducted in optical microscopy and scanning electron microscopy (SEM). The principle of SEM is illustrated simplified in Figure 5.2. In SEM, an electron beam is emitted by a cathode, accelerated by an anode, focused by condenser lenses and then directed to the sample surface using a scanning coil. The beam is then scanned over the sample surface point-by-point. On the sample, the beam interacts with a certain volume, which depends on the acceleration voltage.

When incident electrons from the beam are deflected, backscattered electrons (BSE) are generated, which are detected by a (usually annular) BSE detector. BSE can be up to the incident electron energy and the electron yield, corresponding with the image brightness, depends on the atomic number. Heavier elements appear brighter in BSE imaging.

If an incident electron (or a BSE) collides with a valence electron, the energy transfer can agitate the valence electron, being emitted as a secondary electron (SE). SE imaging is more surface sensitive due to the low energy of the emitted SE, whereas only those close to the sample surface can exit the sample. Due to this, SE imaging is well suited to show the sample topography. The vacancy left behind by the excited SE can be filled by electrons from a higher energy level. The excess energy is then emitted as characteristic x-ray.

In both optical microscopy and SEM, it is possible to analyze the magnet microstructure (in light microscopy, etching is usually required) and domain structure using the magneto-optic Kerr effect. For an optical microscope, polarization is required. To image magnetic domains in SEM, SE imaging with lowered acceleration voltage combined with mirror-quality polishing is required. Magnet powder can also be analyzed in SEM.

To prepare samples for SEM analysis, powder can be mounted on a conductive adhesive pad, alternatively, when a bulk magnet is monitored, it can be hot- or cold invested, ground, polished and if required, further treated by either ion polishing, etching or sputtering with a conductive material to avoid charging the surface during SEM analysis. The surface must be of mirror quality with minimal scratches for at most ideal results. For this, an NdFeB-specific method using water-free diamond polishing was developed and refined by the author. First, the magnet samples are invested in conductive resin, then ground with #320, #500, #800 and #1200 grinding paper in water. Then, they are ground once again for a prolonged time at #1200 in a water-free lubricant. Subsequently they are polished in three steps, using water-free diamond solutions at 6 µm, 1 µm and 0.25 µm. It was found that 3µm oftentimes dislodges oxides in the triple points. Therefore, longer polishing is done at 1 µm, followed by at least 12 min of polishing at 0.25 µm at low rotational speed. Between the grinding steps, an ultrasound bath is used. Between the polishing steps, the samples are merely gently but thoroughly wiped off with a soft cloth and isopropanol to ensure that the brittle oxide phases on the surface are not dislodged. Furthermore, it is important that the magnets are demagnetized to avoid distortion of the image. However, as even a demagnetized magnet has magnetic domains which cancel each other out overall, the beam can be distorted due to the magnetized domains. It is advisable to align the beam frequently and work with higher acceleration voltages when visible distortion occurs.

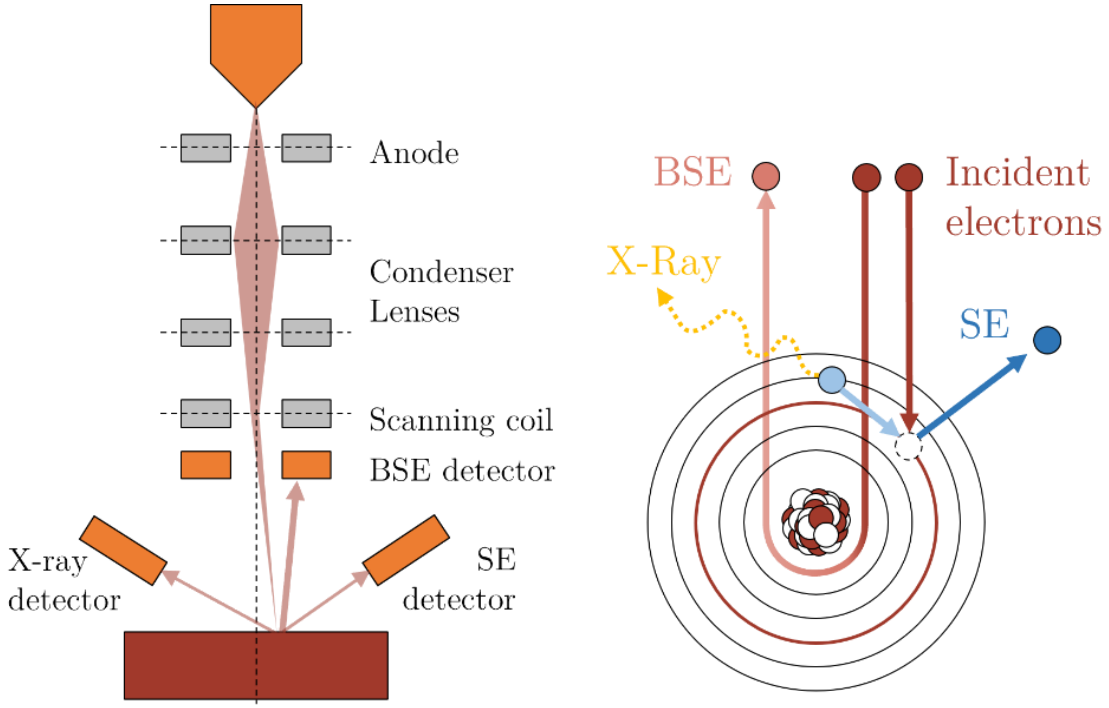


Figure 5.2: Basic principle of SEM imaging.

For chemical analysis, several methods are available. For less-accurate but localized and somewhat surface sensitive analysis, electron-dispersive x-ray spectroscopy (EDS) is used, where the characteristic x-rays generated from interactions of the incident electrons are collected and dispersed by energy to count and quantify incoming x-rays to quantify the specific emission lines. The achieved spectrum can be used to quantify; however, the detection and accuracy are limited, especially when it comes to light elements and elements which have close-by characteristic x-ray lines. The accuracy can be improved by optional calibration.

It should be mentioned that more reliable methods like electron energy loss spectroscopy (EELS) or more surface sensitive methods like auger electron spectroscopy (AES), x-ray photoelectron spectroscopy (XPS) or secondary ion mass spectrometry (SIMS) may be advisable for some applications. However, they were not readily available for this investigation. Overall, the available methods were sufficient for all applications except the detailed analysis of surface passivations and phosphatizations.

For more exact compositional analysis of metallic bulk material without certain spatial resolution, inductively coupled plasma with optical emission spectroscopy (ICP-OES) was used. In ICP-OES, the material is digested and diluted to be fed into a plasma torch, where the dissolved materials are vaporized and ionized, then falling back to lower energy states and emitting light with characteristic wavelengths. The emissions generated in the plasma torch are split into finely resolved by a prism and an Echelle-lattice into wavelength ranges of few pm. A solid-state detector collects electrons. The fanned out light emissions then strike the pixel matrix of the solid-state detector. The energy is collected and analyzed. A spectrum is generated and then analyzed for the line locations and intensities. For ICP-OES, it is very important to measure the materials against a tuning solution which comprises the to-be-analyzed elements for calibration and selecting characteristic lines which avoid error from spectral interference, line broadening, self-absorption and scattering as well as issues from sample preparation. The principle of ICP-OES is shown in Figure 5.3.

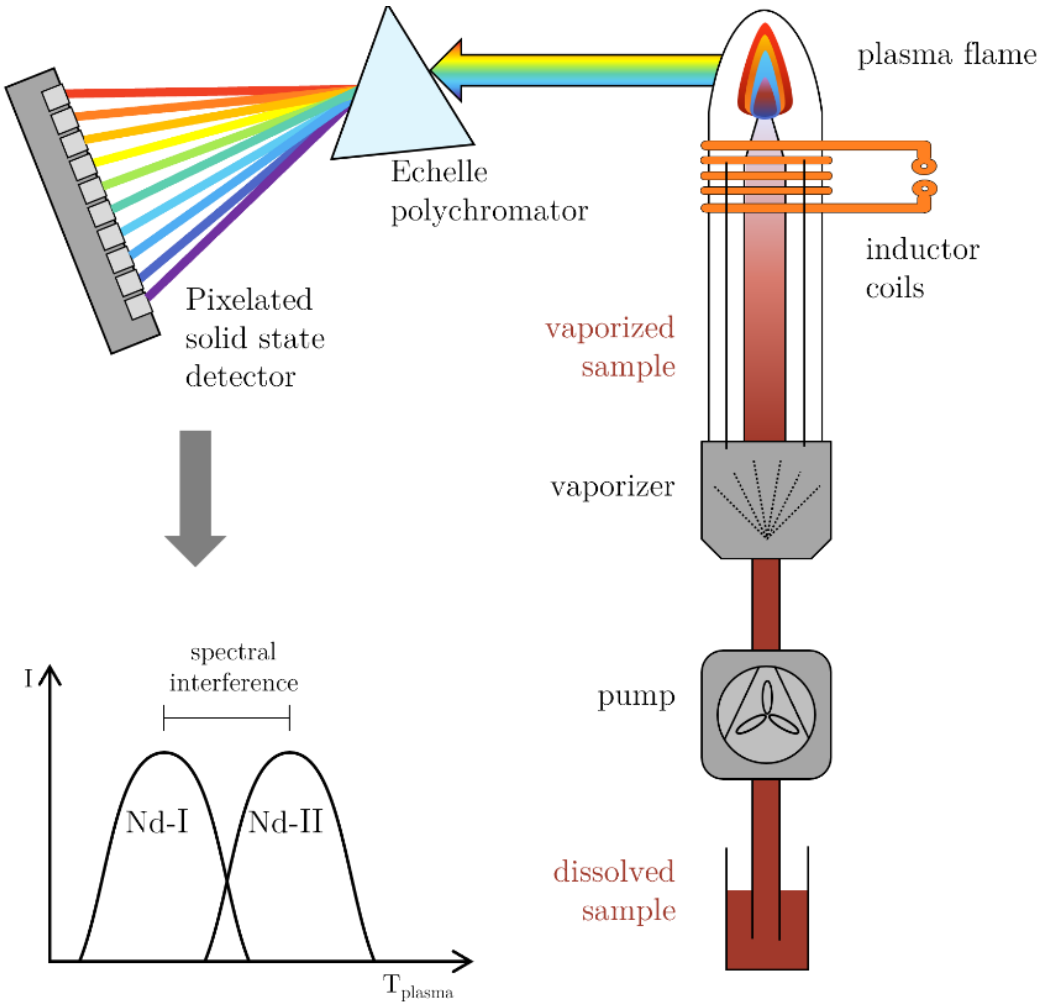


Figure 5.3: Basic principle of ICP-OES.

For organic material analysis, hot gas extraction is used. For analysis of O, N and H, inert gas fusion technology is used. Samples are weighed and inserted into a graphite crucible. Powders are sealed in a Sn capsule and a Ni capsule to avoid further oxygen uptake. The sample is heated in the crucible to liberate O, N and H and O reacts with the graphite, forming CO and CO_2, a noble gas acting as a carrier flushes the hot gas out.

The quantification is achieved by using non-dispersive infrared detectors (NDIR) and thermal conductivity sensors (TC). NDIR detects gas species by wavelength-specific IR absorption and TC cells by changes in heat conduction within the gas stream.

First, CO and CO_2 are measured by NDIR cells, then CO and H are oxidized to CO_2 and H_2O, which are detected by a second set of NDIR cells. Subsequently, the measured analyzed are scrubbed from the gas stream, whereas the remaining N can be detected via a detection system using a third set of NDIR and TC.

The analysis of C and S is conducted in another device using ceramic crucibles. Here, the sample is combusted in a stream of O, oxidizing C and S to CO, CO_2 and SO_2, which are flushed through a drying agent and measured in an NDIR cell to detect SO_2. The analytes are then oxidized to CO_2 and SO_3 and SO_3 is filtered out. Finally, CO_2 is measured. The principle of hot gas extraction is visualized in Figure 5.4.

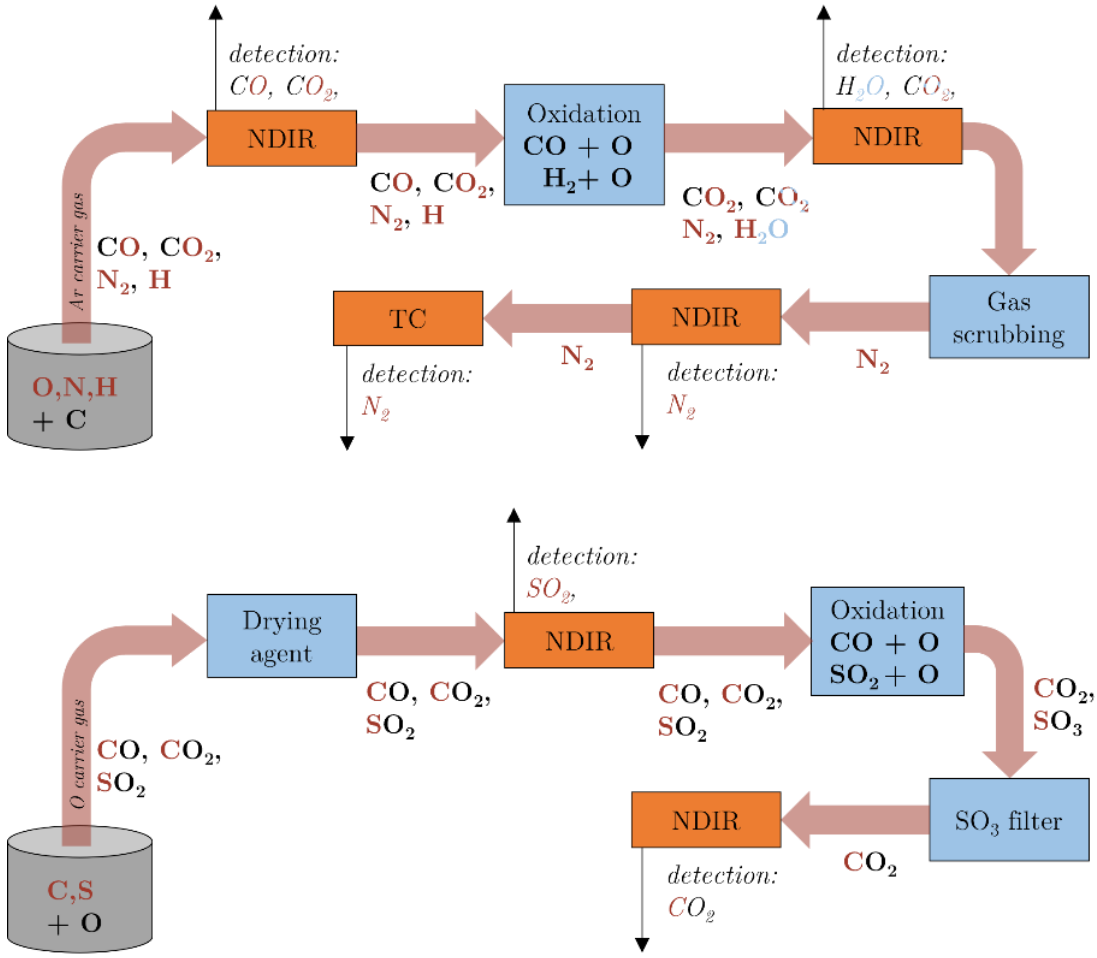


Figure 5.4: Hot gas extraction for analysis of organic constituents.

For analysis of the magnetic properties, a “Hystograph” is used, where the magnet receives a controlled slow magnetization and demagnetization, while the flux density B is recorded with a fluxmeter coil and integrated over the sample geometry. Polarization J is calculated by subtracting the applied magnetic field from the overall flux density. By this, a hysteresis is recorded, showing the B/H and J/H curve with all relevant properties. The interpretation of the hysteresis and magnet properties was explained in section 4.1.2.5.

Finally, for particle size analysis, a laser diffraction particle size analyzer is used, which can be operated wet and dry. For density calculation, a scale equipped with a specific density kit is used after which the density can be calculated by the Archimedean principle.

5.2 Publication I: Processability and Separability of Commercial Anti-Corrosion Coatings Produced by In Situ Hydrogen-Processing of Magnetic Scrap (HPMS) Recycling of NdFeB

The first major, peer reviewed study was conducted to investigate two important questions: First of all, what machine designs can be processed in situ, without demounting the magnets from the rotor, and secondly, what contaminants can be separated from the powder afterwards. In situ treatment and powder separation potentially offer a largely shortened processing path by omitting time-consuming manual handling primarily from disassembly, but also from coating removal and cleaning to separate the contaminants from the bulk magnet. Powder separation may be more constrained in terms of what is viable, but it can be conducted in a highly automatized way.

The study is meant to extend a prior investigation where the author was involved [53] and was conceptualized and executed by the main author. The ICP-OES analyses and methodical support were provided by Co-Author and ICP expert Peter Fleissner. Supervision, funding acquisition and project administration were provided by the doctoral supervisors Prof. Dr. Carlo Burkhardt and Prof. Dr. Spomenka Kobe. All co-authors assisted in proofreading and review.

The paper was published on 21. May 2024 in the Journal Materials under the DOI:10.3390/ma17112487. The manuscript along with review reports are available open access. The paper was distinguished by an editor's choice award.

The manuscript has been inserted in the following using identical text and figures; however, the formatting, section numbers, figure numbers, table numbers and reference numbers are adapted to be seamlessly integrated into the dissertation document. The references are integrated in the overall reference section.

Processability and Separability of Commercial Anti-Corrosion Coatings Produced by In Situ Hydrogen-Processing of Magnetic Scrap (HPMS) Recycling of NdFeB

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Abstract

The recycling of NdFeB magnets is necessary to ensure a reliable and ethical supply of rare earth elements as critical raw materials. This has been recognized internationally, prompting the implementation of large-scale legislative measured aimed at its resolution; for example, an ambitious recycling quote has been established in the Critical Raw Materials Act Successful recycling in sufficient quantities is challenged by product designs that do not allow the extraction and recycling of these high-performance permanent magnets without excessive effort and cost. This is particularly true for smaller motors using NdFeB magnets. Therefore, methods of recycling such arrangements with little or no dismantling are being researched. They are tested for the hydrogen-processing of magnetic scrap (HPMS) method, a short-loop mechanical recycling process. As contamination of the recycled material with residues of anti-corrosion coatings, adhesives, etc., may lead to downcycling, the separability of such residues from bulk magnets and magnet powder is explored. It is found that the hydrogen permeability, expansion volume, and the chosen coating affect the viable preparation and separation methods as recyclability-relevant design features.

Keywords: HPMS; NdFeB recycling; coatings; IPMSM; design for recycling

5.2.1 Introduction

The benchmark of having to recycle 25 % of NdFeB by 2030 was set by the Critical Raw Materials Act (CRMA) [6], recognizing the significant gap between the projected supply and demand of NdFeB magnets due to their increasing use in high-power permanent magnet applications such as electric vehicles or wind turbines [15]. Rare earth deposits are scarce and difficult to mine: primary mining and chemical processing pose a threat to workers and the environment [293]. Recycling could break the vicious circle of supplying environmentally critical materials such as NdFeB to the clean energy transition and electromobility. Hydrogen-processing of magnetic scrap (HPMS) for sintered NdFeB enables short-loop recycling, avoiding multiple mechanical and chemical refining processes [39]. By using hydrogen to embrittle and pulverize the magnets, recycling can take place from the powder stage, where the powder is prepared for a subsequent shaping and sintering process. This is energetically favorable compared to hydro- and pyrometallurgical recycling [33].

Due to their high reactivity, NdFeB permanent magnets must be protected from corrosion during operation. Corrosion protection is usually achieved through the use of a suitable coating. However, the production of high-performance magnets via HPMS requires the high purity of the processed recyclate powders, with the need for a low oxygen and carbon content, as well as a low content of other unwanted chemical elements, which may be present in the form of coatings, adhesives and other residues, thus affecting the quality of the recycled magnet. If detrimental, such residues must be separated from the NdFeB bulk either before or after the HPMS process.

For large applications such as wind turbines, electric vehicle traction motors or computer hard disk drives, automated dismantling and extraction of the NdFeB permanent magnets is suggested for commercial viability [140, 155]. This process is hereby referred to as ex situ, as the magnets are removed prior to the HPMS, allowing for the removal of the coatings and adhesives, which is advantageous to achieve non-contaminated NdFeB powder. However, other applications, particularly small motors, which may not be worth dismantling due to the small amount of material involved but which occur in large volumes, are also important sources of scrap. In order to present a solution for the recovery of these important scrap sources for the circular economy, the processing of coated and passivated NdFeB in a non-dismantled state (in situ) is being investigated.

5.2.2 Materials and methods

In this section, the used general methods (Section 5.2.2.1), the test subjects and exact test setups (Section 5.2.2.2) are explained, followed by a short review of commonly used coating materials (Section 5.2.2.3).

5.2.2.1 Methods

Hydrogen-processing of magnetic scrap (HPMS) is a patented process developed by researchers at the University of Birmingham [133]. Magnets are placed in a reactor, which is then evacuated and filled with pure hydrogen at adjustable pressures at laboratory temperature. The Nd in the grain boundaries reacts with hydrogen to form $NdH_{\sim 2.7}$ and the hard-magnetic $Nd_2Fe_{14}B$ matrix phase accommodates a number of interstitial hydrides, expanding by up to 5 % in volume [21]. This process is illustrated in Figure 5.5, together with a common subsequent processing route by pressing and sintering. The decomposition of the Nd-rich phase reduces the integrity of the matrix, leading to embrittlement, and the volume expansions cause the grains to be pushed apart, resulting in coarse, friable aggregates.

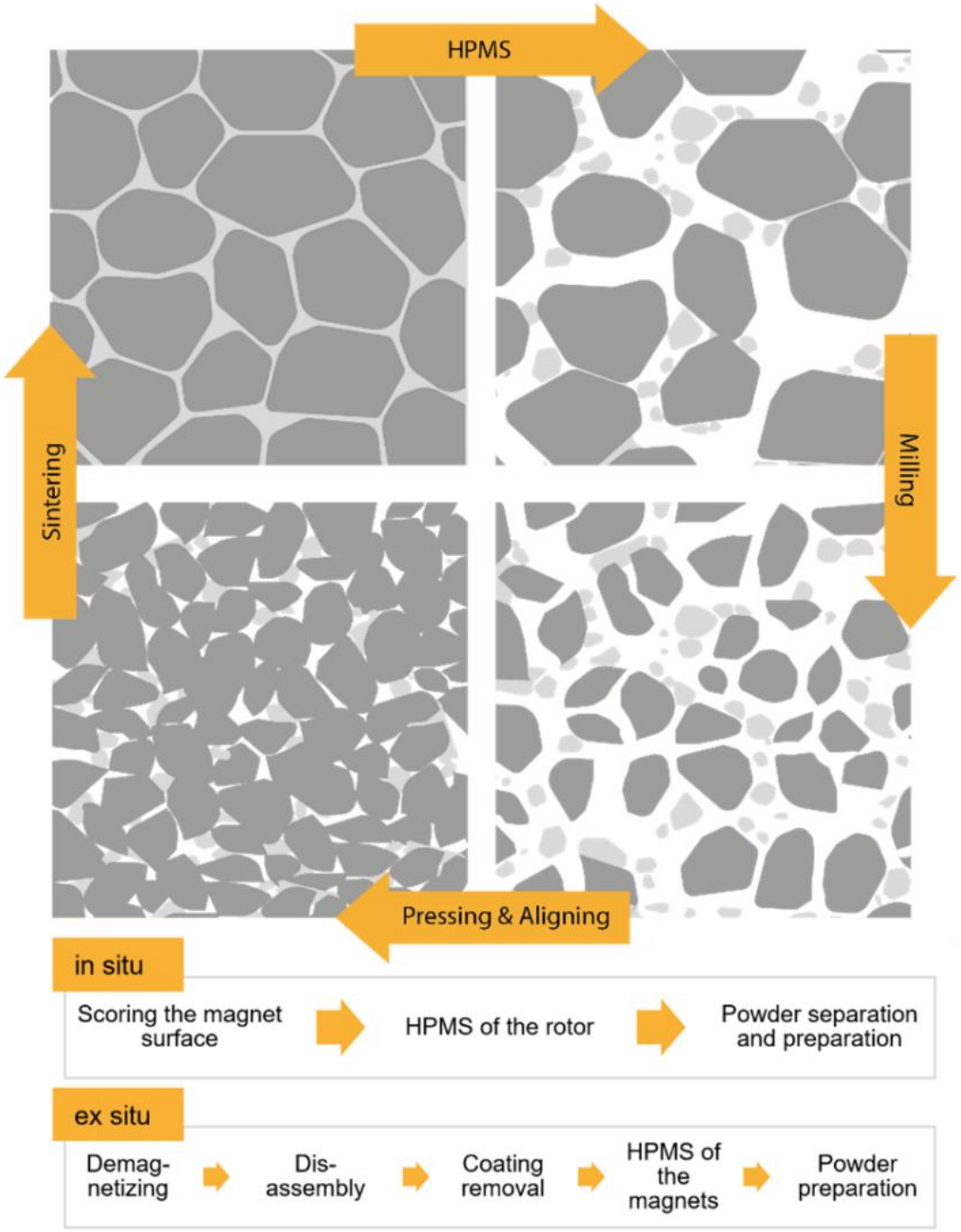


Figure 5.5: Circular lifecycle of NdFeB using HPMS and resintering, process comparison of in situ and ex-situ treatment in HPMS seen below: in situ treated magnets are exposed to hydrogen while still mounted and decrepitated to powder, which must then be removed and separated from coating residues; ex situ-treated magnets are demounted and their coatings are removed prior to treatment.

The material is kept in a bespoke reactor in a hydrogen atmosphere until the hydrogen consumption is reduced to almost zero, assuming the reaction has reached equilibrium. The powder is then partially degassed by evacuation and the powder is removed from the reactor. The result after the HPMS can range from a single crystal powder with particle

sizes below 10 µm to aggregates several mm in size. This powder has to be milled to counteract the grain growth during sintering. To initiate this process, the hydrogen must have a pathway to enter the microstructure at the grain boundaries. Dense coatings must therefore be removed or partially fractured. If the coating has not been completely removed prior to the HPMS, it may disintegrate, remain as mostly uniform carcasses or flakes, or adhere to the individual pieces of NdFeB [53], and the residues are typically separated by a manual post-treatment such as sieving or sifting.

If in situ HPMS is performed on a non-dismantled array, a hydrogen pathway may be provided by damage to the coating. However, the magnet may not disintegrate properly if there is insufficient space for expansion [51].

5.2.2.2 Experimental

A range of EOL and production scrap rotors less than 100 mm in diameter were used in this study. As there were no EOL rotors available with an Ni-based coating, only relatively fresh production scrap, hard disk drive magnets with different Ni and Ni-Cu coatings were added, as the behavior of the material can vary greatly depending on the corrosion state of the NdFeB bulk, and these supplementary magnets were necessary to obtain representative results in the powder separation experiments. An overview of the samples is provided in Table 5.1. Several internal permanent magnet synchronous motor (IPMSM) rotors as well as surface-mounted permanent magnet synchronous motor (SPMSM) rotors with an outer rotor (OR) or inner rotor (IR) arrangement were used. The rotor types and the test method are shown in Figure 5.6. The coatings were identified and characterized by SEM-EDS. A short review of the relevant characteristics and variations of the common anti-corrosion coatings relevant to this study is provided in Section 5.2.2.3 Materials to support the later discussion.

Table 5.1: Samples tested in the investigations. Supplied in a magnetized condition.

Rotor Sample	**Motor Type**	**Magnet Mounting**	**Coating**	**Scrap Type**
1 IR-IPMSM-Zn	Inner rotor	Internal, press-fit	Zn	Prod. scrap
2 IR-IPMSM-Zn	Inner rotor	Internal, press-fit	Zn	Prod. scrap
3 IR-IPMSM-Zn mix	Inner rotor	Internal, glued	Zn	End of life
4 IR-IPMSM-P	Inner rotor	Internal, press fit	Phosphate passivation	Prod. scrap
5 IR-SPMSM-ZnEp	Inner rotor	Surface mounted, glued	Zn-Epoxy	Prod. scrap
6 OR-SPMSM-NiCuNi	Outer rotor	Surface mounted, glued	Ni-Cu-Ni	Prod. scrap
Disassembled magnet samples		**Source**	**Magnet mounting**	**Scrap type**
7 Ni-Cu-Ni		Sample (6)	Demounted	Prod. scrap
8 Ni- and Ni-Cu-based mix		Hard disk drives	Demounted	End of life

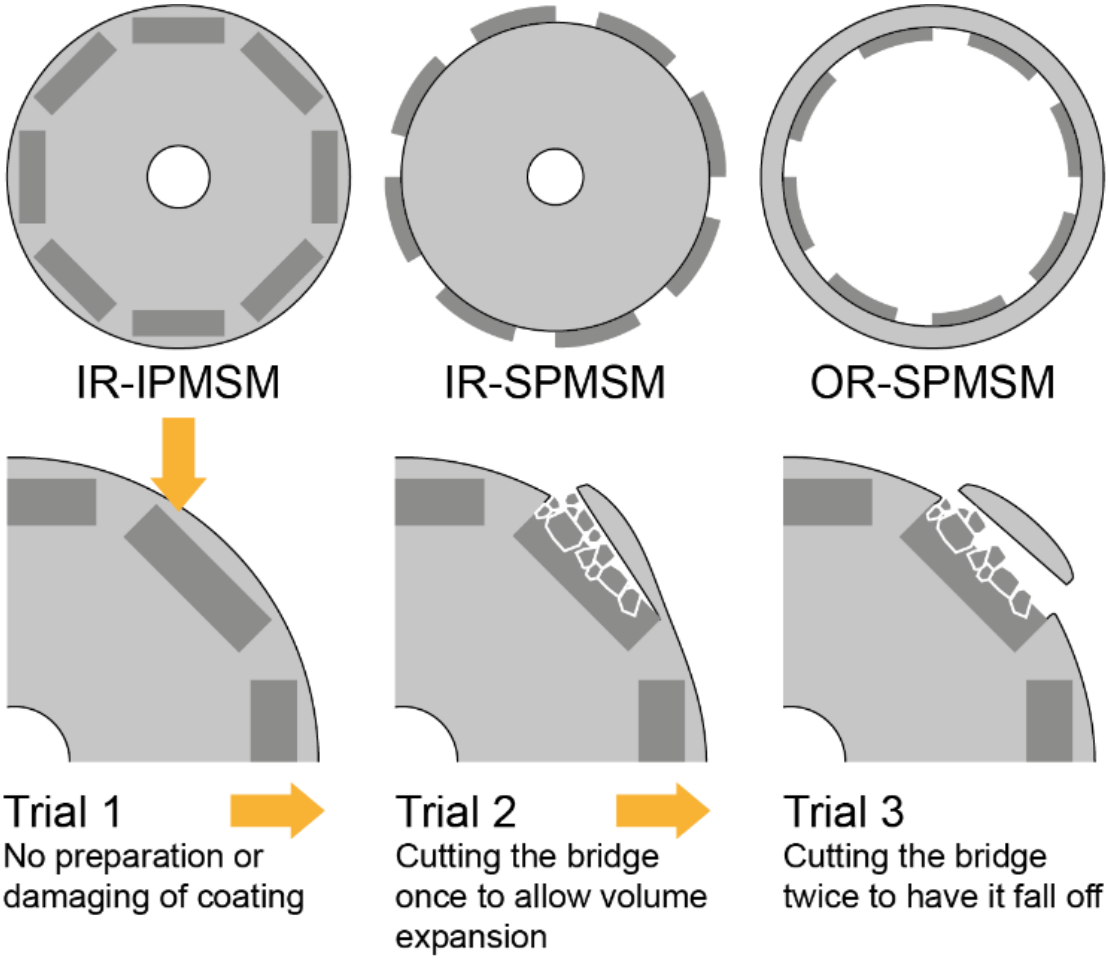


Figure 5.6: Tested rotor arrangements visualized: inner rotor (IR) arrangement of internal permanent magnet synchronous motors (IPMSMs) as well both IR and outer rotor (OR) arrangements of surface mounted permanent magnet synchronous motors (SPMSMs). The preparation methods for the in situ trials are visualized below.

As a baseline for the in situ testing, rotors with passivated magnets were tested without any preparation. They were exposed to hydrogen to determine if the passivation was permeable. Tests on rotors with metal-coated magnets or magnets with passivations that were found to be non-permeable to hydrogen in previous tests were carried out after damaging the coating by scoring or sandblasting. For the HPMS-processing trials, a reactor containing rotors with attached magnets was filled with hydrogen at 3 bar. When the pressure dropped below 2 bar due to the ongoing reaction, the reactor was refilled until the pressure drop per hour was less than 0.1 bar. If the magnets did not disintegrate due to a lack of expansion volume, another attempt was made with the bridges cut once, allowing the bridge to deform and thus to accommodate the expanding magnetic material, as shown in Figure 5.6. If this method also failed to induce a reaction, the bridge was cut on the other side as well. As a result, it fell off when the magnet was decomposed and demagnetized. Additional tests were carried out with variations in the HPMS-processing parameters to increase the particle size of the aggregates to avoid clogging by stuck particles.

The resulting powders, as well as additional HPMS powders from the single magnet samples 6 and 7 with metal coating remnants, were further tested in separation experiments

by milling, followed by sieving, in a further development of an earlier method reported in Burkhardt et al. [53]. Alternatively, chemical separation was investigated by exposing the powder to an acidic liquid. This method is of secondary consideration and is further worked on by other members of the research group. ICP-OES was used to monitor the effectiveness of the separation. ICP samples are digested in aqua regia solution with 5 % nitric acid using an MARS6 microwave, heating the digestive solution to 200 °C. The solution was then diluted and measured in the iCAP 7400 ICP-OES against a custom tuning solution.

Laboratory-scale vibratory ball-milling was carried out using a RETSCH mm500 nano with 3 10 min cycles at 35 Hz with a steel ball to powder ratio of 1.6:1. A Haver-Bröcker EML 200 sieve cascade with 1000 µm, 500 µm, 250 µm, 125 µm, 63 µm, 45 µm and 25 µm sieves was used for the sieving trials and jet milling with an integrated hydrocyclone stage was tested by an external supplier on a few Ni-Cu-Ni-coated magnets as a method of coating separation.

5.2.2.3 Materials

Anti-corrosion coatings are used due to NdFeB magnets being susceptible to corrosion, which is due to the corrosion potential of the bimetallic nature of the material, leading to intergranular corrosion through the formation of corrosion batteries [106, 294]. If the highly reactive Nd in the Nd-rich phase is in an ionic bond, e.g., by oxidation, it cannot be hydrided and embrittled, nor can it act as a liquid phase during sintering. The corrosion behavior of the material itself can be improved by alloying the NdFeB system with suitable metals to reduce the highly negative standard electrode potential of the Nd-rich phase. Alloying with elements such as Al, Co, Cr, Ga, Nb, Pb, P, Sn, Ti, Zr, Ho or Ce can improve the corrosion resistance of the magnets [295–301]. Common metallic coatings are Zn [104, 302, 303], Ni [304–306], Ni-Cu-Ni [307, 308], or less commonly, Sn [53], Al [309–311] and Zn-Ni [107]. Metallic coatings are often deposited by electroplating [303, 305]. Other methods of depositing metallic coatings include sputtering [312], vacuum evaporation [310] or electroless plating [304]. Also common are various types of passivation [53, 313, 314]and organic coatings and adhesives such as epoxy resins [108, 315] and composite coatings such as epoxy resins filled with ceramic and metallic particles [316, 317] or even multi-layer coatings using a combination of passivation, metallic coatings and epoxy coatings.

Ni-based coatings, including Ni-Cu composites, can be produced by a variety of methods with varying benefits, but they are generally considered to provide superior corrosion protection due to their high heat resistance, durability and impermeability. Ni and Cu coatings are cathodic due to their higher standard electrode potential. They are therefore less susceptible to corrosion than the various phases of NdFeB and cannot be used as a sacrificial anode for the highly reactive neodymium-rich phase. As a result, the requirement for the coating density is high. Coatings are often multi-layered with Cu and various types of Ni alloys. High-P (high-phosphorus) Ni coatings have excellent mechanical and anticorrosion properties compared to regular Ni coatings due to their high density and amorphous structure [304, 305], but they also have a porous structure [107]. Not exclusive to high-P Ni, but very common, are problems with poor adhesion and bloating of the coating. This is caused by the immersion of the magnet in electrolytes, where the grain boundaries are infiltrated and the resulting corrosion produces hydrogen under the coating, causing the coating to bloat and break [107]. Ni-Cu-Ni is a common nickel-based multilayer coating often observed on end of life magnets and provides excellent corrosion protection [29]. SEM images of some Ni-based coatings are shown in Figure 5.7.

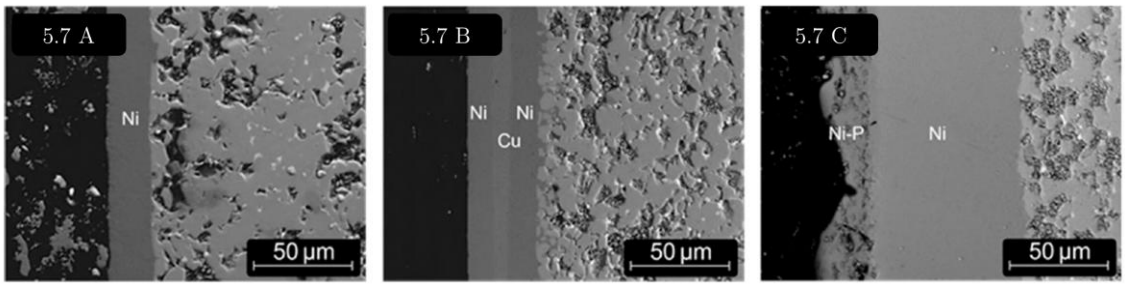


Figure 5.7: SEM-BSE images of single Ni (A) Ni-Cu-Ni (B) multilayer Ni with Ni-P (C) as observed in the samples used in this investigation. Samples shown in (A,C) are derived from mixed scrap samples 8 and (B) is derived from sample 6/7. Hitachi FlexSEM 1000II (Tokyo, Japan), W cathode, 15 kV, BSE-COMP, 200× magnification.

Zinc-based coatings offer the advantage of being able to act as a sacrificial anode due to having a lower standard electrode potential, but they have the disadvantage of not being durable at higher temperatures [303, 305]. Zinc coatings therefore do not need to be fully dense and can be relatively thin (about 10 µm [106]), while still providing good corrosion resistance to various chemically abrasive environments at lower temperatures due to their anodic nature [108]. Zinc coatings are inexpensive and add only small amounts of non-magnetic volume due to their low thickness [318]. Similar to Ni-based coatings, Zn-P is also an option, with higher corrosion resistance and excellent properties, to facilitate adhesion between NdFeB and, e.g., Ni-P or epoxy [107].

Passivations are suitable for well-protected designs, low-erosion environments, or undercoatings to protect the magnet before or during the application of the main coating. NdFeB can be surface passivated by exposing the magnet to an acidic liquid [107]. Different types of passivation have been observed [53], which still need to be categorized and analyzed in terms of their recyclability. The clear identification of passivations requires more advanced surface analysis methods, such as XPS, so only magnets with known passivations can be considered. Unlike the coatings mentioned above, some passivations are permeable to hydrogen, which can be advantageous for hydrogen treatments to recycle the magnet. Phosphating is a very popular treatment due to its improved corrosion resistance compared to other passivations [107], but it was found by Burkhardt et al. [53] that phosphatizations can be impermeable to hydrogen, which has been confirmed several times by processing trials since the end of that study.

5.2.3 Results

5.2.3.1 In situ processing trials

The sample rotors and magnets were tested with different combinations of methods, starting with the least effort and then gradually adding preparation steps to successfully produce clean NdFeB powder from the sample, as described in Section 5.2.3.2.

IPMSM: Passivated and Zn-coated magnets mounted on IPMSM rotors were tested for their processability by in situ HPMS. The rotors were not demagnetized beforehand. The phosphate-passivated magnets of sample 3 showed in the first attempt (no preparation) that the coating was not permeable to hydrogen, confirming the observations of Burkhardt et al. [53]. The Zn coatings were also impermeable. In a second attempt, the externally visible ends of the magnets were damaged to allow hydrogen access. It was observed that

the lack of expansion volume can cause the coarse HPMS powder to become stuck in the steel sheet stacks. In one rotor (sample 1), it was possible to remove the powder by hitting the rotor with a mortar, but in the other cases, this was not practical. In a further test, the magnet pockets were cut as described in Section 5.2.3.2. In one case, involving a relatively large magnet (sample 2), a single cut was sufficient to cause slight deformation, with the magnet powder being loose after the treatment. In other cases (sample 3, including several similar end of life rotors), cutting on both sides was the only viable option to separate the magnet powder from the stacks. As an alternative to cutting the stacks, attempts were made to vary the hydrogen pressure and treatment time, with lower hydrogen pressures resulting in finer powder (see Figure 5.8 A). A treatment at the standard pressure used of 3 bar for 1 h resulted in a Dv50 of 330 µm and a Dv90 of 632 µm, while at 1 bar and 1 h, the Dv50 value reached 280 µm and the Dv90 value reached 546 µm. The particle size distribution was measured with a Malvern Panalytical Mastersizer 3000 (Almelo, The Netherlands) with an Aero-S dispersion unit at 4 bar, using the Mie theory and a slit size of 1.25 mm for the measurement.

SPMSM: For SPMSM rotors, it was assumed that the exposed magnet surfaces would need to be sandblasted or otherwise damaged to allow the NdFeB material to disintegrate and fall off, leaving the coating backs and adhesives behind. This was not the case, as large sections of the coating backs came off, some with the adhesive still visibly attached. This was even the case for a type of magnet with a thick Ni-Cu-Ni coating with low adhesion, which can be easily peeled off by hand (see sample 6, as shown in Figure 5.8 B). A simple method of dismantling is thermal demagnetization, which breaks down the adhesive. This should be followed by either cleaning, if the coating residue can be removed from the powder, or removing the coating altogether.

Creation of hydrogen access: In configurations where soft magnetic swarf or powder material is generated by the hydrogen access method, e.g., by sandblasting to remove Ni-based coatings or by rotary cutting of steel stacks or the coating surface, it has been found that the generated material adheres to the still magnetized NdFeB permanent magnets. As the removal of the soft magnetic swarf or powder material is very challenging, such means of providing hydrogen access are not recommended in this case due to the risk of contaminating the HPMS powder. The presence of soft magnetic phases such as α-Fe in a sintered NdFeB magnet is known to be highly detrimental to the magnetic properties [87, 319].

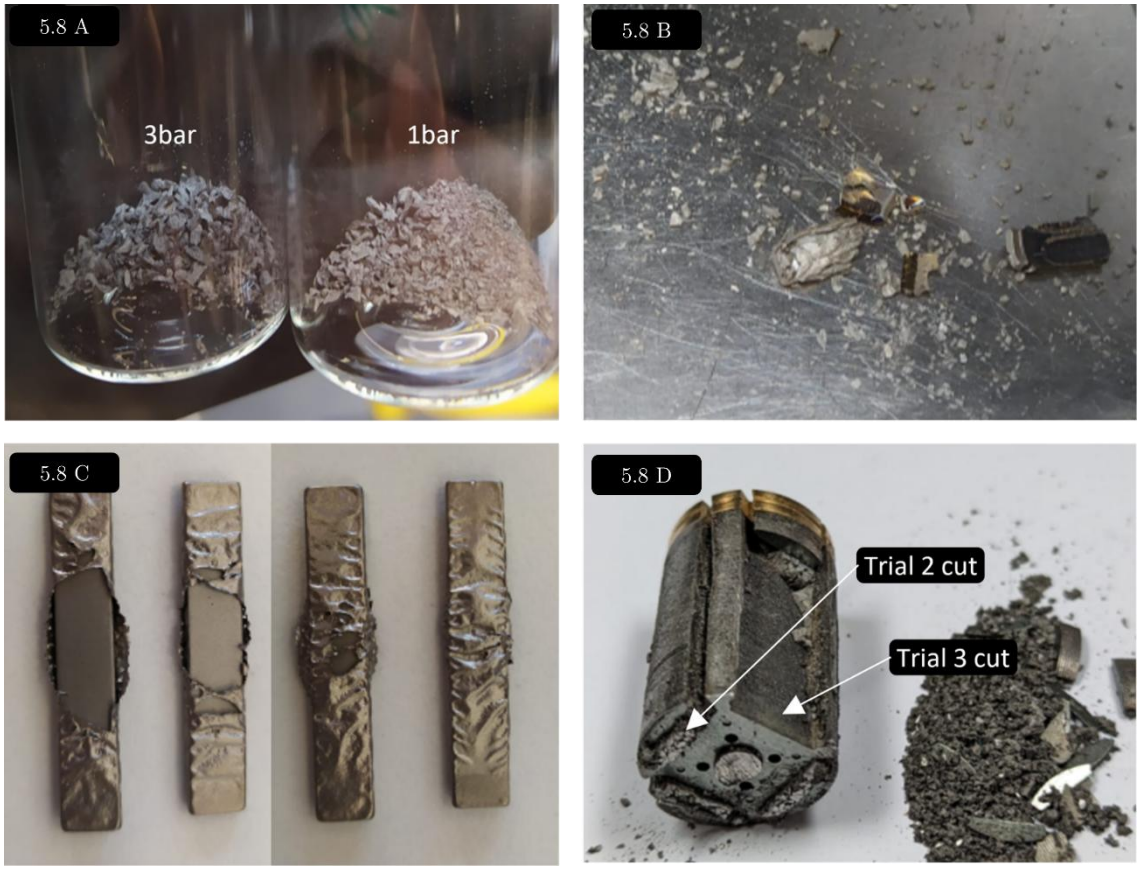


Figure 5.8: Comparison of the same type of magnets (mounted in sample 1) being treated at 3 bar and 1 bar for 1 h imaged by Leica M205 C (Wetzlar, Germany), 10× magnification (A), powder contaminated by coating backs and adhesive after in situ testing of sample 6 (B), failed sandblasting of sample 7 magnets (C) and results after cutting sample 3 once and twice, showing the decrepitated powder intermixed with rotor sheet parts next to the rotor (D). For sample details, see Table 5.1.

5.2.3.2 Coating separation trials

Coating separation of Ni-based coatings: Mechanical removal of coating residues from the HPMS powder was tested on previously removed magnets and on magnet powder produced by in situ treatment. Sieving to separate the coating flakes and NdFeB material after the HPMS proved to be challenging, as the flakes and HPMS powder overlapped significantly in terms of the particle size, resulting in a low yield. Relying on the high ductility of the Ni and Cu coatings, an attempt was made to mill the HPMS powder with all the coating residues still present. The results of these experiments, both conducted on mixed production scrap and mixed end of life scrap, are seen in Figure 5.9. For lightly corroded production scrap, excellent separation was achieved in the subsequent sieving process, with Ni-Cu-Ni flakes ending up mostly in the 125 µm sieve, with small amounts in the fraction above 63 µm, and milled NdFeB powder without Ni and Cu residues below 25 µm. For highly oxidized end of life scrap, the separation after milling was less successful. As expected, advanced corrosion interfered with the process, resulting in non-hydrided pieces and therefore large powder agglomerates, some of which could not be completely broken down during milling. Here, the sieve fractions between 25 µm and 125 µm contained larger quantities of NdFeB as well as low levels of Ni, which was thought to be

due to a brittle, cellular high phosphorous Ni layer found only in the magnets used in this particular batch of powder. Sandblasting worked for most Ni-based coatings tested, with one exception (sample 6/7 (demounted)) where the coating was deformed rather than removed (see Figure 5.8 C), which was due to the high ductility of the material and its uncharacteristically low adhesion to the magnet.

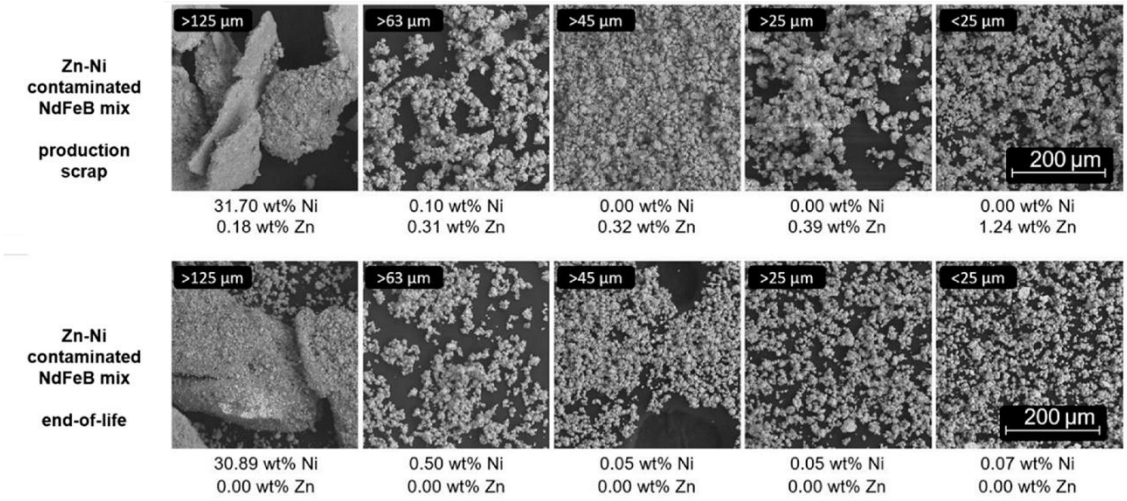


Figure 5.9: Ni and Zn content of sieved fractions determined by ICP-OES of low-corrosion production scrap (sample 7, sample 2, same manufacturer) and high-corrosion end of life scrap (sample 8) – the amounts of powder in the images are not indicative of the amounts found in each fraction. Hitachi FlexSEM 1000II, W cathode, 15 kV, BSE-COMP, 200× magnification.

Coating separation of Zn-based coatings: For the Zn coating residues, separation by sieving before and after milling was not possible. After sieving, most of the Zn ended up in the main NdFeB fraction, with a particle size below 25 µm. The Zn content of the powder was successfully reduced to a level not measurable by ICP by immersing the powder in a 1 molar citric acid bath for 30 min at room temperature. As an alternative, sandblasting has been shown to be a fast and reliable method of removing Zn coatings. The material losses are minimal in sandblasting. Sandblasting Zn-coated magnets with a coating thickness of an average of 18 µm resulted in an average total decrease in thickness of the sample of 24 µm per side.

5.2.4 Discussion

In order to assess the suitability of a coating for recycling by the HPMS, several issues need to be considered. Firstly, a magnetic coating is selected to provide optimum protection in the operating environment to ensure the desired function over the lifetime of the magnet-containing component. Good corrosion protection is also beneficial for HPMS recycling, as the high oxygen uptake in EOL magnets leads to Nd oxides that prevent proper wetting of the grain boundaries during resintering of the HPMS powders. This impairs the decoupling of the grains, reduces coercivity and affects the density and thus the remanence in recycled magnets [49, 316]. Secondly, easy removal of the magnets from the array and good separability of the coating from the magnetic material are key to successful recycling. Under certain conditions it is possible to treat small IPMSM rotors in situ without disassembly.
Since the HPMS powder becomes stuck in the stacks as a result of the volume expansion, only a few rotors could be processed without further mechanical measures by simply

creating hydrogen access. Reliable processing is only possible by either cutting into the bridge over the magnet on both sides and allowing the bridge to fall off, or by using low hydrogen pressure and omitting the cutting step. There may be limitations if the material is highly oxidized. It is believed that tumbling during the HPMS would also be useful to clear the assembly of already degraded powder, but this has not been possible with the existing laboratory-scale HPMS reactor configuration. For SPMSM rotors, it was found that disassembly could be easily achieved by thermally demagnetizing the rotors so that the adhesive dissolves and the magnets fall off, allowing the rotors to be treated in a disassembled state.

Ni-based coatings, including Ni-Cu-Ni, provide good and also tailorable chemical and mechanical protection, which relies on both the layering of the coating as well as the chemical properties like the phosphate content. Whilst sandblasting worked for almost every magnet tested, it may not be entirely reliable for Ni-based coatings if their adhesion to the magnet is inadequate, allowing them to deform rather than be removed. Separation of Ni-based coatings by sieving may be the preferred method for most small motor applications, in which case sieving the ground HPMS powder is usually less laborious than extracting the magnets combined with individual sandblasting. Nonetheless, some losses in terms of the NdFeB material yield must be expected due to it sticking to the coating carcasses before milling (see Figure 5.8 B) as well as after milling (see Figure 5.9, fractions >125 µm). Difficulties may be encountered with unsuitable (but rarely used) high-P Ni coatings, which display a brittle, cellular structure, causing them to mill finely, or a high degree of oxidation of the HPMS powder, so that the separation is not as clear. As a result, high amounts of NdFeB may be present in the sieve fractions between 25 µm and 125 µm, possibly containing some Ni contamination. In these cases, a decision must be made as to whether the contamination is tolerable for blending with cleaner material or whether the powder may be unusable when obtained by this route due to oxidation. This may impact the yield of recyclable powder produced.

For **Zn-coated magnets,** disassembly and mechanical removal of the coating by sandblasting or grinding should be preferred. With the investigated specimen, mechanical powder separation was not possible. Chemical removal with acids is possible, as Zn is more reactive than NdFeB, but a mechanical method that does not produce liquid waste or rely on wet chemical processes and subsequent drying is preferred to avoid waste and excessive effort. Theoretically, small amounts of Zn may be tolerable in the recycled material, as Zn evaporates during the sintering of recycled NdFeB magnets before the densification temperature is reached. The beneficial effects on Zn addition have been found by adding Zn via single-stage hot working [320], which is an entirely different procedure with mechanisms that cannot be compared to a possible Zn integration via sintering. However, larger amounts can damage the equipment or cause pores and fractures in the magnets due to the high vapor pressure.

For **passivations** whose low residual content due to their low thickness is not expected to significantly affect the magnetic properties of the resintered magnet, hydrogen permeability and good corrosion protection are most important features for successful HPMS recycling. Phosphate passivations provide the best corrosion protection but are impermeable to hydrogen. In practice, the type of passivation is often unclear unless specified or fully analyzed. Therefore, such magnets should be assumed to be hydrogen impermeable and the appropriate recycling treatments described above should be used. To increase recycling rates, a specific labeling, as foreseen in the Critical Raw Materials Act [6], is considered mandatory.

5.2.5 Conclusions

The recyclability of small motors with different coatings in various arrangements was evaluated and compared. No one-size-fits-all solution can provide good-quality powder at low costs for every design.

If the disassembly of small NdFeB magnet assemblies is not possible for technical or economic reasons, in situ treatment at low hydrogen pressures or by cutting the steel bridges is recommended. For passivated magnets, damage to the coating is sufficient, while Ni-based coatings (with the exception of a few high-P coatings) can be easily separated from the HPMS powder by milling and sieving, and Zn can be chemically removed, but in the latter case dismantling is the preferred option. In some assemblies, such as SPMSMs, dismantling of the magnets by thermal treatment can be very straightforward, making it ideal for automation.

The effects of the thermal treatment on the interaction of the contaminant (particularly Zn and epoxy) and the magnet should be the subject of further research. The tolerable level of contamination for the possible contaminants should be tested in further studies, as perfect separation is both technically and economically unrealistic. Based on this, it should be investigated what yield of recyclable NdFeB powder of both highly contaminated and barely contaminated samples with Ni-based coatings could be achieved with the presented method of milling and sieving. The milling process should be optimized for an industrial-scale method like jet milling to avoid material losses, which could be the subject of further investigation. The recycling of Ni and Ni-Cu scraps with residues of NdFeB could be subject to further research, too, as both materials are valuable and critical materials.

In general, knowledge of current coating and fixation methods greatly improves the chances of efficient recycling while recognizing that new recycling technologies may be developed between the design of current magnet-containing products and their end of life. Adequate and standardized labeling [54] is one important factor in increasing the quality and yield of recycled NdFeB powder generated via the HPMS short-loop recycling route.

Author Contributions

Conceptualization, L.G.; methodology, L.G. and P.F.; execution of experiments and measurements, L.G. and P.F., investigation, L.G.; formal analysis, L.G.; writing—original draft preparation, L.G.; writing—review and editing, C.B. and S.K.; visualization, L.G.; supervision, C.B. and S.K.; project administration, C.B.; funding acquisition, C.B. All authors have read and agreed to the published version of the manuscript.

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References

The references are integrated into the overall reference section of the dissertation to avoid duplication.

5.3 Publication II: Effects of Thermal Demagnetization in Air on the Microstructure and Organic Contamination of NdFeB

The second major, peer reviewed study was conducted to investigate whether thermal demagnetization can be applied before recycling without detrimental effects on the contamination. The study explores thermal demagnetization as another shortened-loop, multi-purpose method to demagnetize, to demount adhesive bonds and to remove organic contaminants. The study was planned and the experiments were conducted by the main author. While conducting this study with a focus on material contamination, which is most important for recycling, a sister study was headed by Dr. Tomaž Tomše to explore the effects of thermal demagnetization on reusability [225], with whom both studies were conceptualized to be complementary. Dr. Fabian Burkhardt further joined the conceptualization.

ONH/CS measurements and methodological, as well as software support were provided by Rosario Moreno López, imaging with a nano-SEM and calibrated EDS measurements were executed by Pierre Kubelka. Supervision, funding acquisition and project administration were provided by the doctoral supervisors Prof. Dr. Carlo Burkhardt and Prof. Dr. Spomenka Kobe. All co-authors, but primarily Dr. Fabian Burkhardt, assisted in proofreading and review.

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The manuscript has been inserted in the following using identical text and figures; however, the formatting, section numbers, figure numbers, table numbers and reference numbers are adapted to be seamlessly integrated into the dissertation document. The references are integrated in the overall reference section.

Effects of Thermal Demagnetization in Air on the Microstructure and Organic Contamination of NdFeB Magnets

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Abstract

Demagnetization is an essential step for the demounting and safe handling of end of life NdFeB. Thermal demagnetization in air is a straightforward option to demount adhesive-fixed or segmented magnets. However, this process is suspected to increase the uptake of contaminants like O, C and Zn from coatings and adhesives, potentially degrading the recyclate quality. This study tests the effects of thermal demagnetization in air at 400 °C for 15 to 240 min on variously coated samples with different initial oxidation levels. Furthermore, the possible reversal of the contaminant uptake is explored. Samples with low previous oxidation levels showed significant uptake in oxygen with a minimal diffusion depth, while the uptake depended on the used coating. The best protectiveness was achieved with Ni-Cu-Ni with an increase in oxygen of only around 30 %. Epoxy (up to ~130 % O uptake) and Zn coatings (up to ~80 % O uptake) disintegrated during the treatment and offered less protection but still made a difference compared to uncoated samples (up to ~220 % O uptake). Samples with high initial oxidation levels show no clear tendency towards further oxygen uptake and the carbon uptake is generally low, likely due to contemporary epoxy coatings featuring a passivation underneath as a barrier layer. Zn infiltration, which carried organic debris, was observed. Short demagnetization times proved to be favorable for limiting the depth of the diffusing contaminants. Mechanical coating removal after thermal demagnetization in air can mitigate the contaminant uptake, producing clean, recyclable end of life material.

Keywords: thermal demagnetization; NdFeB recycling; NdFeB demagnetization

5.3.1 Introduction

The recycling of metallic end of life (EOL) scrap, which includes permanent magnets (PM), poses significant challenges due to the tendency of ferrous scrap to stick together. This issue has led to the recommendation of avoiding PM in favor of design for recycling [11]. However, there are instances, such as in motors used in battery electric vehicles (BEV), handheld power tools and others, where avoiding PM may not be technically favorable: the lower energy efficiency and torque density of PM-free motors [234, 236] would have to be compensated by larger batteries, lower power or shorter usage times. This underscores the need for a solution that allows for the recycling of PM, especially high-performance NdFeB magnets made from critical raw materials.

An environmentally friendly recycling method is the hydrogen processing of magnetic scrap (HPMS) with a subsequent resintering of the produced NdFeB powder [45]. A comprehensive overview of hydrogen-based recycling methods and routes is given in Burkhardt et al. [321]. For this, the majority of scraps needs to be demounted and, thus, demagnetized before the treatment, as the presence of strong magnetic fields will complicate the removal. A rare exception are scraps suitable for in situ treatments [49, 190], as the HPMS process produces demagnetized powders [21].

While various methods are available for demagnetization, thermal demagnetization stands out for its simplicity and applicability. In comparison to electromagnetic demagnetization with a decaying field, known as degaussing, thermal demagnetization is a highly accessible and rather fail-proof method, as long as a sufficient temperature above the Curie temperature is reached. Significantly higher temperatures and long hold times may degrade the magnet and coating materials, though. Conversely, degaussing requires a tailored tool to generate targeted reverse fields of sufficient strength, and it may lead to residual magnetization [183]. Such tools may also lack the flexibility recyclers require with diverse scrap.

In contrast, thermal demagnetization only requires a furnace that can accommodate the specimens and maintain a temperature above the specific Curie temperature of the magnetic scrap [183], and, if necessary, the degradation temperature of present adhesives. This is an added benefit of thermal demagnetization: the decomposition of adhesives before demounting further reduces the forces to remove the magnets from the assembly. Using an ambient atmosphere accelerates the degradation of polymers compared to an inert atmosphere, where the carbon-rich contents are pyrolyzed, leaving carbon-rich char residue [180]. Not requiring inert process gas and an infrastructure to introduce them is beneficial for cost-efficiency and the environment. On the other hand, the uptake of organic contaminants like oxygen and carbon at elevated temperatures is assumed, which would deteriorate the magnetic properties of recycled magnets made from the affected scrap, since the formation of oxides and carbides hinders the formation of a continuous grain boundary Nd-rich phase [92, 196]. Also, the elevated levels of carbon present during sintering lead to the disproportionation of the hard magnetic phase due to the binding of dissolved Nd, resulting in the formation of α-iron, further reducing the coercivity [111]. This especially applies to the intended direct recycling method HPMS, as here, the powder is meant to be used as is without any kind of chemical reduction treatment, underlining the necessity to limit and monitor the contamination uptake.

The application of coatings and the process of passivation are common measures to protect permanent magnets from corrosion and general contaminant uptake while also offering further benefits depending on the type of coating. For example, epoxy resin-based coatings are common due to their electrically isolative nature to limit eddy currents. Other common coatings are Ni-based like Ni-Cu-Ni multilayered coatings and Zn-based coatings. These three coating types are most commonly found in end of life scrap together with combination coatings like Zn-epoxy.

In this paper, the effects of a combined treatment to thermally demagnetize and to decompose fixation adhesives and organic coatings are tested in the context of recyclability via hydrogen processing and resintering. The EOL magnets with different coatings treated by the combined treatment are analyzed in SEM for microstructure and coating integrity and in ONH and CS before and after. This serves to quantify, understand and mitigate the contaminant uptake of C and O. Additionally, the uptake of O and Zn near the magnet surface or coating are monitored in SEM-EDS. This study is limited to the contamination of common coatings induced by thermal demagnetization in air.

5.3.2 Materials and methods

5.3.2.1 Materials

For the investigation, EOL magnets, as well as scrap NdFeB magnets available from different waste streams, were screened. Samples were chosen to both represent common NdFeB coatings, as well as EOL magnets with a low initial oxygen content, which are hereby classified as contamination samples (lc), (defined as <1000 ppm O) and those with higher initial oxygen contents, hereby classified as higher contamination (hc) (defined as >1000 ppm O). The high contamination samples are expected to be mostly saturated with oxides, even though they have a high variation of initial oxygen contents before treatment. One low initial contamination and one higher initial contamination sample type were taken for each of the following coatings: nickel-copper-nickel (Ni-Cu-Ni), zinc or zinc-epoxy (Zn or Zn-Ep) and epoxy (Ep), as well as uncoated magnets (uc). Each sample group comprises individual magnets received from the same source (e.g., the same manufacturer or the same EOL electric machine). In contrast, the origin of the waste streams of the individual magnets varied.

The samples were selected from the available range of end of life and production scrap magnets acquired in several research projects to reflect the aforementioned criteria and to have similar dimensions.

The particularly suitable lc-Ep sample group (lowest initial contamination, low initial variance before treatment, favorable geometry) was used to create uncoated magnets lc-uc for testing by removing the coating by sandblasting prior to the treatment. Due to sample preparation constraints, this test's range of usable magnets was limited to smaller-dimension magnets with low thicknesses below 4 mm. The magnets are shown before and after treatment in Figure 5.10.

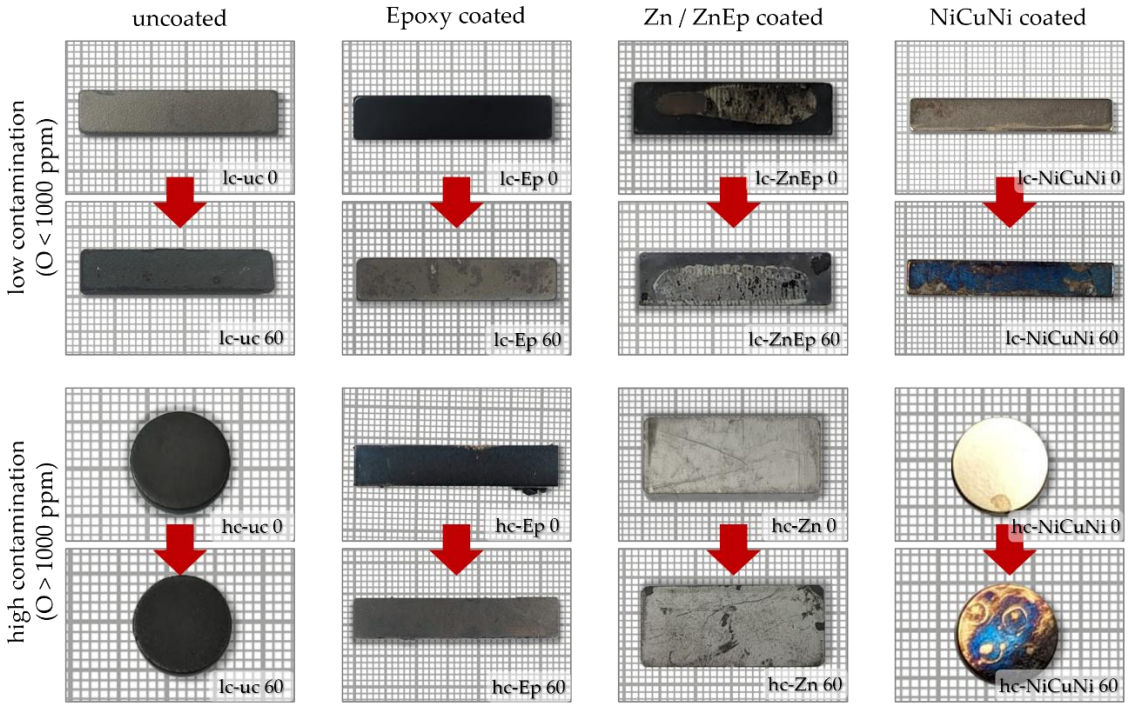


Figure 5.10: All samples used, before and after demagnetization. The change in visual aspects by demagnetization (signified by the arrows between the images) is shown. For demagnetization and for removing organic coatings and glues, the samples were treated at 400 °C in air for 60 min.

5.3.2.2 Thermal demagnetization

Thermal demagnetization was carried out in a Heraeus thermicon P muffle furnace at 400 °C in an ambient atmosphere for a minimum of 15 min and a maximum of 240 min. The temperature was chosen as it is sufficient in both thermally demagnetizing and decomposing present organic resins.

The base alloy of NdFeB has a Curie temperature Tc of 312 °C [108], but it can be enhanced by doping the alloy with heavy rare earth and transition metals, like cobalt and aluminum [117, 177].

The decomposition of basic epoxy resins begins at approximately 200 °C, and nearly complete mass loss is achieved at around 400 °C in air [322]. In previous thermal demagnetization trials with temperatures of up to 400 °C, all adhesive connections present in the samples were separable and the majority of them disintegrated. Exceptions were segmented magnets, where the exposure of the adhesive layer to air is limited and special heat-resistant adhesives with suitable additives were used.

To produce the lc-uc samples from lc-Ep samples, sandblasting was performed using a corundum abrasive at 6 bar compressed air in an HGH 1100 I sandblasting chamber, fully removing the epoxy coating and the passivation layer below.

5.3.2.3 Preparation and analysis methods

Epoxy coatings were carefully ground off with #500 grit corundum sandpaper to determine the samples' oxygen and carbon content. This step is crucial as the coatings predominantly consist of organic compounds, which would interfere with the accuracy of the results. Metallic coatings were not removed. To reduce the influence of the order of measurements, all samples were placed in air for 24 h ± 2 h after thermal demagnetization treatment.

Oxygen values were determined using the LECO ONH836 (LECO Corporation, St. Joseph, MI, USA). Carbon values were measured using the LECO CS744 (LECO Corporation, St. Joseph, MI, USA). The samples were not cut to defined dimensions in order to avoid contamination with water or organic cutting fluids, which would falsify the O and C content. Therefore, samples for both measurements were produced by impact fragmenting the treated magnets and choosing fragments in the weight range of 0.35g - 0.70 g. Due to the apparent relevance of surface effects, only nearly prismatic samples, where approximately half of the sample surface is covered by the coating or the exposed surface area, were considered to keep the treated surface to fragment volume ratio as consistent as possible.

For optical observation and EDS analysis, a Hitachi Flex-SEM 1000 II (Hitachi High-Tech, Tokyo, Japan) with a W-cathode and an Oxford Instruments micsF+ x-stream-2 EDS (Oxford Instruments, Wycombe, UK) as well as the field emission FEI Nova nanoSEM (FEI Company, Hillsboro, OR, USA) with an Oxford Instruments ULTIM MAX 65 EDS (Oxford Instruments, Wycombe, UK) were used.

5.3.3 Results and discussion

5.3.3.1 Visual aspect

Figure 5.10 shows samples before and after a 60 min thermal demagnetization treatment at 400 °C. The uncoated sample lc-uc significantly darkened. The uncoated sample hc-uc had a darker appearance beforehand, likely due to prior surface oxidation, but it also darkened further.

The epoxy disintegrated progressively during thermal demagnetization. Therefore, the original protective function of the coating is impaired, which should be kept in mind for the further processing after demagnetization. The demagnetized magnets should be immediately processed further or stored in protective gas.

By itself, thermal demagnetization is not a suitable means to fully remove epoxy coatings. The decomposition of the organic resins leaves a charred residue as the compounds are not fully volatilized. After 60 min, such residues can be fully removed with a dry cloth. Below the epoxy, the passivation is visible; see Figure 5.10 lc-Ep 60 and hc-Ep 60 as opposed to lc-uc 60 and hc-uc 60 without a passivation layer.

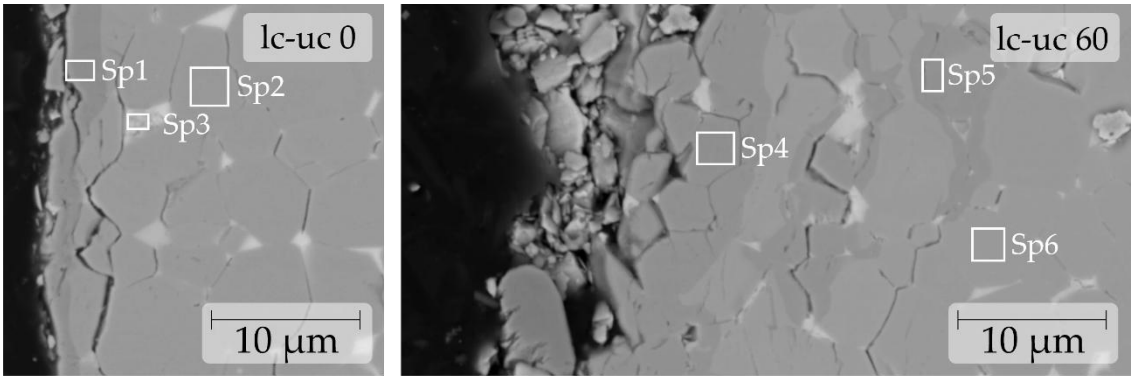
The Ni-Cu-Ni coatings are tarnished and turn blue- and orange-colored.

The Zn-epoxy coating of lc-ZnEp shows a dark gray color, and while the coating flakes are burnt off, it appears as if the lower layer has darkened. The Zn coating of hc-ZnEp appears thinner and dark areas emerge where the coating is scratched.

5.3.3.2 Microstructure

The microstructure is monitored close to the coating or the exposed magnet surface. Bulk images from the middle of the samples are omitted as no significant differences between the treated and non-treated samples are seen. When the magnets are observed on the Scanning Electron Microscope (SEM) in combination with EDS before and after treatment, the most prominent detectable changes are the diffusion of contaminants and the formation of cracks between the grains.

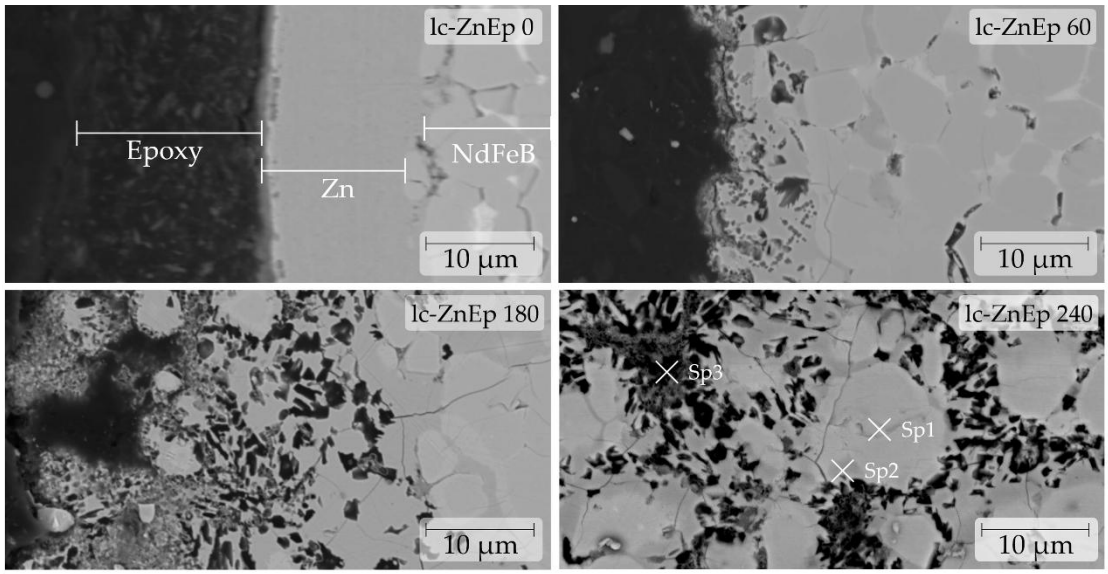
In Figure 5.11, enhanced oxygen contents can be seen in the matrix grains propagating along the grain boundaries. Unlike the visible change seen for the oxygen uptake, the carbon diffusion itself was not possible to monitor on SEM-EDS due to the loss of sensitivity of the energy dispersive x-ray spectroscopy (EDS) sensors and the BSE contrast with the decreasing atomic number and a low proportion of carbon in the material. Generally, SEM-EDS only yields qualitative results for oxygen too, as more elaborate techniques (e.g., TEM-EELS) would be necessary to reliably quantify the local oxygen content.



	Sp1	Sp2	Sp3	Sp4	Sp5	Sp6
O [wt. %]	6.8	n.d.	11.3	5.5	4.7	n.d.
σ_O [wt. %]	0.3	n.d.	0.4	0.3	0.3	n.d.

Figure 5.11: The diffusion of oxygen in lc-uc during demagnetization is visualized by SEM. EDS measurements should only be interpreted for O detection and rough quantification. Hitachi FlexSEM 2500× BSE, 20 kV.

An exception for the detection of carbon are large spots with high carbon content, which can be spotted easily in backscattered electron imaging (BSE). This was observed in the treated ZnEp samples. Figure 5.12 shows the deep diffusion of C and O particles, possibly pieces of epoxy in grain boundaries infiltrated by a dark gray phase, identified as Zn-rich, replacing the Nd-rich phase. While the diffusion of Zn along the grain boundaries of NdFeB has been reported before [323], it is also seen that the infiltrating Zn-rich phase appears to act as a carrier to loose organic residues.



	Sp1	Sp2	Sp3		Sp1	Sp2	Sp3		Sp1	Sp2	Sp3
Zn [wt. %]	2.8	68.8	45.4	C [wt. %]	8.0	10.8	40.5	O [wt. %]	n.d.	n.d.	4.4
σ_{Zn} [wt. %]	0.6	1.6	1.4	σ_C [wt. %]	0.9	1.6	1.5	σ_O [wt. %]	n.d.	n.d.	0.6

Figure 5.12: The diffusion of Zn and epoxy residues is visualized by SEM. EDS measurements should only be interpreted as a qualitative analysis. Hitachi Flex SEM 2500× BSE, 20 kV for lc-ZnEp 0-lc-ZnEp 180. FEI NanoSEM 3500× BSE with Oxford Instruments Ultim Max 65.

With the increasing demagnetization time, the Zn-epoxy infiltration progresses deeper into the material and pushes apart the grains, which is documented by the SEM images seen in Figure 5.12. In the first hour, the infiltration depth is within several micrometers but reaches up to 150 µm depth at some spots after 240 min. Overall, the Zn layer appears to be disintegrated even though the demagnetization is carried out below the melting point of 419.5 °C [324].

5.3.3.3 Quantification of oxygen and carbon uptake

In a first screening test, the samples of each magnet type were exposed to the demagnetization treatment at 400 °C in air for 15 min, 30 min, 60 min, 120 min, 180 min and 240 min with one sample for each time and sample group. This test aimed to observe if there are differences in the oxygen and carbon uptake depending on the initial contamination and coating. The results are seen in Figure 5.13, Figure 5.14 and Figure 5.15.

To establish the maximum possible error within the constraint of limited numbers of samples, the most sensitive and most readily available samples (lc-uc) which also showed very consistent initial values were tested ten times per demagnetization hold time. The error bars of the lc-uc graph in Figure 5.13 represent the minimum and maximum O values measured for each time, giving an idea about the variance in oxygen levels caused by the demagnetization treatment.

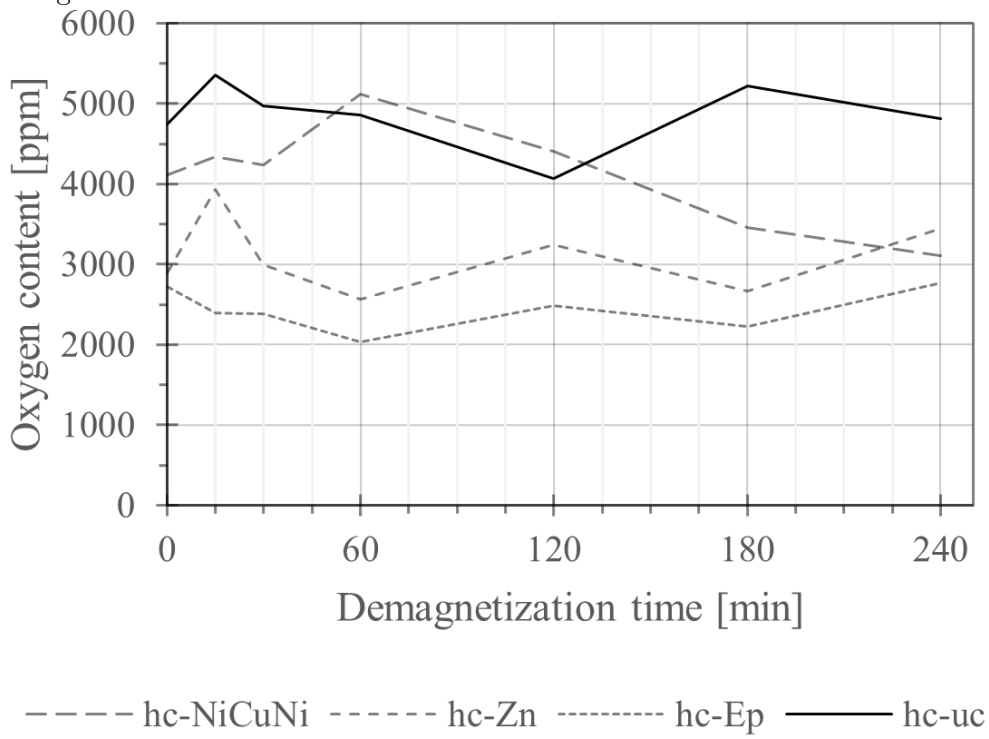


Figure 5.13: Development of the oxygen content of low contamination samples over demagnetization time. Measured with Leco ONH836. Single measurements were taken for all samples except for the most sensitive sample, lc-uc, which was measured ten times to illustrate the expected maximum range of oxygen values at each time point.

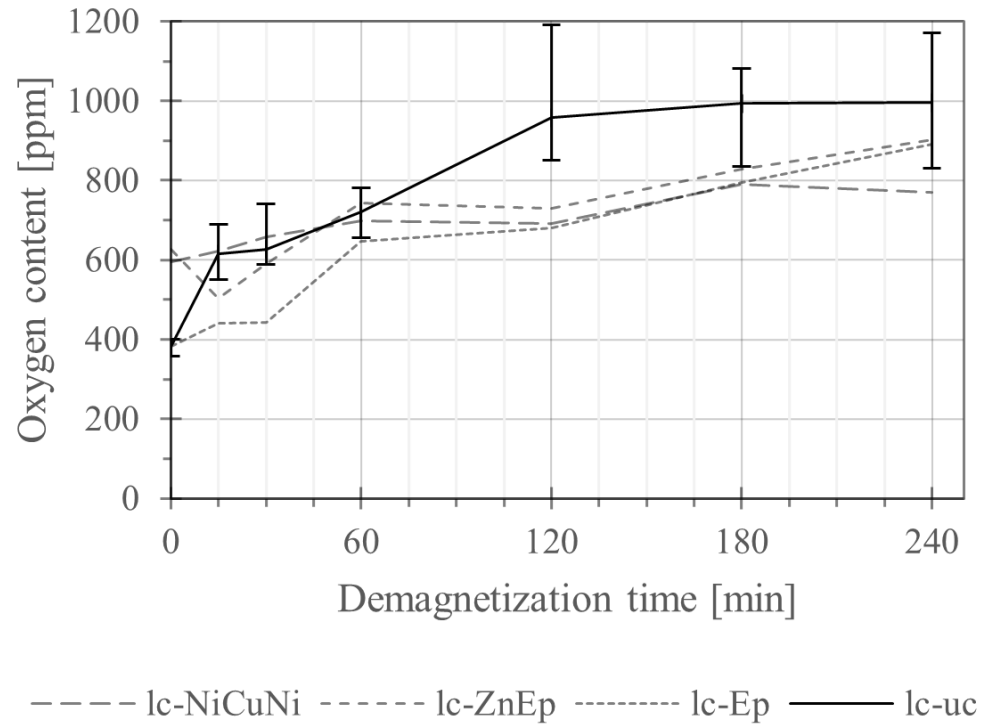


Figure 5.14: Development of the oxygen content of high contamination samples over demagnetization time. Measured with Leco ONH836.

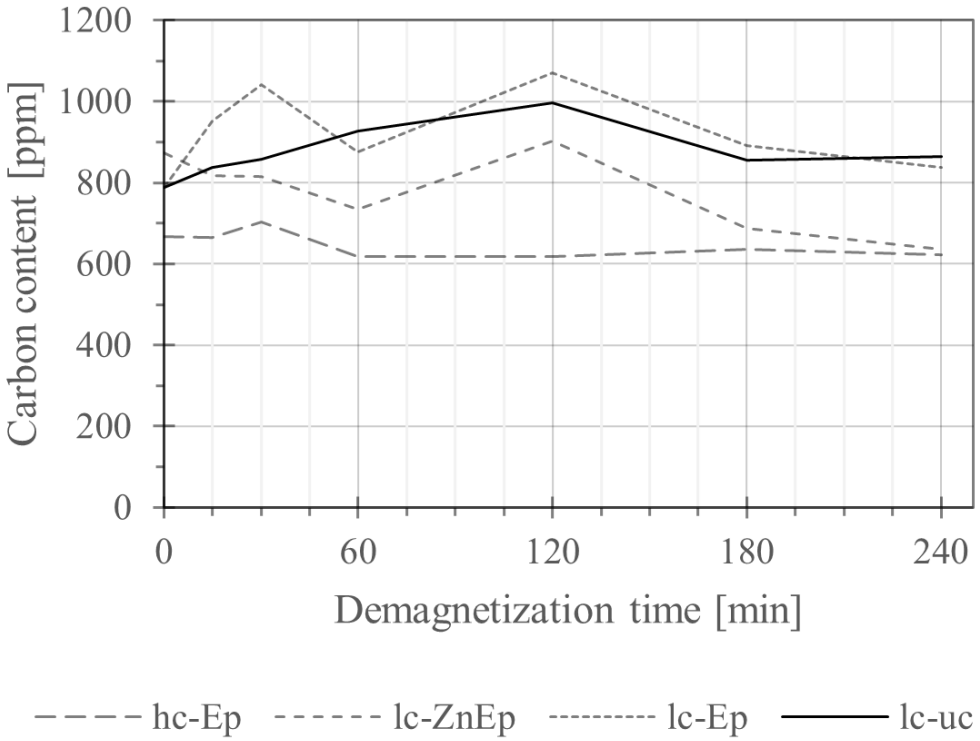


Figure 5.15: Development of the carbon content of carbon-relevant samples over demagnetization time. Measured with Leco CS744.

Figure 5.13 shows the development of the oxygen content over time for low contamination (lc) samples, defined as samples with an initial oxygen content below 1000 ppm. Here, the oxygen content of all thermally demagnetized samples increases over time.

As expected, the increase is highest for the uncoated lc-uc sample, where the oxygen content at 240 min increased by ~160 % compared to the initial value. After 120 min, there is barely any further uptake measured, likely due to saturation of the surface with oxides.

For the lc-Ep sample, the increase in this timeframe was ~130 %. Both samples had a very low starting oxygen content, which may have contributed to the substantial increase. For the lc-ZnEp, the increase was ~45 %, but ~80 % from the minimum value, as a dip in the curve is seen at 15 min, which is likely attributed to the considerable variation of values as seen in the error calculated for lc-uc. The lowest increase in oxygen content was seen in the lc-NiCuNi sample at ~30 %. This shows that while there is a tendency for the magnets to pick up oxygen, the uptake differs based on the used coating and its individual protectivity. It is further indicated that the saturation of the surface layers with oxides hinders further uptake.

Figure 5.14 shows the same measurements for high contamination samples (hc samples, defined as samples with an initial oxygen content above 1000 ppm) which do not show a tendency to increase. This is attributed to the high variance of initial values compared to the likely very low oxygen uptake due to the samples being mostly saturated with oxides already.

The change in carbon content is further monitored in Figure 5.15. Here, only the epoxy or Zn-epoxy-coated samples and, for comparison, the uncoated lc-uc sample are shown. The Zn- and Ni-Cu-Ni-coated samples are omitted as they do not contain carbon. Again, there is no apparent increase, which may be the consequence of the following: the initial carbon content of lc-ZnEp, lc-uc and lc-Ep falls within 800-850 ppm, which is relatively high considering that 1000 ppm is reported to be the approximate limit to the C content around which the coercivity drops dramatically [111, 213, 325]. Due to the high atomic mass of Nd compared to impurities like O and C, as well as the low amount of active Nd-rich phase in the overall magnet matrix, the effects of seemingly low contamination should not be underestimated.

For hc-Ep, while the initial carbon content is moderate at ~670 ppm, the O content is high at ~2720 ppm. Therefore, both saturation and a considerable variation in values may prevent significant carbon uptake.

Another important factor seems to be the usage of passivation underneath the contemporary epoxy coatings, which likely functions as a barrier against (carbon) diffusion.

In contrast to the presented results, moderate carbon uptake was measured in one specific sample analyzed in a screening experiment before this study, but it could not be appropriately repeated due to a lack of specimens of this sample type. What sets this sample apart was that it was an old, rather outdated magnet design featuring no passivation below the coating.

5.3.3.4 Reversibility of the contaminant uptake

The visible discoloration is shown Figure 5.10, as well as the SEM-EDS analysis of the surface areas are seen in Figure 5.11 and Figure 5.12. Figure 5.11 shows that the uptake of oxides is very localized in the close-to-surface regions of the magnet. The same is assumed for the hypothetical uptake of carbon. Therefore, experiments were conducted by taking (i) non-demagnetized samples, (ii) demagnetized samples and (iii) demagnetized samples that underwent sandblasting to remove all visible discoloration.
Sandblasting the darkened layer for 1 - 2 s per surface until no more changes in surface coloration are visible on average removes 10 - 25 µm of material from each side (measured

on Samples lc-uc). In Figure 5.16, it is seen that the O values were successfully lowered, approximately reaching the initial values.

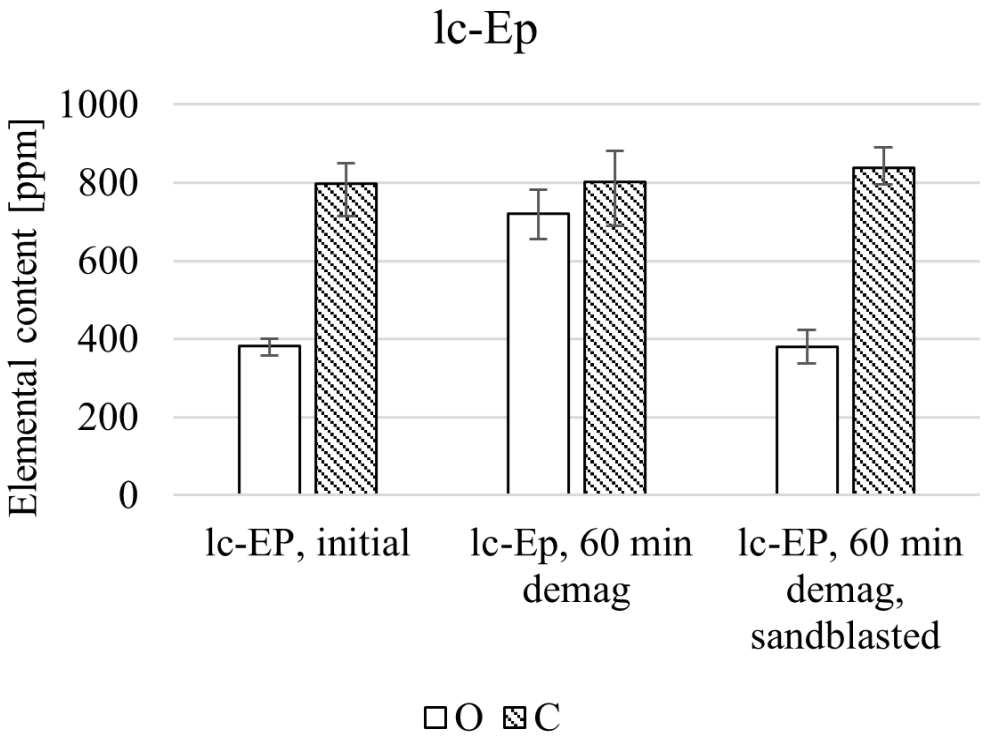


Figure 5.16: Comparison of the average oxygen and carbon content of lc-Ep before and after demagnetization as well as after the subsequent sandblasting of the darkened surface layer. Measured with Leco ONH836 and CS744.

The oxygen and carbon content could not be reduced below the initial values before demagnetization. This also applies to samples with very high initial oxygen content, such as hc-uc. As explained previously, these samples are assumed to be saturated with oxides and carbides. They appear to have oxides distributed throughout the material instead of only within a limited depth near the surface.

Therefore, the effect of thermal demagnetization on organic contamination can be successfully mitigated by ensuring that mechanical coating removal for all samples is carried out only after the thermal demagnetization process. This method only works in a limited depth and leads to some material loss.

Coating removal has to be carried out regardless; while Ni-based coatings can be removed from the powder [39, 190], Zn coatings, epoxy coatings, glues and oxide layers should be removed to avoid the contamination of the material to be resintered.

Zn residues evaporate below the sintering temperature, depositing on and damaging the equipment. Due to their high vapor pressure, high concentrations of Zn may further impair the density and integrity of the magnet.

Under certain conditions, the Zn coating can be removed, despite the infiltration caused by thermal demagnetization. As seen in Figure 5.12, the Zn infiltration depth increases over time and only reaches a few µm after 60 min but can go up to approximately 150 µm after 240 min. Therefore, a subsequent sandblasting treatment could remove the infiltration if the demagnetization time is kept as short as possible. Regarding Zn infiltration, monitoring the surface color alone does not facilitate the targeted removal of the Zn contamination, as it has similar optical characteristics to non-infiltrated NdFeB material.

The excessive carbon uptake from the remaining carbon-rich contamination will reduce coercivity by forming carbides, reducing the active Nd-rich grain boundary phase [53] and disproportionating the hard magnetic phase [111].

5.3.4 Conclusions

In this paper, the development of the oxygen and carbon content of samples with variable coatings and initial contamination levels during thermal demagnetization was tested, and it was trialed whether the contamination uptake can be mitigated. A general overview of the samples, sample designations and testing parameters is given in Table 5.2.

Table 5.2: Sample classification and designation as well as the testing parameters and condition of the tested samples summarized to facilitate the understanding of the report.

Sample classification	**Testing Parameters**	
Initial oxygen content <1000 ppm → low contamination (lc) >1000 ppm → higher contamination (hc)	Thermal demagnetization conditions Temperature: 400 °C Atmosphere: Air	
Coating/surface type	**Variation of the condition of tested samples**	
• Uncoated • Epoxy-coated (-Ep) • Zinc-epoxy-coated (-ZnEp) • Zinc-coated (-Zn) • Ni-Cu-Ni-coated (-NiCuNi)	Treatment • Untreated • Thermally demagnetized • Thermally demagnetized and sandblasted	Hold time • 15 min • 30 min • 60 min • 120 min • 180 min • 240 min

In the following recommendations for the magnets' design, the thermal demagnetization parameters and the integration of this process into the recycling process are given: Demagnetization times should be as short as possible to minimize the uptake of unwanted contaminants, including O, C and especially Zn. After 15 min at 400 °C, all magnets were fully demagnetized. Longer hold times may be necessary for larger magnets.

Generally, thermal demagnetization with the added objective to facilitate the removal of adhesive-fixed magnets or to split segmented magnets is better employed in air, as it allows faster and more complete decomposition [180]. While demagnetizing in a protective atmosphere may be beneficial to avoid oxygen uptake and preserve coatings, which would be helpful for reuse applications, it is not recommended for recycling. High costs for inert process gas and the required infrastructure to introduce it can be circumvented. The method's shortcomings, namely the possible contamination with O, C and Zn, can be mitigated by adjusting the order of processes and processing parameters or avoided using suitable coatings. The mechanical removal of coatings should be generalized for all samples without sorting and employed after thermal demagnetization, not before. This order is essential, as the O content increases strongly in uncoated samples.

The demagnetization temperature is best kept low but should be high enough to disintegrate all organic adhesives. Using a hold temperature of 400 °C proved to be sufficient and the adverse effects of the treatment were successfully reversed.

Among the tested coatings, Ni-Cu-Ni stands out as the most effective in protecting the magnets from oxidation during thermal demagnetization, as demonstrated in Figure 5.14. It showed the lowest relative oxygen uptake, consistent with Ni-Cu-Ni's excellent performance in corrosion tests like the salt spray test [53]. It functions well as a physical barrier to chemical and oxidative attacks. Overall, Ni-Cu-Ni, as well as other Ni-based coatings, are a promising choice for preserving the chemical properties of magnets during their lifetime as well as during separation for recycling.

Overall, thermal demagnetization in air as a preparative measure before recycling via HPMS and resintering, which is a recycling technique with a high sensitivity towards contamination, is viable. For less sensitive methods like hydrometallurgical recycling, where the material can be sufficiently purified as it is first leached and then the metals are extracted [326], it is also viable. It can be regarded as a simple and cost-effective procedure that can be carried out by any recycling facility owning at least a simple furnace operating at up to 400 °C. Due to the formation of hydrocarbon fumes during the decomposition of adhesive and organic coatings, suitable waste gas filtration or treatment must be employed. Due to widespread legislation to avoid air pollution, like the EU-wide Industrial Emissions and Livestock Rearing Emissions Directive (IED) [327] or the US Clean Air Act (CAA) [328], among others, such gas treatment facilities should be available at most recycling facilities.

Therefore, this simple method can be employed without reservation if the abovementioned conditions are met, especially for separating NdFeB from other materials and safely handling scraps.

In a possible, more advanced version of this process, mechanical coating removal could be aided by an optically controlled system to minimize material losses. A system like this could remove any surface layers with a different shine or color than the NdFeB alloy, removing coatings, passivation layers and unwanted oxide layers.

Furthermore, it is acknowledged that there is a lack of published research concerning the effects of contamination originating from various elements beyond those introduced by thermal demagnetization as presented in this paper. Only a few studies have investigated the influence of contaminants on pure rare earths [329], on the properties of NdFeB magnets specifically and on the structural incorporation or substitution of elements in NdFeB [77]. The research and review of these fields should be greatly encouraged.

Author Contributions

Conceptualization, L.G., F.B. and T.T.; methodology, L.G. and R.M.L.; validation, L.G. and T.T.; formal analysis, L.G.; investigation, R.M.L., L.G. and P.K.; data curation, R.M.L.; writing—original draft preparation, L.G.; writing—review and editing, F.B., R.M.L., P.K. and S.K.; visualization, L.G.; supervision, C.B. and S.K.; project administration, C.B.; funding acquisition, C.B. All authors have read and agreed to the published version of the manuscript.

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References

The references are integrated into the overall reference section of the dissertation to avoid duplication.

5.4 Publication III: Effects of Contamination on the Recyclability of NdFeB Permanent Magnets via Short-Loop Processing: Review of Common Contaminants and Study on Ni Coating Residues

The third major, peer reviewed study was conducted to investigate the impact of different contaminants qualitatively and quantitatively, using a combined approach of broadly reviewing literature on documented effects and extending this by an own, practical investigation where there was a lack of information.

The study is meant to complement the other studies and overall, give guidance to recyclers on the requirements towards their individual separation strategies.

The review and planning of the studies were conducted by the main author along with most of the execution of the trials. The methodology development was supported by Dr. Fabian Burkhardt and during the trials, assistance was received by Nicolas Moll. Considerations regarding carbon contamination were supported by Stefan Rathfelder, who is an expert on the shaping-debinding-sintering method, where carbon contaminations play a large role.

Supervision, funding acquisition and project administration were provided by the doctoral supervisors Prof. Dr. Carlo Burkhardt and Prof. Dr. Spomenka Kobe. All co-authors assisted in proofreading and review.

The paper was published on 29. October 2025 in the Journal Recycling under the DOI: 10.3390/recycling10060200. The manuscript along with review reports are available open-access.

The manuscript has been inserted in the following using identical text and figures; however, the formatting, section numbers, figure numbers, table numbers and reference numbers are adapted to be seamlessly integrated into the dissertation document. The references are integrated in the overall reference section. Please mind that the journal demanded that the Materials and Methods section must be integrated between the discussion and conclusions, which diverts from the prior two papers.

Effects of Contamination on the Recyclability of NdFeB Permanent Magnets via Short-Loop Processing: Review of Common Contaminants and Study on Ni Coating Residues

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Abstract

Short-loop recycling of NdFeB trades a reduced ecological burden for a higher sensitivity towards contamination, as the powder is usually further processed as-is. In this investigation, the known effects of common contaminants (O, C, Ni, Cu, and Zn) introduced due to product design choices, namely from coating material and adhesive residue or a lack of corrosion protection, are reviewed. This study focuses on the impact of such contaminants on the magnetic properties and microstructure of recycled magnets via HPMS and resintering. Because of the lack of information regarding the practical effects of metallic coating residues, the impact of Ni contamination on the properties of resintered NdFeB magnets is assessed. HPMS processed scrap powder is blended with Ni powder and recycled by sintering. It is found that Ni partially substitutes Fe in the φ-phase, as expected from the literature review, leading to detrimental effects on the coercivity and remanence. The formation of an α-(Fe, Ni) phase is observed. The acceptable limit of Ni contamination without detrimental effects is found to be around 0.25 wt.%; however, due to the substitution in the φ-phase, the contamination is irreversible via short-loop recycling and would accumulate over multiple lifecycles.

Keywords: NdFeB; rare earth permanent magnets; HPMS; short loop recycling; design for recycling; contamination; nickel coating; Ni-Cu-Ni

5.4.1 Introduction

The recycling of rare earth permanent magnets, especially NdFeB, has received widespread scientific attention, and consequently has also been addressed by different European Acts, prominently the Critical Raw Materials Act [6].
A suitable recycling method that offers direct recycling with reduced environmental impact is the hydrogen processing of magnetic scrap (HPMS) process, which was developed and patented by the University of Birmingham [44, 45, 47, 321]. This method, as well as different preparation and separation processes, has been researched in European Commission-funded projects and is on track to be implemented on an industrial level to promote sustainability and the resilience of the supply chain [330]. However, there is an open question remaining, which shall be addressed in this paper: What are the tolerable

levels of contamination of common contaminants from residual coatings, adhesives, and corrosion for recycling via HPMS and resintering? This especially includes some well-researched contaminating elements but also Ni, where prior evidence on the effects has been insufficient to judge the impact.

As HPMS employs direct recycling in a short loop, the possibilities to alter the chemical composition of the to-be-recycled magnets are rather limited, especially compared to traditional methods like hydrometallurgical and pyrometallurgical recycling where unwanted contaminants can be removed in the process. This necessitates the separation of possible contaminants prior to the production of the final recycled magnet. However, ideal separation may require extensive efforts and lower the economic viability of the recycling operation, whereas it is sensible to first determine how the properties of the magnet deteriorate in conjunction with the presence of the contaminants.

Possible contaminant sources are corrosion, residues from coatings, adhesives, and other adjacent materials like coolants and fillers, among others. The main contaminating elements researched in this paper are C, O, Ni, Cu, and Zn. The literature research is expanded by experiments regarding the effect of adding Ni to the material.

5.4.2 Literature review: effects of common contaminants on sintered NdFeB magnets

5.4.2.1 Carbon contamination

Carbon uptake can happen through various sources of contamination. A possible source is that polymer coatings are commonly applied for their protective and electrically isolating nature [108, 186], especially in high-speed electric machines, for example, electric traction motors. Furthermore, adhesives, used for segmenting and attaching magnets to the periphery, are usually based on polymer materials as well [16, 331]. Carbon-rich fillers used in both polymer coatings and adhesives, for example, carbon black or some types of fiber reinforcement, are also commonly employed. Carbon can also be introduced if the magnet-containing component includes any other polymeric features, which are not removed before the magnetic material is crushed or milled. Other potential sources are solvents, coolants, binders used in metal injection molding and powder extrusion molding, and other gaseous and liquid carbon-rich materials potentially coming into contact with the magnet material during its lifetime or recycling [332]. According to Hartwig et al. [213], the presence of solid carbon-rich impurities in the material during sintering seems more detrimental than the exposure to carbon-rich atmospheres at higher temperatures. In the following Figure 5.17, several studies where multiple carbon values are reported in conjunction with magnetic properties are compiled to illustrate the effect of carbon contamination.

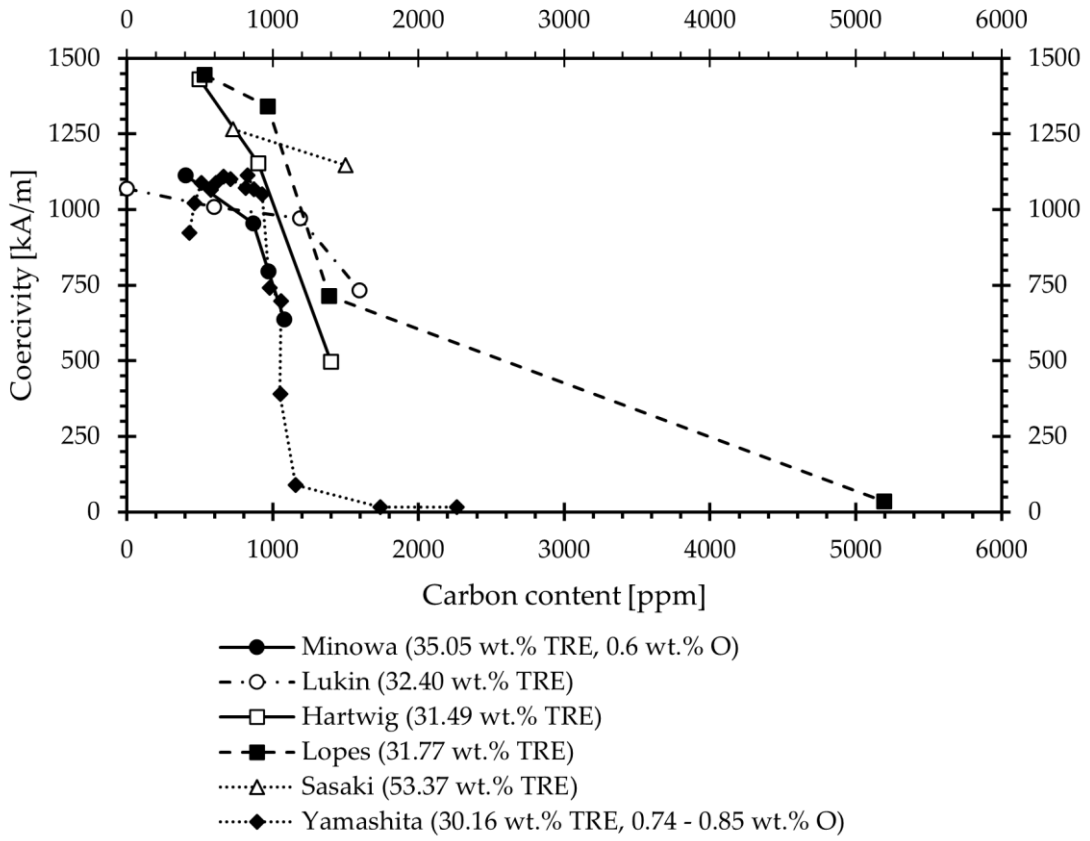


Figure 5.17: Data from different studies on sintered NdFeB magnets showing the coercivity in relation to the carbon content, including results from Minowa et al. [332], Hartwig et al. [213], Sasaki et al. [333] before and after annealing, Lukin and Szymura [334], Lopes et al. [111], and Yamashita [325]. Total rare earth contents (TRE) and some additional information has been added for better interpretation.

A high C content causes the formation of tetragonal and bcc-Nd carbides, accumulating in the triple junctions, which reduce the amount of active α-Nd in the grain boundaries and thus also the coercivity [110, 333]. Within the actual grain boundaries, no carbon is found [333]. C can partially substitute B in the hard magnetic $Nd_2Fe_{14}B$ phase, also referred to as the φ-phase [77, 110]. However, the relative amount of C found in the φ-phase compared to the triple points is minimal [333]. If C-rich residues are still present during sintering, where the amount of free Nd is greatly increased due to the solubility of Nd from the φ-phase in the Nd-rich phase, it can lead to disproportionation, as the Nd is bound in the form of carbides. This leads to the formation of α-Fe proportionally in accordance with the C content, greatly reducing coercivity [110, 111, 213]. It is also seen that densification does not properly take place in the presence of significant amounts of α-Fe [111, 213].

Overall, it is known that C is more sensitive to the magnetic properties than O and N [332]. Commonly, 0.1 wt.% is referred to as the limit of tolerable C content, which is supported by data from several studies, which have been compiled in Figure 5.17. Conversely to the reported detrimental effects, mostly regarding coercivity, Lukin and Szymura [334] report that the corrosion resistance, remanence, and the maximum energy product can be increased with carbon addition. The effects of carbon contamination on the remanence are less prominent and rather inconsistent across studies, including our own experiments.

It is evident that an elevated carbon content should be avoided by properly removing carbon-rich substances like the coating, which can, for example, be removed by grinding, sandblasting, or chemically from the bulk magnet, as shown recently by Dickinson-Lomas et al. [186].

5.4.2.2 Oxygen contamination

NdFeB is highly prone to corrosion due to a high electric potential difference between the matrix φ-phase and the intergranular Nd-rich phase, leading to the formation of corrosion batteries and intergranular corrosion [106]. Furthermore, rare earths like Nd are highly reactive and thus oxidize quickly [105]. Oxidation of powdered material is especially critical, as the alloy has a high affinity to oxidize [102], rendering the material potentially pyrophoric [115]. The effect of oxygen contamination on the coercivity is shown in Figure 5.18, where different studies that report oxygen values in conjunction with magnetic properties are compiled.

Oxidation of the Nd-rich phase can lead to the very common bcc-Nd_2O_3 or the less abundant fcc-NdO, as well as its polymorphs [335]. Furthermore, Nd hydroxides and other metalorganic corrosion products can form based on the media to which the magnet is exposed [336].

Like contamination with C, contamination with a critical level of O also results in a reduction in coercivity, due to hindering the formation of a continuous Nd-rich phase [102, 118]. With regard to the studies compiled in Figure 5.18, no clear tolerable limit that applies across alloy compositions can be determined. According to Zakotnik and Tudor [47], the presence of oxides inhibits excessive grain growth which speaks to possible benefits when having a moderate O content if there is no sufficient alloy addition to mitigate grain growth. Furthermore, Fukagawa et al. [337] report that cubic Nd oxides effectively suppress the nucleation of reverse domains, whereas a certain amount of oxygen would be beneficial for the development of high coercivity.

Furthermore, the presence of oxides can also affect processing during recycling, since when hydrogen processed, oxidized areas do not properly fall apart into fine, friable powder but larger aggregates [24]. These have also been shown not to mill properly, whereas they can be removed by sieving and processed separately [190], for example, to reduce the oxygen content by applying the Neoleach method [205] as an extension of HPMS, or in severe cases, long-loop hydrometallurgical [326] or pyrometallurgical [338] processing can be applied. When applying the Neoleach method or jet milling, the oxidized Nd-rich phase is removed and can be replaced by the addition of fresh NdH_x. When a thermal process introduces oxidation, for example, thermal demagnetization, a superficial oxide layer is formed which can be removed simply by grinding off the discolored surface [176].

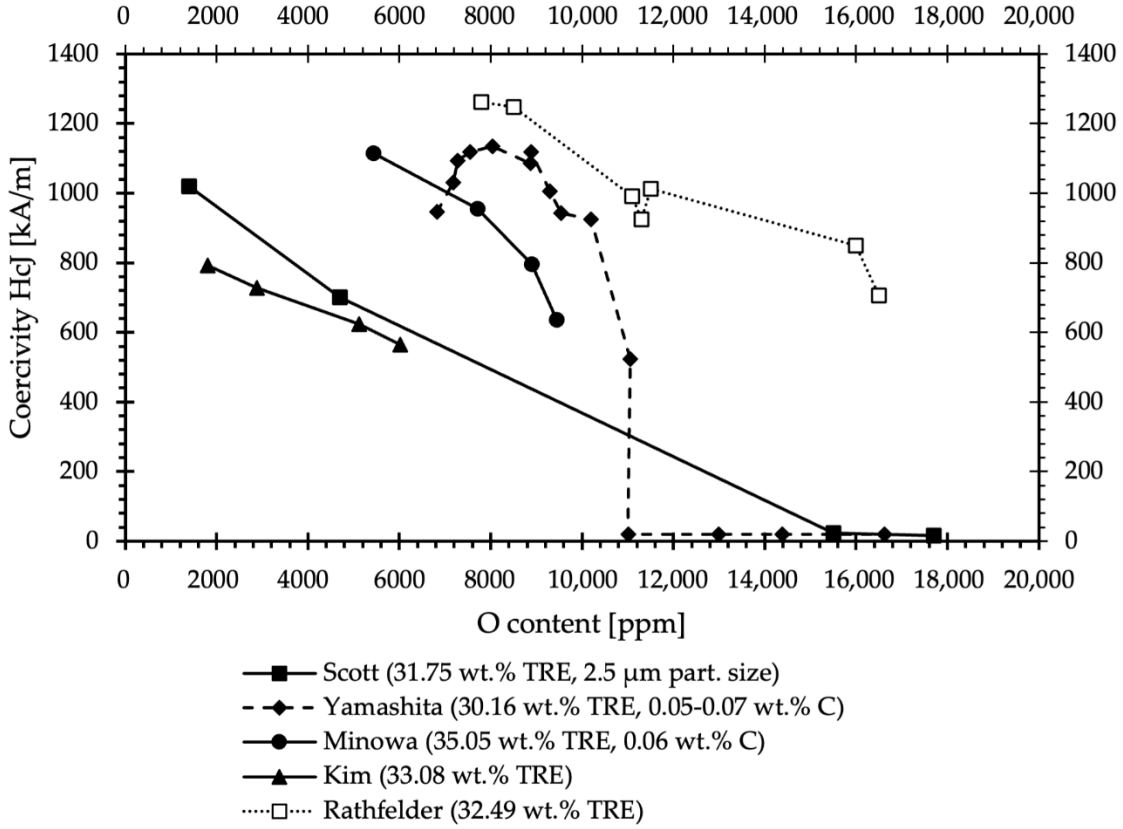


Figure 5.18: Data from different studies on sintered NdFeB magnets showing the coercivity in relation to the oxygen content, including results from Scott et al. [102], Yamashita [325], Minowa et al. [332], Kim [339], and extended data from Rathfelder et al. [215] (The data was provided by the authors of Rathfelder et al. [215]. The data was not explicitly published in the paper but was made available upon request. The measured samples were produced in an identical way as in the mentioned study). Total rare earth contents (TRE) and some additional information has been added for better interpretation.

5.4.2.3 Copper contamination

Copper uptake is possible from coatings, as Ni-Cu-Ni coatings are a popular, highly protective variant [308]. Similarly to Ni, Cu is rather ductile and can be removed alongside the Ni layers by milling and sieving of the powder; however, mechanical removal can be troublesome [190]. It is questionable, though, if Cu removal itself is necessary, as it is deliberately used as an alloying element in NdFeB magnets. Most NdFeB alloys contain some amount of copper. According to an internal analysis of ICP-OES data derived from 195 end of life NdFeB magnets, the magnets most commonly contain up to 0.2 wt.% Cu, as 165 of the 195 magnets fit into that range. Few samples contain more, with the highest amounts measured ranging around 1-3 wt.% of Cu.

According to Sagawa et al. [77], Cu has a very limited solubility in the φ-phase, being able to replace 0.2 of 14 Fe atoms. Further, Cu is soluble in the Nd-rich phase and Nd oxides, and it can form new phases. Different binary and ternary systems are reported for Nd, Cu, and Fe [340, 341].

It is well known that Cu can be added to improve the coercivity of permanent magnets; however, there is a trade-off of a lowered Curie temperature and remanence reported by

Pandian et al. [342]. Several research groups investigate grain boundary modification with Cu-rich alloys. For example, Wan et al. [343] introduced a Pr-Cu alloy via the dual alloy method into a NdFeB sintered magnet matrix, adding a total of 6.48 wt.% of Cu, improving the coercivity significantly, and reducing the remanence. Therefore, it seems that even a higher Cu contamination may be tolerable as the magnetic properties are changed but may be beneficial to some applications.

5.4.2.4 Zinc contamination

Zn uptake is possible from coatings, both the coatings of permanent magnets and some parts attached to them, as Zn is a popular metallic coating due to its anodic nature. Zn can generally be removed from bulk material by grinding or sandblasting; however, prior thermal demagnetization, which can be employed in conjunction with or prior to demounting, can lead to complications. Zn infiltrates the grain boundaries of the NdFeB magnet at elevated temperatures and extended times [176, 323], reaching deep into the magnet material. This can be avoided by keeping the demagnetization time relatively short [34]. Mechanical removal from the powder, like achieved with Ni-Cu-Ni coatings, was not viable in prior trials [53]. However, in preliminary tests that need further investigation, the Neoleach method [205] appeared promising to be able to remove Zn residues from the φ-phase grains along with rare earth oxides.

Regarding the effects of Zn addition on sintered NdFeB magnets, not much is known, likely due to Zn evaporating below the sintering temperature, at 907 °C, in atmospheric pressure [324]. Horikawa et al. [344] produced Zn-coated NdFeB powders made from milled sintered magnets by exposing the powders to Zn vapor at 550 °C, which led to the amorphization of the powder surface and formation of α-Fe, which differed from the effects of other coating elements like Yb and Mg. This decreased the magnetic properties of the powder, which were not resintered in the study [344]. However, in hot deformed magnets Zn can be integrated to improve the magnetic properties [320].

The evaporation of Zn during the process of sintering NdFeB powder should be seen as rather critical, as the equipment can be damaged by the deposition of a Zn layer and due to the high vapor pressure of Zn, which may cause structural damage to the brown part and a lack of densification. Preliminary tests were conducted, sintering samples with low Zn addition, which led to deposition of Zn on the inside of the sintering equipment, leading to the tests being discontinued immediately. The results of the test are shown in Appendix 5.4 A.

5.4.2.5 Nickel contamination

Ni can be introduced into the magnet matrix due to the widely used Ni and Ni-Cu-Ni coatings, as well as some more uncommon types with additional layers of Au or Sn [53]. In a prior study, it was found that the removal of Ni is possible by milling and sieving, as the coating flakes do not mill as finely as the magnet powder due to their ductility [190]. However, there are two caveats: First of all, some high-phosphate Ni layers are rather brittle and can mill as finely as the magnet powder, leading to a small residue (0.07 wt.% in the fraction below 25 µm) [190]. Secondly, with highly oxidized material, more magnet powder falls into the higher sieve fractions, where there is also more Ni present [190]. This leads to the question of how much Ni is tolerable, especially after multiple recycling procedures, and if Ni accumulates in the material.

Ni has a partial solubility in the Fe-sublattice of the $Nd_2Fe_{14}B$ elementary cell, substituting up to 1 out of 14 Fe atoms [77], which is equal to ~5.41 wt.% of Ni. The addition of Ni leads to an increase in the Curie temperature; however, it also results in a

steep decline of the magnetization, and consequently also the remanence [77]. Theoretically, several intermetallic compounds of Ni and Nd are also possible, potentially forming in the grain boundaries [345]. A study by Trujillo Hernández et al. [346] produced powders meant for exchange spring magnets via ball-milling and heat treatment with a $Nd_{16}(Fe_{76-x}\ Ni_x)B_8$ system, detecting $Nd_2Fe_{14}B$ (φ-phase), α-Fe, $Nd_{1.1}Fe_4B_4$, and $NdNi_2$. This shows both that there is a risk of introducing Ni at the milling stage aside from brittle Ni residues and that a NdNi phase is likely to be found. Furthermore, the study reaffirms the conclusion that the saturation magnetization is decreased and shows that Ni may preferentially replace Fe at the 16_{k2}, 8_{j2}, and 4_c sites of the φ-phase [346].

While there is currently no further data about the effects on the magnetic properties related to the nickel content available, it can be assumed that a higher Ni content will significantly deteriorate the magnetic properties.

Due to the relatively high thickness of Ni coatings, comparably high Ni levels can be introduced accidentally when the recycling procedure and/or the magnet design are unfavorable; thus, this has to be investigated further. In the following, the effects of Ni addition during NdFeB recycling via HPMS and resintering are investigated.

5.4.3 Results

After the preliminary experiments, screening with 0.025 wt.%-10 wt.% Ni, the relevant range to be tested extensively was determined.

Sintered compacts with a Ni addition of 0.25 wt.%, 0.5 wt.%, 0.75 wt.%, 1 wt.%, 2 wt.%, 3 wt.%, 4 wt.%, and 5 wt.% were produced along with reference samples without any Ni addition (0 wt.%). Four samples of each contamination level were produced; however, as some samples had to be excluded due to issues with the respective sintering run resulting in excessive oxidation or a lack of densification, some had to be repeated until the target number of acceptable samples was successfully produced. Oxygen levels after processing were found to be between 0.07 wt.% and 0.39 wt.%, with an average of 0.21 wt.% in the sintered (and partially annealed) parts for the acceptable samples, which were taken into account.

For all samples of the main tests, the density ranged between 91 % and 99 %, with no clear tendency to decrease or increase. In the preliminary test, the samples with 5 % and 10 % show a drop in density to 81 % in the case of a 5 % Ni sample and 67 % for the 10 % Ni sample. The density is given relative to the density measured in the original magnet, which was determined to be 7.59 g/cm^3.

The effects of Ni addition on coercivity and remanence are shown in the following 5.19 and Figure 5.20.

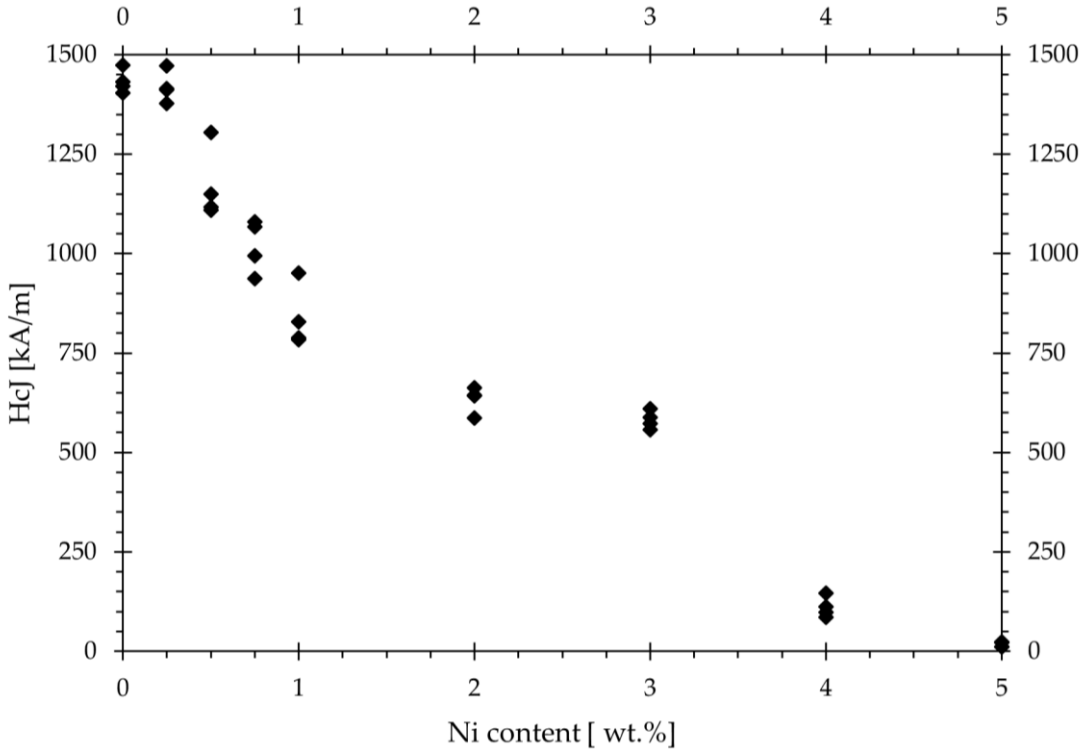


Figure 5.19: Influence of Ni contamination on the coercivity of sintered NdFeB compacts, with four samples per Ni content level, as sintered, measured with the Hystograph HG 200 at 23 °C. Measurement error is 0.4 % for the determined coercivity (internal test, n = 50).

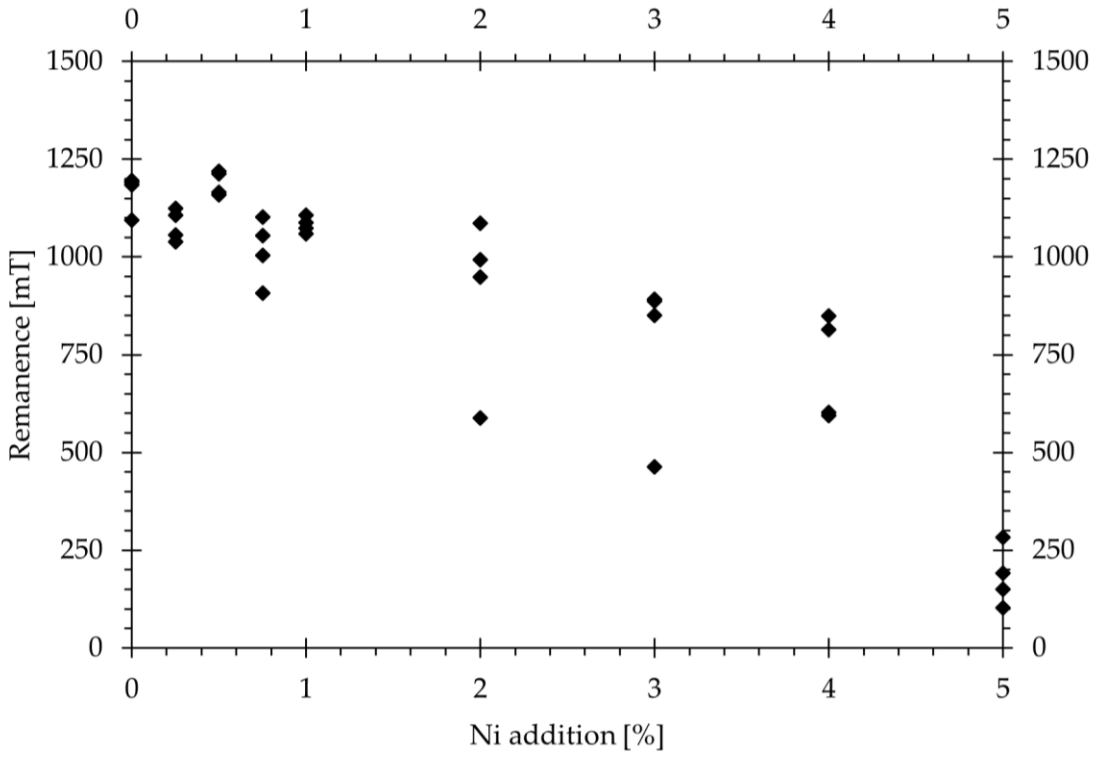


Figure 5.20: Influence of Ni contamination on the remanence of sintered NdFeB compacts, with four samples per Ni content level, as sintered, measured with the Hystograph HG 200 at 23 °C. Measurement error is 0.3 % for the determined remanence (internal test, n = 50).

The samples were investigated via SEM-EDS. The following Figure 5.21 shows the changes in the microstructure with increasing Ni content of 0 wt.% (Figure 5.21 A), 1 wt.% (Figure 5.21 B), 5 wt.% (Figure 5.21 C), and 10 wt.% (Figure 5.21 D). It can be noted that between 0 wt.% and 0.25 wt.% there are no tangible differences in microstructure observable in SEM and no Ni detectable in EDS; however, starting at 0.50 wt.%, some Ni can be measured by EDS.

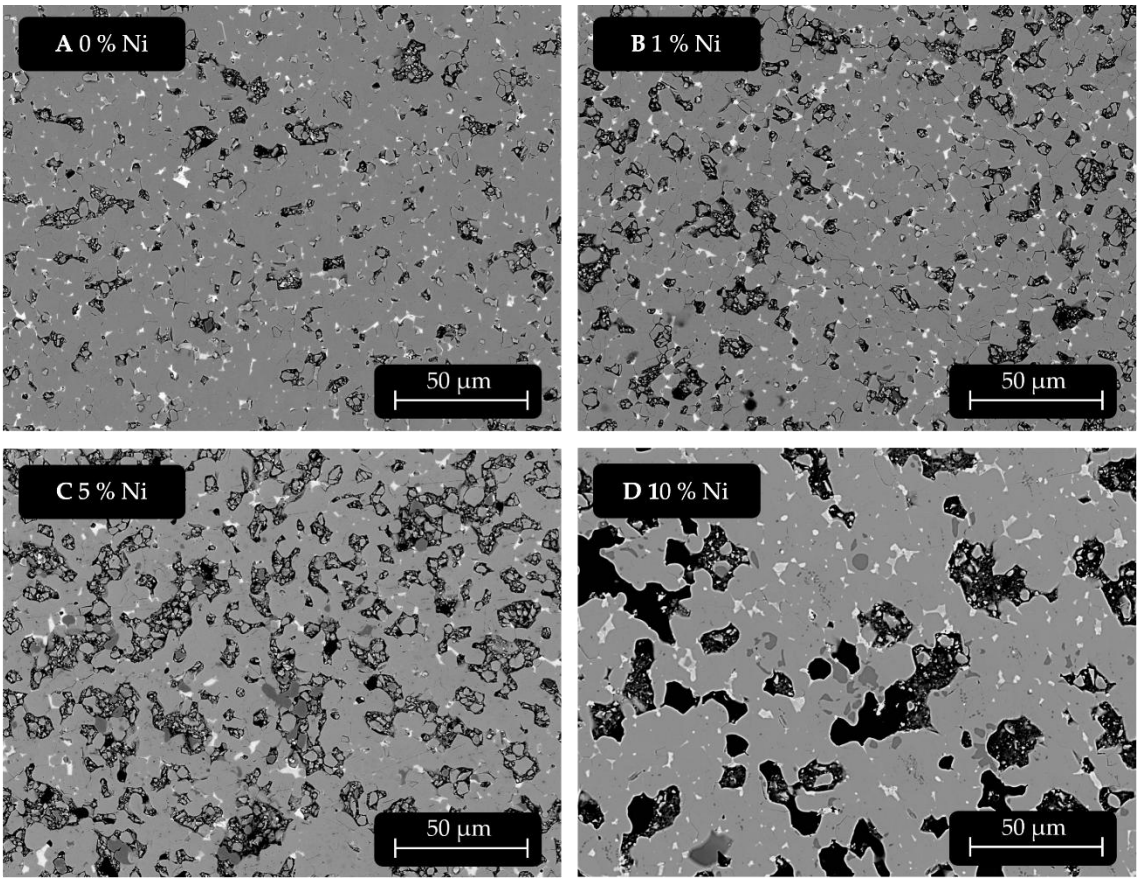


Figure 5.21: Effects of Ni addition on the microstructure: comparison of no addition (A) with 1 wt.% (B), 5 wt.% (C), and 10 wt.% (D). The sample with 10% (D) is from the preliminary experiments with the same parameters, but a different sintering furnace. Hitachi FlexSEM, 15 kV acceleration voltage, BSE-COMP mode, 500×.

In Figure 5.22 and Table 5.3, some exemplary EDS measurements are shown, where Figure 5.22 indicates the measured spots and Table 5.3 compiles the corresponding measured values in wt.%. The standard deviation is given in wt.% as well.

In Appendix 5.4 B, the microstructure of the original magnet and the recycled magnet without nickel addition are compared. Also, in Appendix 5.4 C, SE mode images are shown to support the observations in Figure 5.21 and Figure 5.22 regarding polishing defects and porosity.

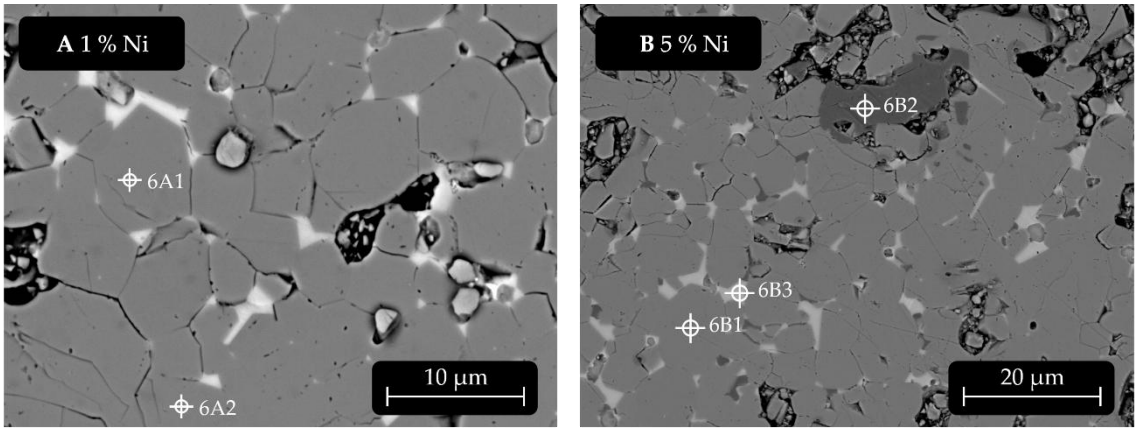


Figure 5.22: Energy-dispersive X-ray measurements on the different phases of samples with 1 % Ni content (A) and 5 % Ni (B) content, recorded with 20 kV acceleration voltage. The measurement spots are indicated, for which the measured elemental compositions are given in Table 1.

Table 5.3: Measured elemental compositions of the spots indicated in Figure 5.22.

Point	**6A1**		**6A2**		**6B1**		**6B2**		**6B3**	
Description	φ (1 %)		φ (1 %)		φ (5 %)		α-(Fe,Ni)		Nd-rich (5 %)	
Element	wt.%	σ	wt.%	σ	wt.%	σ	wt.%	σ	wt.%	σ
Fe	65.8	0.3	65.7	0.3	64.2	0.2	97.5	0.1	26.5	0.1
Nd	18.8	0.1	18.9	0.1	18.6	0.1			35.6	0.1
Pr	5.7	0.1	5.6	0.1	5.6	0.1			14.3	0.1
Dy	6.2	0.3	5.8	0.3	6.9	0.2			3.4	0.2
Ni	**1.3**	0.1	**1.2**	0.1	**4.7**	0.1	**2.5**	0.1	**17.7**	0.1
Co	2.3	0.1	2.4	0.1					1.1	0.1
Cu									1.5	0.1
Si			0.2	0.0						
Al			0.2	0.0						

5.4.4 Discussion

The coercivities measured for different Ni additions show that there is barely any effect from the addition of 0.25 wt.%.

At 0.5 wt.% and higher, the coercivities drop significantly, and at 5 wt.%, which is almost equal to the maximum substitution rate in the φ-phase, barely any measurable coercivity remains. The effect of the Ni additions on the remanence initially yields a fairly consistent remanence with additions of up to 1 wt.%, after which the remanence starts to drop consistently; however, it does not drop not quite as drastically as seen for coercivity.

When comparing the SEM images shown in Figure 5.21, it can be seen that the samples with 1 wt.% and 5 wt.% Ni addition are more affected by sample preparation. This may indicate that a brittle phase was formed or the integrity of the material is otherwise impacted. The sample with 10 wt.% Ni addition shows what appears to be a darker phase

forming, which looks similar to α-Fe as well as large cavities, reflected in a loss of density. Furthermore, grain growth can be observed. The dark grey phase α-(Fe, Ni) can be found increasingly with higher additions in samples with at least 3 % Ni addition. Below this level, no such phase was spotted. This phase is very similar to the typical α-Fe phase resulting from any kind of disproportionation of the φ-phase.

The measured Ni content in the φ-phase tends to be lower than the added relative amount, but close to the originally introduced level. Ni appears to substitute and displace Fe in the φ-phase, as expected based on the research literature. However, some amount of Ni is also found in the α-(Fe, Ni) phase. Additionally, a high Ni content can be measured in the Nd-rich phase.

The presence of a soft magnetic Ni-containing phase in the grain boundaries, as well as the formation of α-(Fe, Ni), explains the drastic loss of coercivity. The loss of remanence can be attributed to the uptake of Ni into the φ-phase, as elaborated in Section 5.4.2.5.

The variation in density is likely attributed to factors like oxygen uptake during recycling, excess Ni and Fe consuming active Nd and hindering re-densification, and possibly variation induced by the lab-scale equipment used for pressing and sintering. The variation in density most probably contributes significantly to the variance in the measured magnetic properties. However, the lack of density in some samples does not clearly correlate with the degree of variation observed.

It is known that, for example, excessive carbon contamination can lead to a lack of densification and formation of α-Fe in the NdFeB microstructure, which was reviewed in Section 5.4.2.1 in more detail, of which some may be reiterated at this point: The reason for this is that during sintering, the liquid phase dissolves Nd from the hard magnetic phase, which is able to bond with contaminants. The active liquid phase is thus reduced and the re-distribution of material required for successful densification is hampered. In the case of Ni, it is suspected that a similar mechanism leads to a decrease in density and higher porosity as a result: Ni replaces Fe and excess Fe is precipitated and forms α-Fe. Excess Fe and Ni should form a regular or Ni-rich φ -phase with any excess Nd and B, the latter usually being added with some excess to avoid the formation of α-Fe. Further, some direct binding of Nd by Ni is likely as well, as expanded on in Section 5.4.2.5 and supported by the EDS measurements in Table 5.3.

It is concluded that the most prominent effect is on the coercivity, and the acceptable limit of Ni contamination should be set at around 0.25 wt.%. However, it must be kept in mind that the contamination accumulates in the φ-phase and is therefore irreversible. This can pose a limitation to employing short-loop recycling on already recycled material (as opposed to the first-time recycling of magnets from virgin material), even while utilizing supporting techniques like Neoleach or jet milling, which can only reverse contamination that has accumulated in the grain boundary phase, not in the φ-phase.

5.4.5 Materials and methods

During preliminary experiments, excessive oxygen uptake during sample transport and equipment failure rendered some samples unusable which were therefore discarded; however, limited data of these trials was identified as acceptable and has been marked accordingly.

For the main experiments, production scrap magnets with exceptionally low oxygen contamination, excellent microstructure, and good consistency of magnetic properties were chosen and processed by removing the epoxy resin coatings via sandblasting, HPMS processing at 3 bar hydrogen and room temperature, partial degassing at 500 °C for 1 h, and ball milling to an average particle size Dv50 of 6.67 µm (measured dry with the laser

diffraction particle size analyzer Mastersizer 3000 (Malvern Panalytical Ltd., Almelo, the Netherlands)).

The experimentally determined chemical composition of the magnets used is shown in Table 5.4 and has been determined using the ICP-OES for metallic constituents. ICP samples are digested in aqua regia solution using a MARS6 microwave (CEM Corporation, Matthews, NC, USA), heating the digestive solution to 200 °C. The solution was then diluted with 5 % nitric acid and measured in an iCAP 7400 ICP-OES (Thermo Fisher Scientific Inc., Bremen, Germany) against a custom tuning solution. For organic constituents, the LECO ONH836 (LECO Corporation, St. Joseph, MI, USA) and LECO CS744 (LECO Corporation, St. Joseph, MI, USA) were used. After processing, oxygen levels were measured again.

Table 5.4: Chemical composition of the to-be-recycled magnet used, given in wt.% for each element, for an average of 3 samples.

Al	B	Co	Cu	Dy	Fe	Ga	Nd	Pr
0.24	0.95	2.45	0.16	3.48	67.23	0.16	20.28	6.35
± 0.003	± 0.003	± 0.010	± 0.001	± 0.019	± 0.205	± 0.006	± 0.070	± 0.037
Tb	Sn	P	Si	O	N	H	C	S
0.47	0.01	0.33	0.24	0.04	0.05	0.00	0.08	0.00
± 0.003	± 0.001	± 0.006	± 0.001	± 0.002	± 0.004	± 0.000	± 0.011	± 0.000
Total rare earth content: 30.58								

The recycled powder is modified by the addition of different amounts of nickel powder < 10 micron/99.9 % by chemPUR Feinchemikalien und Forschungsbedarf GmbH (Karlsruhe, Germany), mixing, and preparing pressed and sintered magnets. The addition levels shown in the Section 5.4.3 were chosen based on preliminary screening experiments to provide a more detailed overview in the more realistic lower-contamination range below 1 wt.% of Ni and a more broad overview in the higher-contamination range up to 5 wt.%.

The magnet powder was filled into a custom silicone mold and vacuum-sealed, aligned at 2000 V in a K-series pulse magnetizer from Magnet Physik Dr. Steingroever GmbH (Cologne, Germany), isostatically pressed at 10 t in a SJYP 40T manual CIP press by Xiamen Tmax Battery Equipments Limited, (Xiamen, China), and then sintered at 1100 °C, with a heating rate of 600 K/h up to 1030 °C and 200 K/h from 1030 °C to 1100 °C and a hold time of 30 min at 1100 °C. The sintering furnace is not externally cooled and slowly cools down to room temperature until the vacuum is reversed and the furnace tube is removed. A Nabertherm R 50/500/12 tube furnace with a C 550 Controller (Nabertherm GmbH, Lilienthal, Germany) with a custom Inconel 635 tube is used in conjunction with a vacuum system consisting of a Pfeiffer Vacuum HiScroll 6 (Asslar, Germany), and a Pfeiffer Vacuum HiCUBE TC 110 (Asslar, Germany) vacuum pump. The vacuum at sintering temperature is roughly $5 \times 10{-}5$ hPa.

Samples that did not properly densify or oxidize visibly and excessively due to leaks in the setup were excluded.

The produced samples were characterized with SEM-EDS using the Hitachi Flex-SEM 1000 II (Hitachi High-Tech, Tokyo, Japan) with a W-cathode and an Oxford Instruments micsF+ x-stream-2 EDS (Oxford Instruments, Wycombe, UK); underwent density measurement via the Archimedean principle, using the Sartorius Entris 124I-1S scale and the YDK density determination kit (Sartorius GmbH & Co. KG, Göttingen, Germany);

and their magnetic properties were recorded using the Hystograph HG 200 from Dr. Brockhaus Messtechnik GmbH & Co. KG (Lüdenscheid, Germany).

5.4.6 Conclusions

The effects of different contaminants have been reported in several studies and are reviewed in Section 5.4.2 of this paper. Compiling the results of different studies reaffirms the known acceptable limit of carbon contamination of around 0.1 wt.%. For oxygen contamination, the results are less conclusive, and it is assumed that the effects strongly interact with other microstructural and/or chemical parameters; however, in the case of oxygen, most studies do not show a distinct drop (with the exception of Yamashita [325]) but rather a linear decrease. Researching existing studies, a lack of data regarding the practical effects of contamination with metallic coating residues is found. It is likely that Zn does not remain in the magnet material after resintering. During preliminary trials, experiments involving Zn contamination were found to be impossible without significant damage to the sintering setup due to the evaporation of Zn. This is shortly elaborated on in the Appendix 5.4 A.

The effects of Ni contamination on short-loop recycled and resintered magnets have been researched methodically in this work, and Ni was found in the φ-phase, the Nd-rich phase, and the α-(Fe, Ni) phase. A tolerable limit of approximately 0.25 wt.% can be identified when the goal is to produce magnets that show almost identical properties to uncontaminated ones. Beyond that level, a nearly linear decline can be seen. Therefore, the acceptable limit can be increased, depending on the individual requirements. However, due to irreversible accumulation in the φ-phase, the long-term effects in multiple recycling cycles should not be underestimated. It is therefore recommended to recyclers to declare the Ni content of their recycled magnets if a full removal is not possible within technical and economic means. This allows future recyclers to choose the correct treatment option for Ni-contaminated scrap, as the contamination could be reversed with long-loop recycling or the material could be blended with Ni-free material to lower the total content.

Regarding the effects of different, deliberately added alloying elements and also the interaction of these with the contaminants reviewed and researched in this work, more scientific work is essential. Similarly, the influence of the metallic contaminants over multiple recycling cycles including non-short-loop routes is an important topic for future research as already achieved for organic contaminants by Zakotnik et al. [196]. Furthermore, it could be researched whether contaminants like Ni can be compensated for by the addition of other material, for example, as performed with the addition of fresh Nd to compensate for Nd bound by organic contaminants [196]. NdFeB-recycling-related research groups are highly encouraged to follow up and reproduce the presented results with different base magnets.

Author Contributions

Conceptualization, L.G. and F.B.; methodology, L.G. and F.B.; software, L.G.; validation, L.G., F.B. and N.M.; formal analysis, L.G.; investigation, L.G. and N.M.; resources, C.B. and S.K.; data curation, L.G. and S.R.; writing—original draft preparation, L.G.; writing—review and editing, F.B., N.M., S.R., S.K. and C.B.; visualization, L.G.; supervision, C.B., S.K. and F.B.; project administration, C.B. and S.K.; funding acquisition, C.B. All authors have read and agreed to the published version of the manuscript.

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Abbreviations
The following abbreviations are used in this manuscript:

BSE-COMP	Backscattered electron compositional imaging mode
CS	Carbon-sulfur analysis via hot gas extraction
EDS	Energy-dispersive x-ray spectroscopy
HPMS	Hydrogen processing of magnetic scrap
ICP-OES	Inductively coupled plasma optical emission spectroscopy
ONH	Oxygen-nitrogen-hydrogen analysis via hot gas extraction
SE	Secondary electron imaging mode
SEM	Scanning electron microscopy

Appendix 5.4 A. Effects of Zn contamination

No extensive study on the effects of Zn contamination could be performed due to the damage to the equipment which was evident even when processing pressed compacts with 0.025 – 0.075 % Zn addition. In the sintering run, ~0.1 g of Zn was added; however, the entire glass furnace tube used in the preliminary tests was metallized after the treatment, and the samples showed a high porosity. Figure 5.23 shows the metallized glass tube. Detrimental effects on the vacuum system are unfortunately expected to arise if similar experiments are carried out several times. Figure A2 shows the microstructure of the Zn-contaminated magnets. No Zn was found remaining in the sintered magnets in ICP-OES; however, due to the very low initial addition, this may be an error or it may be different with higher additions. The experiments were discontinued immediately, and we do not recommend replicating them without sufficient protection of the vacuum system.



Figure 5.23: Photograph of the glass sintering tube after sintering samples with a total Zn content of around 0.1 g.

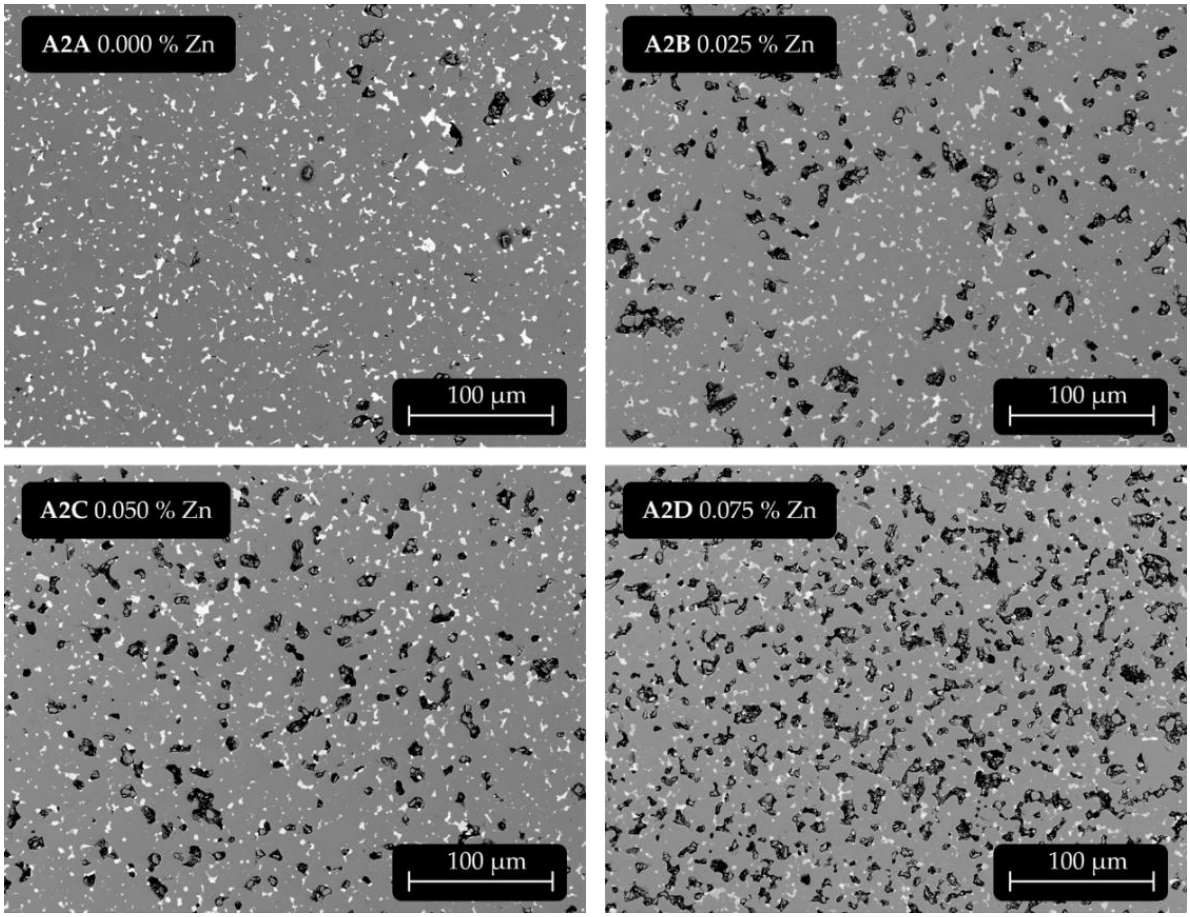


Figure 5.24: Porosity seen in samples with small Zn addition, image from the preliminary trials. Hitachi FlexSEM, 15 kV acceleration voltage, BSE-COMP mode, 200×.

Appendix 5.4 B. Microstructure of the original scrap magnet before recycling

For comparison, the microstructures of the magnet prior to recycling and the recycled magnet without Ni addition are shown next to each other.

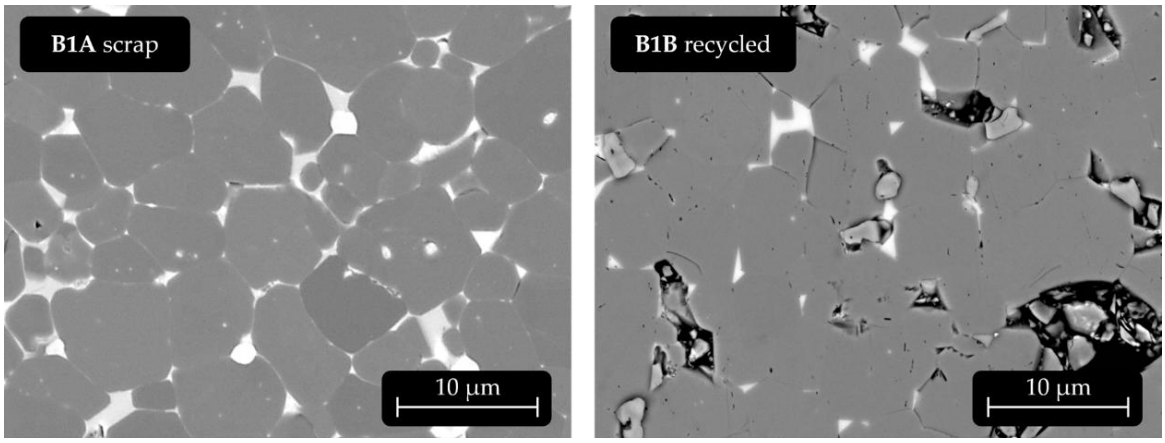


Figure 5.25: Comparison of the scrap magnet and the recycled magnet microstructure. The recycled magnet is without Ni addition. No NdHx addition or annealing. Hitachi FlexSEM, 15 kV acceleration voltage, BSE-COMP mode, 2500×.

Appendix 5.4 C. SE images of the microstructure

Supporting the evaluation of the microstructural topography, an SE image of recycled magnets without Ni contamination and with 3 wt.% Ni contamination are shown to complement the BSE images used throughout the publication.

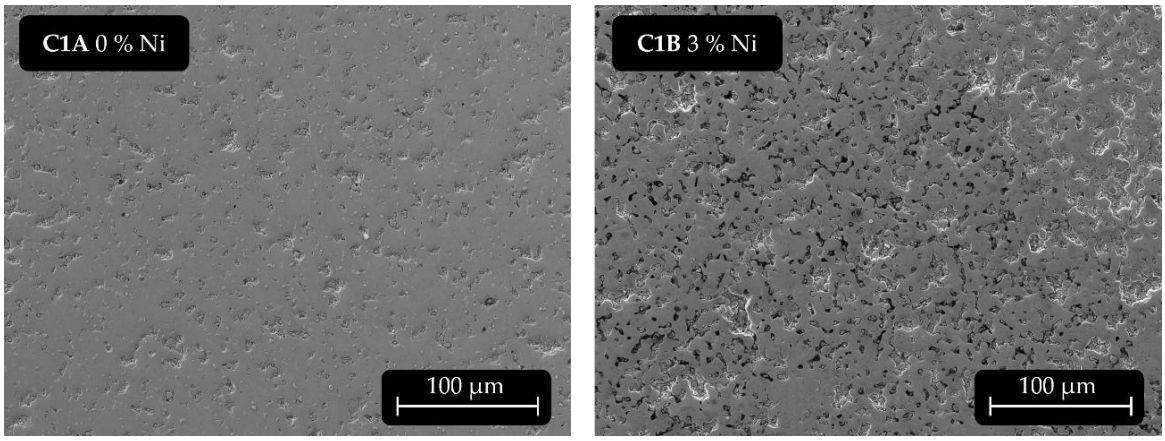


Figure 5.26: SE-images of recycled magnet containing 0 wt.% and 3 wt.% addition of Ni. Please note that the bright areas in C1B are caused by charge on the borders of defects and porosities. Hitachi FlexSEM, 15 kV acceleration voltage, SE mode, 2500×.

References

The references are integrated into the overall reference section of the dissertation to avoid duplication.

5.5 Minor Investigations

Extending the studies published and presented in chapters 5.2 - 5.4, some additional investigations were conducted, which may be published separately in the future more extensively. Here, a short overview about them will be given in the following Table 5.5.

Table 5.5: Minor investigations, study descriptions.

Study title	**Hydrogen permeability of surface conversions of NdFeB**
Study design and main results	Different passivated and phosphatized samples have been received, either with limited or no information on the conversion type. Some of the known types are Zn-phosphatized magnets, thermally passivated magnets, "anorganic coatings". The passivated magnets have been tested with regular HPMS parameters and mostly showed good permeability with an initiation time under 30 min and full decrepitation achieved below 60 min. However, some of the tested magnets took longer or did not decrepitate due to a lack of permeability. Furthermore, some magnets fell to a rather coarse powder.
Current limitations	As both visual cues and SEM-EDS analysis do not yield a clear idea of what passivation or phosphatization is present, for publishing, surface sensitive analysis should be conducted prior to publishing.
Purpose of the study	Giving guidance to designers how to choose a recycling-friendly passivation.

Study title	**Recycling-friendliness of segmentations**
Study design and main results	Block magnets, segmented magnets and snakeline magnets, both filled and unfilled, were received for testing. Snakeline magnets are not fully cut, rather, a meandering line is cut in, which can be left bare or filled. It was tested how each version performs in I) direct HPMS, II) thermal demagnetization and HPMS and III) thermal demagnetization followed by mechanical surface removal and finally, HPMS. With performance, in this case, the uptake of contamination is meant, which is mostly the uptake of C. It was found that block magnets and unfilled snakeline magnets performed best in scenario I) and II), followed by segmentation and then filled snakeline. This is due to the adhesive used in segmented magnets and filled snakeline magnets, whereas more adhesive is used in the filled snakeline due to a larger gap. In scenario III), all types of segmentations reached low enough C contamination levels.
Current limitations	The study should be extended with regard to fillers used as "placeholders" in segmented magnets, including glass beads, AlOx, woven mats and any other popular fillers. The removal of such fillers should be tested in jet milling and other methods.
Purpose of the study	Giving guidance to designers how to choose the most recycling-friendly variants of segmentations.

Continuation of Table 5.6: Minor investigations, study descriptions

Study title	Removal of epoxy resins from NdFeB powder.
Study design and main results	Epoxy coated magnets have been broken and processed in HPMS to test different methods to remove the epoxy residues from the powder. Tests included electrostatic separation and magnetic separation, which both failed as the resin stuck to the hard magnetic phase, whereas the particles coated with resin behaved essentially the same as the clean particles. Very few, small, purely resinous particles could be separated. Furthermore, tests with different solvents like acetone and toluene at varying temperatures were conducted, however, while with acetone, visible removal of epoxy from the particles was possible, the carbon content of the powder dramatically increased over 9000 ppm C due to reaction of the powder with the solvent.
Current limitations	So far, none of the methods worked.
Purpose of the study	Providing a way to remove epoxy resins from NdFeB powders for efficient pre-processing.

For each of the studies, some more research has to be conducted which has been limited either due to the availability of samples or testing methods. Readers are invited to reach out for collaboration or further information.

5.6 Summary of the Practical Investigations

5.6.1 Shortened HPMS recycling processes

Especially for smaller motors, design for recycling is very hard to implement, if disassembly should be targeted due the effort unlikely being viable for the low yield of material – their size, weight and choice of features is oftentimes highly restricted [208]. As shredding may lead to low yields with typical sorting technologies, some other options have been explored to greatly reduce the effort required for preprocessing of magnet containing components. It should be mentioned again here, that while typical sorting after shredding may be unsuitable for NdFeB containing scraps, that Ihne et al. [209] studied sorting mechanisms of shredded material containing different hard magnetic and soft magnetic components and propose a suitable workflow.

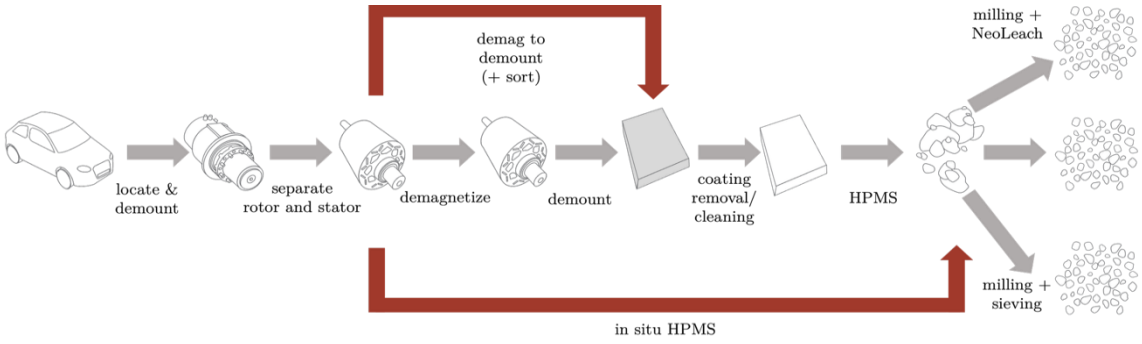


Figure 5.27: Shortened-loop processes and regular pre-processing overview.

5.6.1.1 „Demag to demount“

Designs that rely highly on adhesive fixation can be a good candidate for this method. Usually, adhesives would be seen as not being very recycling friendly, however, if the recycling of the adhesive itself can be neglected, it can be recognized that the disassembly can be highly simplified. For this method, the components are exposed to 400 °C in air atmosphere for ideally just 15 min if the component size allows it, which leads to nearly complete combustion of accessible adhesives. In cases, where the adhesives are deeply embedded, vibration, shaking or tumbling may be helpful. This method has also been identified as being useful for processing segmented magnets. Carbon diffusion into the material was only detected under two specific circumstances: The combination of a Zn coating with an epoxy coating or adhesive, combined with a long demagnetization time, or when no passivation is applied under the epoxy resin, which appears to be very uncommon.

However, in the latter case, the diffusion is limited in depth and can be removed by grinding off the surface. Oxygen diffusion happens regardless of a passivation and depends on the protectivity of the coating. However, also here, the contamination can be removed by removing the discolored surface in a mechanical process, like sandblasting in a drum.

A similar concept was developed by Hitachi for compressors specifically: The compressors are cut open, the rotor is removed and demagnetized and then dropped to remove the magnets by impact [347]. For more complex, low-yield products with several minor metals, pyrolytic disassembly is quite common outside of Europe [115]. Such treatment is carried out at 1500 – 1700 °C over longer hold times and therefore is very energy intensive [115].

5.6.1.2 In situ HPMS and separation from powder

In situ treatment of electric machines means taking the rotors, minimally preparing them and then treating them in HPMS without disassembling the magnets. Afterwards, the decrepitated powder can be removed from the large rotor parts and purified from some, but not all contaminants.

In situ treatment is suitable for designs where either no critical contaminants are applied or removable materials only. Ni-Cu-Ni and Ni coatings are removable from powder by sieving after milling, as the coatings deform instead of milling. Zn, organic coatings and adhesives cannot be removed with this method or other tested methods.

An interesting side effect found, which was also reported by Zakotnik et al. [24] is that excessive buildup of organic contamination leads to larger HPMS powder grains which are less friable and mill significantly worse. It is expected that this is due to already oxidized materials and carbides not hydrating properly and the oxides and carbides being less brittle. Due to that, specifically the oxygen content of the powder can be reduced if after milling and sieving, large particles left in fractions above 63 µm are not added back to the finely milled powder.

Some passivations are hydrogen permeable, other passivations and all coatings have to be scored to create hydrogen access. Another important factor is expansion space. If the reactor is static, enough expansion space must be given inside the magnet pockets. However, if the reactor tumbles the material, the requirements are likely more lenient.

5.6.2 Contamination Impact

Contamination can be introduced by residual materials from coatings, adhesives or even lubricants and coolants. The closer the material is to the magnet, the more likely a contamination by it will occur. Another source of contamination is corrosion. The most important contaminating materials are C, O, Zn, Ni and Cu.

Since separation was a major topic in the studies regarding the shortened recycling processes, which trade less effort for an enhanced risk of contamination, it is clear that separation procedures which offer an extremely high purity likely also require multiple steps, time, processing facilities and materials. Thus, it must be determined what level of contamination is viable.

By means of literature research, it was possible to combine information on the effects of O and C by compiling a number of systematic studies. In terms of Cu, most literature addresses the positive effects of adding Cu deliberately to the alloy. In terms of Zn and Ni, the information is limited. However, Zn effectively cannot be added to a regular sintered magnet as it evaporates before sintering temperature. While this appears to affect density negatively, it could not be further researched as even the smallest possible amount to reliably weigh in caused visible deposition of Zn vapour on the equipment, which poses a risk for the equipment.

In terms of Ni, a systematic study was conducted, where it was found that Ni appears in almost all phases of NdFeB and has detrimental effects both on the coercivity and remanence, as it substitutes Fe in the φ-phase and leads to Fe being ejected from the phase and forming α-Fe.

The presented study offers a brief, first overview but it should be complemented with more research. First of all, more potential contaminants should be factored in, e.g., filler materials in adhesives or passivation layers. Secondly, the accumulation of contaminants in the long-term over several recycling cycles should be studied closely to establish when a material should transition from the short-loop to a long-loop recycling process.

Based on the presented information and the literature research shown in Chapter 4, a thorough design for recycling strategy could be developed. The implementation of the compiled knowledge is addressed in the following Chapter 6.

Chapter 6

Implementation

6.1 Analysis of Motor Design Levels and Recommendations for Design for Recycling

Electric machines, including motors and generators have a few main components: the motor housing, shaft and bearings, which integrate the machine into the periphery, as well as the stator and rotor core, windings and in many cases, permanent magnets [12]. In most cases, there are also sensors and power electronics.

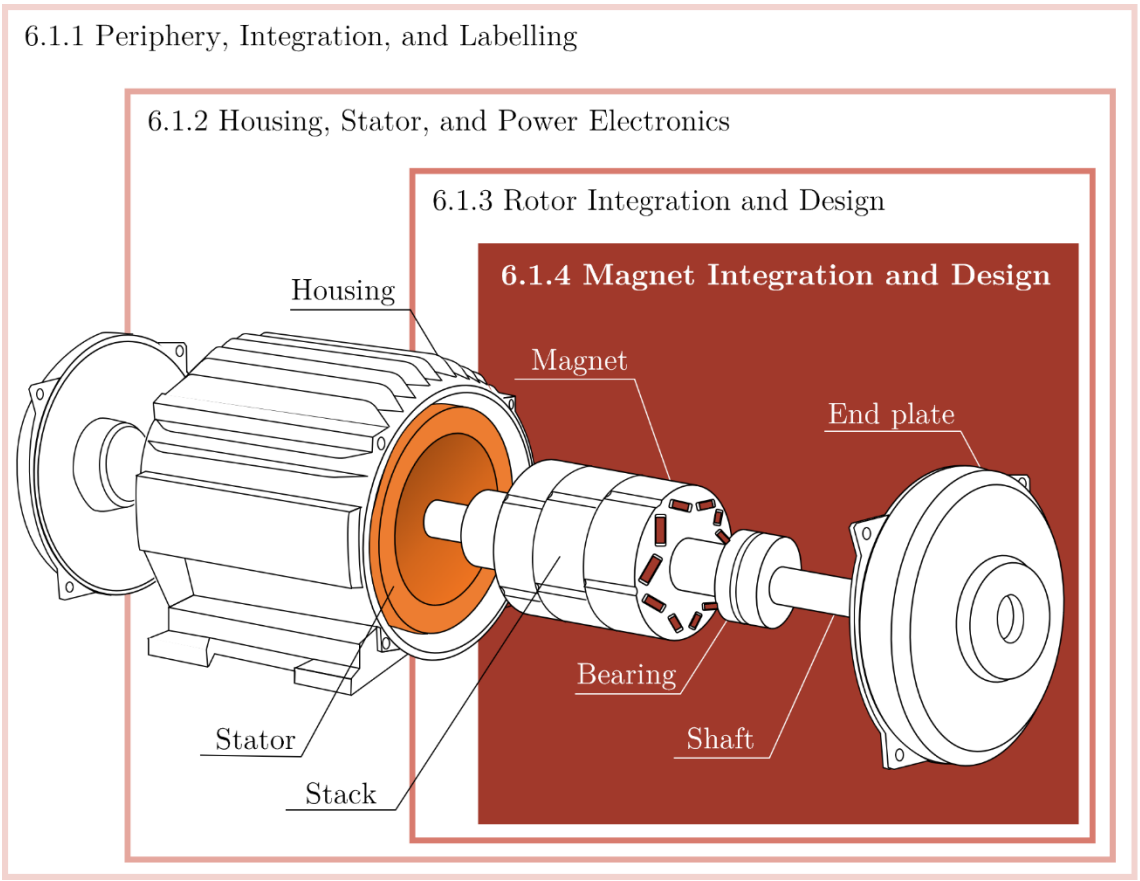


Figure 6.1: Overview image of product levels of an electric machine and sub-chapters related to them.

Some general recommendations can be made, especially regarding the choice of connectors and fixings. For example, while choosing non-permanent fixings appears to be a sound decision for improving recyclability, some applications do not allow their use.

For instance, using threaded fasteners like screws, nuts, and bolts is very straightforward, there is a lot of choices for designs, the potential for load bearing is high and the assembly is a simple, well controlled process if the design allows their application [208]. Other fixing methods without additional materials are possible as well, like snap fits and press fits. Snap fits use undercuts and protrusions, with at least one component being flexible [208]. When aligned, the flexible features can be temporarily deformed when they are joined and once in position, the position is fixed by locking with the flexible feature recovering its original alignment [208]. Such features, while typically irreversible, can be designed to be demounting-friendly while also offering a damping aspect and being rapidly assembled, however, they may not be robust enough for some applications [208]. Press fits are similarly simple to install and can be disassembled if designed properly, offering higher mechanical strength. Negative aspects are the required narrow tolerance control and limited adjustability.

In terms of fixations using additional materials, adhesive fixation turned out to be a conditionally recycling friendly option, at least if recycling the adhesive material is not a priority, as via the "demag to demount" method, they can be easily disassembled. However, for designs foreseen for in situ treatments, they would be highly detrimental.

The main advantage of adhesive fixations is that they are more universally usable than mechanical, removable features, which would need to be tailored to more specific applications.

Overall, however, the possibility for generalization is low. The recycling-friendliness of each feature depends on what preprocessing and recycling methods shall be applied. An overview of the typical and shortened paths was presented in Figure 5.27.

Section 6.1 is meant to give written guidance and explanation to machine designers, however, as a more straightforward and easier to interpret option, a tailored guideline is also offered in Section 6.2.

A systematic analysis of the design levels of IPMSM motors was already conducted and reported in a conference paper [348], however, the study was greatly extended and is no longer limited to IPMSM anymore.

6.1.1 Periphery, integration, and labelling

Good design for recycling for the housing and integration should be targeted mostly to allow good disassembly. Li et al. [349] offer an overview about the factors of "good" disassembly: The first set of factors are of technical nature; difficulty, tools required, damage to the product and safety [349]. The second set are economical, meaning the required time, human resource, equipment required and the value of the product [349]. Finally, also the environment must be considered, minding what waste, pollution and noise are created while disassembling [349].

6.1.1.1 Periphery and integration

For demounting the electric machines efficiently and safely, it should be ensured that any type of power supply can be disconnected swiftly and obviously. Furthermore, it is advisable to consider the ergonomy of demounting. If possible, electric machines should be kept as lightweight as possible with good graspability and accessibility from outside. In terms of graspability, both the proportion of the human body, typical tools and assisting robots should be considered.

Another important consideration is how many components must be removed before the electric machine can be removed as well. Ideally, only components, which also need to be disassembled to be fed into another recycling stream or for operations like decontamination should be removed.

6.1.1.2 Labelling

Labelling can improve the collection, pre-processing, logistics and recovery of materials by avoiding that they are brought into an unsuitable processing chain or processed in a suboptimal way [54]. It is highly advisable to label components which need to be recycled or treated in a specific way, which especially applies to NdFeB. Such labels should be concise and easy to interpret. If more detailed information is required, it should be deposited on a machine-readable data carrier. Such information could be regarding the specific chemical composition, location of components in complex products or the foreseen disassembly sequence.

The ideal location of labels can vary depending on which actor on the recycling chain needs what information. For example, in a complex product, for locating and demounting important components, the label should be placed in the "most likely expected place", for example, near the type plates. Alternatively, it could be made sure it is immediately visible when the product housing or, for example in case of a vehicle, the door or hood are opened.

If the information targets the disassembler of the electric machine, it should be placed on the machine housing. If it is to be used by the magnet recycler, it ideally should be on the magnet holding component or on the magnet. If the ideal positions are not viable due to cost or space constraints, it should be clarified how the information required can be transferred from one actor to another.

6.1.2 Housing, stator, and power electronics

While the housing, stator, and power electronics do not usually contain recyclable sintered NdFeB, they can be of importance for the recycling process due to a range of reasons.

6.1.2.1 Housing design

The housing is a component that serves several purposes: It holds the components safely, provides cooling if required and protects inner materials and components from outside influences. A more protective housing decreases the likelihood of corrosion of inside components, which may lower the requirement towards the protection of the individual magnets. A hermetically sealed housing may allow exchanging an epoxy-coated magnet for a passivated one, for example, as the protective function is shifted to the system level rather than the magnet itself.

Helland [208], for example, discusses the recyclability of epoxy encapsulated motors. This method offers excellent protection of the magnets as well as other benefits like low vibration and isolation of the magnetic field [211]. However, such housing design leads to highly difficult disassembly and limited repair options [211]. Therefore, they can overall be summarized as an unfortunate decision to take in the light of design for recycling, especially in perspective of the waste hierarchy, where repair should be clearly prioritized.

Another important aspect for eco design of the housing is cooling, as an effective cooling will allow to use less HRE, a lighter motor and less permanent magnet material altogether [240]. Common cooling strategies are air cooling with a fan or self-ventilation by the rotor, alternatively with a closed cooling loop inside the housing [350]. Liquid cooling is another possibility, which can be done with bespoke cooling channels in the stator or by immersing the rotor and stator fully or partially in oil. Overall, the ideal method is both effective (for

reference, see Li et al. [350]) and does not bring any corrosive or contaminating media in contact with the magnets. Therefore, cooling by immersing would require extensive cleaning before further processing.
Finally, disassembling the housing should be intuitive and ergonomically viable with common tools. The same rules as explained in section 4.4.2, regarding accessibility, tool choices and graspability can be applied. Suitable fixations should be used, ideally, reversible ones with a short disassembly time. Any means that could complicate typical disassembly methods, for example overmolding of screws, should be avoided.

6.1.2.2 Stator

The main aspect how the stator can influence magnet recyclability is when it is poorly cooled, as again then, measures to protect the magnets against demagnetization have to be employed at the cost of good recyclability and the ecological footprint due to heavy rare earth use.

Secondarily, stator materials could potentially contaminate the rotor. Ideally, the stator and rotor are separated early on to avoid this. The stator-rotor integration should therefore facilitate easy separation of both components.

Finally, skewing of the rotor could be potentially circumvented by skewing the stator instead, which would improve the recyclability of the rotor and the disassembly of the magnets, however, the applicability of this must be evaluated carefully.

6.1.2.3 Power electronics

Several electronics are coupled to an electric machine, which typically include rectifiers, DC-links, inverters and control electronics. While their influence on the recyclability is both complex and indirect, they still should be discussed briefly.

Power electronics influence aspects like the switching frequencies and torque control, which can influence the rotor losses due to eddy currents [351, 352]. When it is required to deal with high eddy currents, several non-recycling friendly features can be inevitable to integrate. Examples are segmentation of magnets and organic coatings. Additionally or alternatively, the magnet alloy may require higher heavy rare earth contents, which have a higher environmental footprint than light rare earths [353]. By employing grain boundary diffusion, the amount of HRE used can be limited, however, the core shell structure is not persistent over recycling as it dissipates during resintering.

Similarly, a poor inverter can induce vibration, potentially enhancing torque ripple [354]. Again, torque ripple has to be counteracted for some electric machines, like electronic power steering especially, by skewing the rotor, which again, is problematic for recycling s. Publication I [190].

While adapting power electronics is far out of the scope of this work, a general recommendation can be given: Any measures to decrease typical, unwanted phenomena like rotor losses and torque ripple will possibly also have a positive effect on the overall recyclability of the NdFeB magnets inside the rotor.

Furthermore, modern power electronics can enable the failure prediction for the electric machine and provide information about the state of the included permanent magnets. For example, monitoring the back-EMF and the d-current may indirectly reflect changes in the remanence of the permanent magnets [355]. The information could potentially be integrated into a digital twin, which can offer a prognosis about changes in magnet quality and the remaining lifetime of the motor components. However, overall, such options are more interesting for reuse and remanufacturing than recycling and likely only applicable for large, high-value electric machines.

6.1.3 Rotor design and integration

6.1.3.1 Rotor-shaft integration

The entire rotor component usually consists of one to multiple lamination stacks mounted on a shaft for torque transmission, balancing discs and bearings [146].

The rotor is often split into several stacks, which are lined up on the shaft. These stacks can be linear or skewed, meaning that they are not arranged with the magnets in line, but rather shifted by a few degrees, usually. Skewing can be done in multiple levels with varying skew angles, s. Figure 6.2.

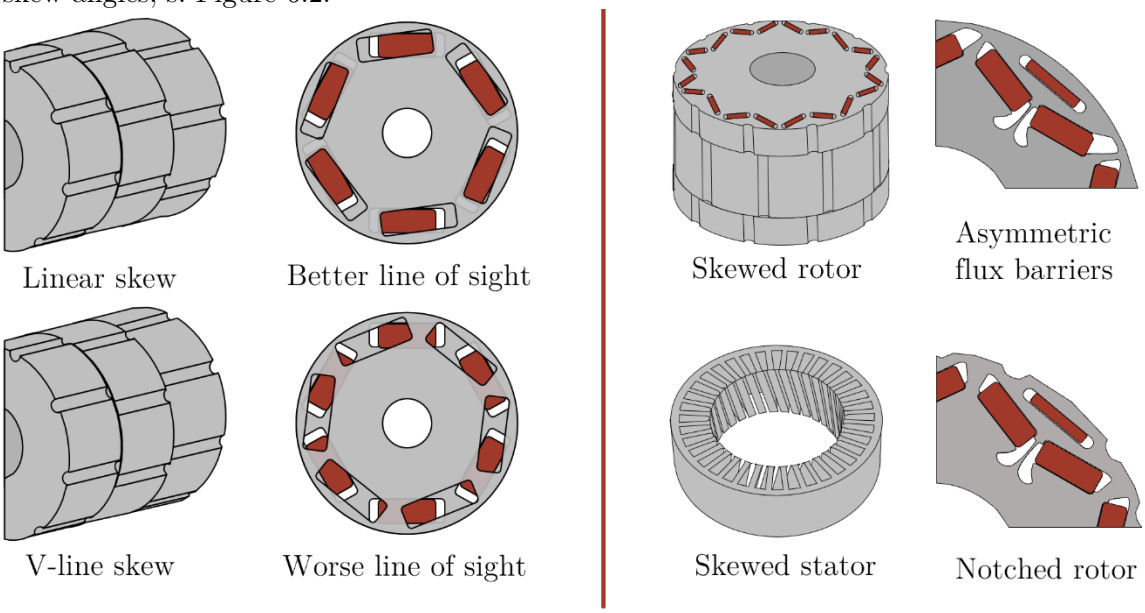


Figure 6.2: On the left, different skew arrangements are seen, as well the dependance of the line of sight on the skew angle. On the right, a skewed rotor is compared with other options to reduce torque ripple, like a skewed stator, asymmetric flux barriers and a notched rotor.

Skewing is employed to reduce torque ripple and noise [16]. From a viewpoint of recyclability, skewing should be avoided as it may cause extra steps for dismantling. For example, if the magnets should be extracted by pushing them out, the shaft has to be expressed first to separate the stacks, as else the magnets cannot be pushed through.

If skewing cannot be avoided at all, it should be considered to allow in situ recycling by ensuring enough expansion space, using a passivation as corrosion protection, avoiding glue and keeping the skew angles so the powder can exit the assembly, s. Publication I [190].

However, there are a few tactics to circumvent rotor skewing, as torque ripple and airgap harmonics can also be improved by other methods, for example by using asymmetric flux barriers, notches on the rotor surface, or stator skew [16]. Differently skewed rotors and alternatives to rotor skewing for reducing torque ripple are shown in Figure 6.2.

The mounting of the stacks on the shaft often is done by a press fit, sometimes the shaft is equipped with serrations to reduce the force to mount and demount, while maintaining high rotational fixation. This is helpful for demounting.

Another option to facilitate demounting is using a feather key or a serrated shaft (s. Figure 6.3) or some other way to retain the rotational position with a form fit, lowering the required pressing force. If the disassembly is foreseen to be done in conjunction with

thermal demagnetization, namely via “demag to demount”, the fixation of the shaft with adhesive is also a recycling friendly feature.

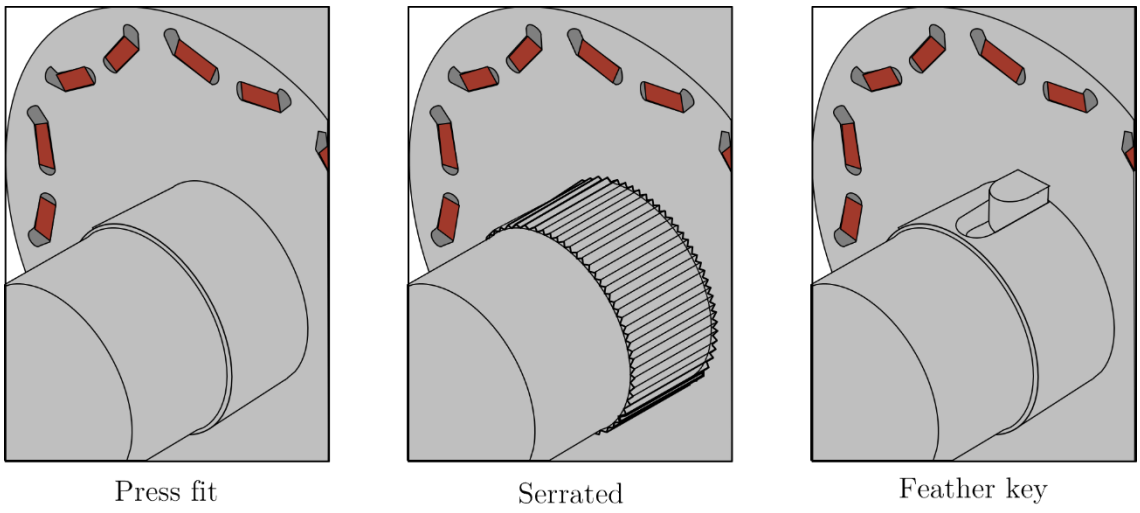


Figure 6.3: Rotor-shaft connections, recycling friendly form fit versions are for example using feather keys or serrated shafts to reduce the demounting force.

6.1.3.2 Rotor layout

Rotors are usually made of electric steel sheets, which are insulated from each other with some type of lacquer and connected by different stacking methods, for example interlocking, backlack (a baked adhesive), dotted glue (partially applied adhesive), lasering or clamping, as shown in Figure 6.4. The different stacking methods have distinct advantages and disadvantages. Backlack, for example, allows placement of the magnets very closely to the air gap due to the mechanical integrity of the stack, which can be beneficial for recycling via in situ treatment. However, the full application of the adhesive both increases the risk of it contaminating the material after demagnetization and may also hinder hydrogen access to the magnets.

Dotted glue allows better hydrogen access, but also less integrity. It can be seen as a middle ground between backlack and interlock methods.

Interlocking is a very convenient, cheap method, where the steel sheets are punch stacked. No adhesive is required; however, the steel sheets are generally coated to isolate them against each other. It is advisable to use a type of coating that does not disintegrate at demagnetization temperature. For this, anorganic coatings are preferable for better recyclability. If an anorganic coating is combined with interlocking, the contamination risk is minimal, while hydrogen accessibility is high, therefore this combination can be regarded as recycling-friendly in general. However, in a “demag to demount” situation, especially, when the shaft is also fixed with adhesive, backlack is a good option to promote full disassembly of all parts.

Overall, the suitability of the rotor stacking method depends on the foreseen recycling path.

IPM rotors
Internal permanent magnet rotors are the most common design used for electric traction motors and are generally widely used due to high power density, high torque density, wide speed range and efficiency [14, 146, 240]. The internal permanent magnets are placed in a pocket in the electric steel stack. This magnet pocket usually has so called flux barriers to direct the magnetic field, reduce the weight of the required permanent magnets, make use of reluctance torque and achieve a large constant power-speed range for field weakening operation [16]. A variant of IPM rotors is the PMSynRM rotor which utilizes reluctance with strongly elongated flux barriers [150]. Some common designs of IPM with different shapes of flux barriers were already shown in Figure 4.29. The most popular designs are reported to be single or double layer "v-shape", flat arrangement up to triple layered, or a mix of v- and flat shaped, which is also called delta-shape [16]. Generally, larger flux barriers which are not filled with adhesive, positively affect recyclability by different paths. Firstly, they provide expansion space for in situ treatments. Secondly, the weight reduction potentially allows shorter demagnetization time and thus, energy consumption. Sometimes, more cavities between the magnets and the shaft are used to reduce weight, improve cooling and the electromagnetic performance by improving reluctance and reducing torque ripple [16]. The same benefits apply here. Another factor regarding magnet recyclability is the use of layered magnet designs, like double or multi-layered "v-shape", "delta shape", or "fir tree" designs. Such designs make in situ treatments that rely on a cut-in from the side unviable and for disassembly, the more magnets need to be removed, the more time has to be allocated to the operation. Less, larger magnets are beneficial for disassembly. It should also be noted that breaking off the magnets is a risk due to high pressing forces during disassembly [146], which again, favors larger magnets with more balanced proportions.

The suitability of coatings will be discussed more in detail in the corresponding subsection, as it depends on a range of factors.

SPM rotors
Surface mounted permanent magnet rotors are inherently recycling-friendlier than IPM, as the magnets are directly accessible and can often be disassembled simply by removing the bandage, if there is any, and then demagnetizing thermally. Generally, outer rotor SPM need less fixation force and thus allow not using a bandage or a large amount of adhesive, which is why they are superior to inner rotor SPM from the perspective of recyclability.

The best feature combinations for SPM can be narrowed down to two options: First, combining fillerless adhesive fixation with a passivation or organic coating for the capability to apply "demag to demount". Applying a Ni or Ni-Cu-Ni coating is sensible as well, however, it is advisable to break the magnets and sieve the powder due to potential issues with removing Ni layers mechanically from the bulk magnet, see Publication I [190]. The second favorable feature combination is using a mechanical fixation method, for example, like the patented solution by Tucker [279], s. Figure 4.35 and a passivation or Ni / Ni-Cu-Ni coating for simple in situ treatment.

6.1.4 Magnet integration and design

The magnet integration and design are the most important set of factors for recyclability. The integration refers to how the magnet is affixed to the rotor, which is decisive for disassembly from the rotor and also the performance of the design in shortened processes. The magnet coating and segmentation, as well as the fixation determine the contamination of the end of life material. And finally, the magnet alloy and production method determine how the microstructure and magnetic properties evolve with recycling.

6.1.4.1 Magnet integration and fixation methods

Adhesive assemblies and inmolding

Generally, adhesive bonding is usually tailored to the materials which are to be bonded, as well as the bonding strength and other requirements. Such additional requirements could be related to processing parameters, like reduced curing times or specific curing conditions, or to the resistance of the resin to outer influences. Maintaining integrity over the entire usage temperature range and any relevant chemical exposure is required. While there is a range of adhesives available in general, epoxy adhesives are most commonly used to affix NdFeB in electric machines and usually the only option in traction motors [153]. Oftentimes, due to formation of eddy currents, fillers in the form of globular, fibrous or even woven materials including other organic substances, metals and ceramics are used to ensure electrical insulation over the whole magnet surface. To produce an adhesive fixture, the surface of the NdFeB magnet, which is usually either passivated or coated must be cleaned and prepared sufficiently to ensure adhesion. Surface roughness, integrity, polarity and charge are especially relevant. Then, the adhesive is applied to surfaces, and the parts are mounted for curing.

In-molding of rotor and stator parts is another possible fixation type which is commonly applied for smaller rotors. For this, the parts are put into a mold and poured over with epoxy. Like this, components are fixed, isolated and embedded in a way that avoids noise [208]. This makes it almost impossible to repair or replace parts and leads to challenging disassembly – any kind of heat treatment can lead to large amounts of fume. Overall, this method is not environmentally friendly and should be avoided.

Alternative assemblies

Press fits are also a possible fixation method, along with different holding features as presented in chapter 4.4.3. A method which is quite frequently seen is using tabs in some of the rotor cutouts which are then bent when the magnet is inserted, holding the magnet in place, see for example, the patent by Kim [241], also shown in Figure 4.35, panel I.

6.1.4.2 Magnet coatings

Several factors determine whether a coating is recycling friendly or not: Firstly, it should be separable from the magnet or any contamination it causes should not negatively impact the properties of the recycled material. Furthermore, it should be protective as organic contamination by C and O are especially critical for recycling. The potential of the material to corrode should not be underestimated – NdFeB is both very prone to corrosion, especially in humid environments [21, 104, 106] and also oftentimes used for applications with a long product lifetime. Highly corroded magnets are generally not considered to be economic for recycling [12]. For smaller consumer electronics, the product lifespan can be as little as 2-3 years [39], while highly relevant scrap sources with motors built in, like passenger vehicles, which are assumed to have a lifetime of 13 ± 3 years [34] and wind turbines, which are assumed to last 20 – 30 years [38, 39] may be exposed to the elements for a very long time if they are not sufficiently protected. While the magnetic properties of the original magnet are only somewhat effected by oxidation, magnets exposed to more corrosive media like acetic acid or salt water show a remanence loss over time, the magnet effectively disintegrating [108]. In the following, common surface protection methods are reviewed for their recyclability.

Passivations and phosphatizations
Passivations and phosphatizations are used both alone and under coatings. Phosphating works by immersing the parts in a bath of dilute phosphoric acid and usually dissolved metal ions, very commonly Zn-phosphate solution. The surface reacts with the bath and forms a protective conversion layer. For passivation, the parts are also immersed or sprayed with a passivation solution which can be a range of different materials. Classically, Cr (VI) solution was used for passivating but has been replaced by less toxic materials [356]. Examples for popular novel passivations are Ce passivations [104, 313] and tungstate passivations [314]. Here, the film forms mainly by adsorption and different reaction mechanisms on the surface like hydrolysis, complexing, etc. [357]. Phosphatization is very commonly used below other coatings. Generally, all large-series magnets coated with epoxy resins appear to have a passivation or phosphatization below the coating [176, 358]. Passivations and phosphatizations are especially recycling friendly due to their very low amount of added material, which is usually not detectable in SEM. This avoids risking significant contamination. Furthermore, some passivations and phosphatizations are hydrogen permeable. One drawback is the limited protectivity, which has to be supplemented by a suitable housing design for corrosive environments. Furthermore, to preserve the benefit of low contamination, the magnets should be affixed with no adhesive. If this is minded, passivated and phosphatized magnets are most recycling friendly.

Epoxy coatings
Epoxy coatings are reported to be very protective against chemical attacks [108], however, they can be osmotic to water, oxygen and ions, leading to sub-coating corrosion [359]. The isolative nature of epoxy coatings makes them well suited, or sometimes even the only choice for applications where the formation of eddy currents must be avoided, for example in high-speed electric machines. In electric traction motors, epoxy coatings are by far the most common choice observed, followed by passivations. A common modification of Epoxy coatings is the use of fillers. Fillers can be of ceramic nature, organic like carbon black, which is likely the most popular filler, polymer fiber and even woven mats, metallic particles, or glass beads [108, 176]. Some fillers like ceramic and organic particles are often used as "placeholders" to ensure full isolation [108, 186]. Others can improve properties like corrosion resistance and microhardness, like titanium nanoparticles [316, 358]. Epoxy coatings can usually be visually identified with ease, especially due to the widespread use of carbon black filler. When epoxy coatings are processed in HPMS, they fall apart and stick to the hard magnetic phase, making it very challenging to remove them. Epoxy residues definitely must be removed as the included amount of carbon is potentially highly critical to recycling due to the effects on the magnetic properties due to forming carbides, substitution within the φ-phase and causing the formation of α-Fe indirectly [111, 333]. Carbon-rich impurities like adhesives, binders and organics coatings being especially problematic compared to for example, carbon rich atmospheres [213]. The removal of epoxy resin from the powder has been trialed in preliminary experiments in this work, however, no method tested seemed promising: Solvolysis attempts lead to an enhanced carbon content due excessive carbon take-up, likely due to the high surface per volume ratio of the powder. It appears, solvolysis as tested and functional for bulk magnets [186] is not suitable for powder due to this. Removing the layer by milling also proved to be counterproductive due to smearing. Electrostatic separation and magnetic separation were also tried; however, it did not yield promising results as the particles behave similarly as merely a thin epoxy layer is attached to the hard magnetic grains.

At the current state, it is assumed that organic materials cannot be removed from powder very well. Thus, epoxy coatings are only viable from the perspective of recyclability, if the coating is removed prior to HPMS.

Zinc coatings

Zn coatings are anodic and provide sacrificial corrosion protection, meaning that coating integrity is less critical than for purely barrier-type coatings [305, 318]. This allows applying relatively thin layers, as the corrosion protection is maintained even in the presence of pores and defects. Furthermore, the coatings are popular due to being cheap.

Zn coatings can be modified with other elemental additions, like Ce, to further improve the protectivity [104, 360] or by "sealing" the Zn layer with a silane layer [318].

Zn can be removed mechanically or chemically from the bulk magnet [158, 190]. Regarding the removal from the powder, a mechanical removal appears challenging [190], a preliminary experiment for chemical removal with dilute citric acid within the Neoleach method [205] also showed promising first results, however, no systematic study on this was found or conducted yet. Therefore, similarly to epoxy coatings, it is assumed the Zn coatings must be removed prior to HPMS to avoid contamination. However, it was found that Zn infiltrates the grain boundaries [323], potentially also acting as a carrier for other contaminants [176]. If Zn coated magnets are thermally demagnetized, they must be treated for a short time only to avoid deep infiltration, for example, for 15 mins.

In hot deforming, small additions of Zn may improve the magnetic properties [320], in sintering however, Zn contamination, even at low levels must be avoided due to having the potential to both damage the sintering equipment and leading to a low density of the magnets due to high vapour pressure when it outgasses during sintering, s. Publication III.

Overall, Zn can be seen as even more critical than epoxy resin due to the low or limited compatibility with several recycling processes. Furthermore, the protectivity is subpar compared to other options [53].

Ni and Ni-Cu-Ni coatings

There are various options for Ni-based coatings of NdFeB magnets, including monolayered and multilayered Ni, as well as Ni-Cu-Ni, whereas the Ni coatings can have varying phosphate contents [304, 305]. Ni has a standard electrode potential of merely -0.25 V, being cathodic compared to NdFeB [305]. Therefore, the coating must be very dense and defect free [305]. Due to this, multilayered coatings are beneficial as they prevent crack propagation and offer a more thorough protection in the first place as well [306]. As Ni does have some chemical weaknesses, e.g. being sensitive towards sulfuric environments, an intermediate Cu layer is helpful [308]. Cu and Ni form a galvanic pairing with Ni acting as a sacrificial anode. The good protectivity leads to good performance in thermal demagnetization. When processed in HPMS, Ni coatings peel off the material and can be easily distinguished from the material [53]. It was reported in a study on the processing of hard disk drive magnets, which are usually all Ni or Ni-Cu-Ni coated, that Ni flakes can be sieved out by a rotating drum during HPMS [39]. The resulting powder can then again be sieved, removing fine Ni particles [39]. However, in several own studies on various Ni- and Ni-Cu-Ni coated scraps, it was found that without prior milling before sieving, only a very small yield of clean material, or a high contamination of Ni within the material can be achieved, as the overlap between the particle size of HPMS material and Ni flakes can be very large [190]. An adjusted workflow with a good yield was presented in Publication I [190]. High-phosphate Ni layers offer better mechanical properties and corrosion protection, but has to be applied in an acidic bath leading to sub coating corrosion, hydrogen generation below the coating and impairing adhesion [107, 305]. The porous structure further mills and contaminates the NdFeB powder [190].

Overall, the removal of Ni residue from powder is acceptable with the method, as in the worst case, the equivalent of up to 700 ppm of Ni was found in the relevant powder fractions (s. Publication I, [190]). Up to 2500 ppm, however, Ni residues do not lead to a tangible deterioration of the magnetic properties (s. Publication III, [361]). However, Ni

accumulates in the φ-phase, which is why a build up over several recycling cycles could still be critical.

Overall, due to the exceptional protectivity and compatibility of Ni-Cu-Ni and Ni coatings with almost all methods except some mechanical removal methods from the bulk magnet, which are hindered due to the thickness and ductility of the coatings, Ni and Ni-Cu-Ni can be categorized as rather recycling friendly coatings.

6.1.4.3 Magnet segmentation

Eddy current generation leads to rotor losses, as the permanent magnets heat up and the magnetic properties are impaired, as more closely described in section 4.3.3. With high-speed machines, eddy currents are especially problematic. A common and effective method to handle eddy currents is segmenting the magnet. The magnet can be split up into several segments either axially or radially, which are then re-attached using adhesive. For example, Tesla Model 3 is radially segmented [16].

From the perspective of recyclability, segmentation must be seen critically, as a carbon-rich material is brought into the material which cannot be simply removed from the outside of the magnet like a coating. The magnet would need to be broken apart. Generally, segmented magnets are regarded as problematic. However, it was found in the investigation of Publication II and the minor investigations, that with thermal demagnetization and impacting the magnets while still hot, the segmentation can be broken up and subsequently be removed.

If segmentation cannot be avoided, using fillers which are also thermally decomposed below 400 °C would be preferable, or using no fillers at all. Another interesting option is using so called “snakeline” magnets, where the volume in which the eddy currents can be generated is reduced by cutting into the magnet without fully splitting it into segments, like for example patented and commercially offered by the Swiss company BOMATEC [331]. While the eddy current reduction is slightly lower, the costs are lower as well and it is possible not to use adhesive to fill the cut in line. However, the benefit towards recyclability is dependent on whether the resulting gap is filled with adhesive for improved mechanical integrity or not, as both filled and unfilled variants are available [331].

With the “demag to demount” process, all variants – segmented, snakeline filled or unfilled can be dealt with s. Chapter 5.5 with the minor investigations. If the material should be HPMS treated without prior demagnetization, an unfilled variant should be used, or, if adhesive can be removed from the powder, at least an adhesive without filler should be used, which is more viable with a snakeline magnet than with a segmented magnet. Overall, especially unfilled snakeline magnets should be considered as a viable, recycling friendly alternative which can even be cost-saving.

Another alternative is replacing the isolative layer with one that is not directly attached to the magnet. This is following the proposition of Hernández-Millán et al. [224] to prioritize insulation materials which are easy to separate from the metals in a motor to improve recyclability. For example, thin, heat-resistant paper could be used or any kind of thin sheet material, for example also a woven net like it was already observed to be used as a placeholder in some segmented magnets. “Classical” segmentation, reported alternatives and a new proposition are shown in the following Figure 6.4.

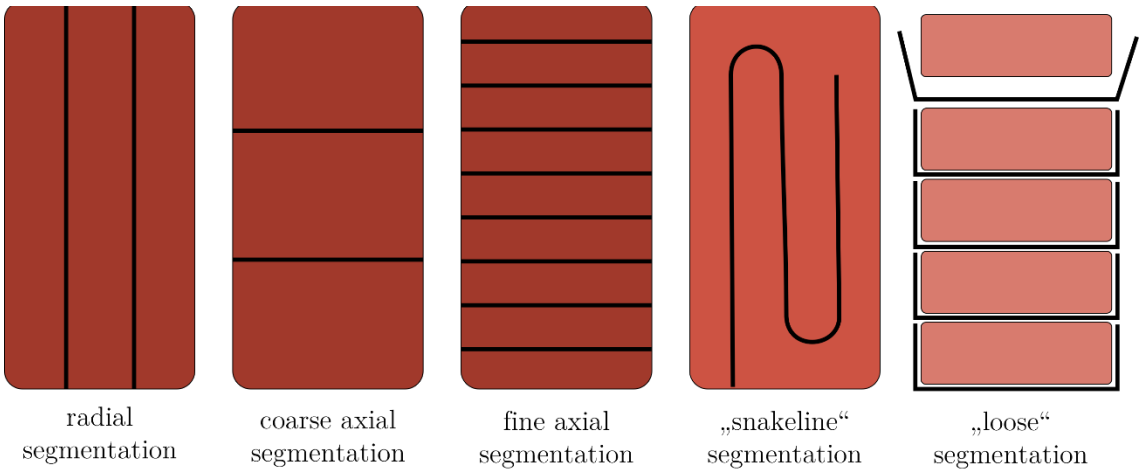


Figure 6.4: Methods to segment magnets, from left to right: Radial and coarse axial segmentation are commonly applied in EV traction motors. Fine axial segmentation is possible in EV traction motors as well, but more common in race cars, sports cars, e.g., Snakeline segmentation is a rather recent, more recycling friendly option. "Loose" segmentation is an own proposal for a recycling friendly option to producers, where thin sheets of material, for example heat resistant paper, can act as "spacers".

As for such concepts, technical adhesives and their processing, often using prolonged curing times, can be avoided, lowered production cost can be assumed. However, the need to adapt current assembly lines arises.

Some more options to reduce eddy currents are limiting the volume in which the eddy currents can be conducted as well, but at a smaller scale: For example, the thickness of the steel sheets in the rotor can be reduced [16]. Past designs have used laminations around 0.3 mm thickness [206], now 0.2 mm laminations are more popular [16]. Furthermore, reducing the grain size can also mitigate eddy current losses, as well as a reduction of conductivity of the grain boundary phase [247].

6.1.4.4 NdFeB magnet alloy

To design a suitable magnet alloy, the “starting material” needs to offer good intrinsic properties, namely high saturation magnetization, Curie temperature and magnetocrystalline anisotropy [39]. These intrinsic properties must be translated into functional extrinsic properties with a high remanent magnetization and high coercivity, which is achieved by appropriate processing and addition of alloying elements [39]. However, in the light of sustainability, there is some more requirements regarding the longevity and recyclability of the magnet material, primarily corrosion protection and the management of grain growth during resintering.

Alloying for improved corrosion protections usually targets the stabilization of the highly reactive Nd-rich grain boundary phase [105], reducing the reactivity and the potential difference between the Nd-rich phase and the φ-phase. Nonetheless, also the corrosion resistance of the φ-phase can be improved, as it is also susceptible to reacting with organic contaminants. In the following, commonly reported alloying elements are listed in Table 6.1 along with an explanation how they can affect recyclability directly or indirectly, as an alloying element or contaminant, and the respective references that claim the possible effects on the overall alloy.

It should be noted that all non-magnetic materials introduced, even if they are generally deliberately introduced, beneficial alloy additions will lower the magnetic properties proportionally. Furthermore, elements which can substitute for Fe or B may disturb the magnetic coupling in the φ-phase elementary cell, decreasing the magnetic properties as well. Effects from different techniques of introducing the alloying elements, like with grain boundary diffusion, are discussed separately in the next subsection. Another consideration should be that different alloys which are mixed for recycling could potentially negatively interact, however this field is rather difficult to explore due to many unreported alloying methods and a large variety of different alloys.

Table 6.1: Review of magnet alloy design for recycling.

Al	• Improves corrosion resistance [300]. • Improves sinterability [39]. • Impairs remanence due to solubility in the φ-phase, substituting Fe [77, 300]. In the φ-phase, 1.5 out of 14 Fe atoms can be substituted by Al [77], effectively erasing the remanence of the elementary cell, while also ejecting Fe atoms, which can form α-Fe [300]. Literature reports it to be exclusively found in the φ-phase [300] or that it is also occurs in the Nd-rich phase and Nd oxides [342]. *Note: it seems to be generally disputed whether Al is deliberately added or just an unwanted contamination from AlOx ceramic crucibles used in alloy production.*
C	• Impairs sinterability be forming tetragonal and bcc-Nd carbides, which accumulate at the triple junctions and deprive the grain boundaries of Nd, thus lowering the coercivity [110, 333]. • Impairs densification due to binding Nd and impairing the formation of a sufficient liquid phase [111, 213]. • Excessive C present during sintering leads to disproportionation of the φ-phase due to binding of Nd, leading to a proportional increase of α-Fe proportionally in accordance with the C content, greatly reducing coercivity [110, 111, 213]. • Can partially substitute B in the φ-phase, which leads to a decrease in saturation magnetization, anisotropy field and Curie temperature due to deformation of the elementary cell and possibly functioning as an electron donor to the 3d orbitals [77, 110]. However, the relative amount of C found in the φ-phase compared to the triple points is minimal [333]. • Magnetic properties are much more sensitive to carbon than to any other organic contaminant [333]. • Magnetic properties drop rapidly at approximately 1000 ppm C, s. Publication III [361]. • Usually introduced as a contaminant.
Ce	• Substitution of Nd with Ce is popular to reduce cost, as Ce is more abundant and cheaper. • Reduction of saturation magnetization and anisotropy field and consequently, remanence and coercivity, reduction of Curie temperature [77, 362, 363]. • Formation of a Fe_2Ce Laves phase is possible, negatively influencing the mechanical properties [363]. • Ce substitution can increase the corrosion resistance by increasing the surface potential of the grain boundary phase [364]. • Note: REE substitution can alter the reaction kinetics in HPMS. When processing magnets with high Ce content for a project, the material decrepitated well, but the HPMS aggregates were unusually soft. • Usually added deliberately, but can be seen as a contaminant.
Co	• Improves Curie temperature [39] and usage temperature [365]. The Curie temperature increases roughly 11 K per added 1 at.% Co [181]. • Improves corrosion resistance, 3.5 at.% of Co are recommended [299]. • Soluble in the φ-phase, as all 14 Fe atoms can be potentially substituted with Co [77]. • Co is reported to alter the reaction kinetics in HPMS processing (without further explanation how)[141]. • Usually added deliberately.
Cr	• Small additions inhibit corrosion [296]. • Can improve mechanical properties [366]. • Larger amounts impair the magnetic properties due to antiferromagnetic interactions when Cr is substituting Fe in the φ-phase [77]. • Usually added deliberately, can be introduced due to contamination with old passivation layers in very low quantities.

Continuation of Table 6.1: Review of magnet alloy design for recycling.

Cu	• Improves sinterability [39]. • Improves coercivity by enhancing grain boundary wetting, with the trade-off of lowered remanence due to addition of non-magnetic material [342]. • Has very limited solubility in the φ-phase [77], but is well soluble in the Nd-rich phase forming different intermetallic phases [340, 341]. • Usually added deliberately, can be introduced as a contaminant from Ni-Cu-Ni coatings.
Dy	• Improves intrinsic coercivity through a higher anisotropy field, allows a higher usage temperature, but decreases the remanence due to antiferromagnetic coupling [77, 81, 365]. • Significantly slows the hydrogen absorption of the alloy and requires longer HPMS initiation and processing times [49]. • Usually added deliberately, often via grain boundary diffusion to mitigate negative side effects and maximize the utility per added amount [24, 87].
Ga	• Increases coercivity by improved grain boundary wetting at 0.5-2 at.% of Ga [367]. • Increases corrosion resistance [298, 368]. • Usually added deliberately to modify the grain boundary phase.
Gd	• Improves Curie temperature, but decreases anisotropy field and saturation magnetization [39, 77, 177]. • Can be added deliberately or as a tramp element from the ore.
Ho	• Improves corrosion resistance [104]. • Improves the anisotropy field slightly, but decreases the saturation magnetization and the Curie temperature slightly [77]. • Added deliberately or as a tramp element from the ore.
La	• Similarity to Ce, La can be used as well to lower the cost of the alloy at the loss of the magnetic properties [362]. • Usually added deliberately, but can be seen as a contaminant.
Nb	• Can be used for grain refinement [369, 370]. • Improves corrosion resistance [368, 370]. • Alters reaction kinetics in hydrogen processing [371]. • Usually added deliberately.
Ni	• Can partially substitute Fe in the φ-phase [77]. Can form several soft magnetic phases in the grain boundaries [343]. • Can lead to formation of α-(Fe,Ni), as substituted Fe is ejected from the φ-phase (s. Publication III [361],[346]). • Leads to a loss of remanence and coercivity (s. Publication III [361]). • Acceptable limits appear to be up to 2500 ppm (s. Publication III [361]). • Most likely to be introduced by contamination with Ni-based coatings and not added deliberately.
O	• Material generally has a high affinity to oxidize [102]. • Forms bcc-Nd_2O_3 or the less abundant fcc-NdO, as well as its polymorphs [335] in the grain boundary phase. • reduction in coercivity, due to hindering the formation of a continuous Nd-rich phase [102, 118]. • remanence can be affected due to misalignment of φ-phase grains which are in direct contact with Nd_2O_3 grains [100]. • May inhibit excessive grain growth [47]. • Cubic Nd-oxides suppress nucleation of reverse domains [337]. • Oxidized Nd does not hydrogenate, leading to larger, HPMS aggregates that do not mill as well [24]. [190], • *Note: there is no clear tolerable limit and the influences of oxygen are complex, likely, some oxygen is beneficial, but too much affects the magnetic properties.*

Continuation of Table 6.1: Review of magnet alloy design for recycling.

P	• Inhibits corrosion [296]. *Note: May be added deliberately or due to procedures like phosphatization, cutting and grinding*
Si	• Inhibits corrosion [372]. • Offers grain refinement [372]. • Can partially substitute Fe in the φ-phase [77]. *Note: It is unclear, whether Si is added deliberately or introduced as a contaminant from processing, however it is found in most magnet alloys.*
Sn	• Inhibits corrosion [373]. • Strong detrimental effect on the magnetic properties [296]. • Can be added deliberately or introduced by uncommon coating types with Sn layers (s. [53]).
Tb	• Strongly increases the anisotropy field and usage temperature [77]. • Less trade-off in terms of the saturation magnetization compared to Dy addition [77]. • Usually added deliberately, oftentimes using a grain boundary diffusion process.
Ti	• Inhibits corrosion [296]. • Increases mechanical strength [366]. • Can potentially improve the microstructure when combined with Ga [374]. • Usually introduced deliberately.
V	• Increases corrosion resistance and mechanical strength [366]. • Can partially substitute Fe in the φ-phase [77]. • Usually introduced deliberately.
Zn	• Evaporates below the sintering temperature [324]. • Can lead to lack of density due to high vapour pressure (s. Publication III [361]). • Can lead to damaged equipment due to Zn vapour deposition (s. Publication III [361]). • Usually introduced as a contaminant from coatings but does not stay in the sintered magnet after sintering. Infiltrates grain boundaries [323], thus can also persist after superficial coating removal.
Zr	• Inhibits corrosion [296, 367, 375]. • Different accords: Some authors claim a reduction coercivity when added to NdFeB [296, 367, 375], others an improvement due to grain refinement [376]. • Usually introduced deliberately. *Note: Based on the different accords, it is likely that the effects greatly depend on processing parameters and limiting the concentrations.*

6.1.4.5 Magnet processing

With a suitable alloy, a good magnet can be produced, this applies both for primary production and recycling. Aside from the end of life magnets being well protected from corrosion and including beneficial alloying elements like grain refiners, it is also helpful to receive magnets with a good initial microstructure and evenly distributed, small grains, which can be achieved by proper processing. Besides this method, sometimes, specific methods to modify the magnet microstructure are employed.

Grain boundary diffusion, for example, should be seen critical from the viewpoint of recycling, as it interferes with recycling via HPMS and resintering [377]. Fine particles are thought to be formed from the high-HRE shells, which could lead to excessive grain growth in resintering. Furthermore, formerly grain boundary diffused magnets which show a distinct core-shell structure before recycling do not show the same after resintering [378]. Therefore, it should be considered that this material is basically wasted by distributing it rather evenly when resintering. However, at this point it should also be considered that grain boundary diffusion allows the improvement of recycled material beyond the original properties of the end of life magnet [24], leading to the need to evaluate whether improving the quality of the recycled material to a competitive grade is worth impacting the future recyclability of it.

Grain boundary modification, where the “normal” Nd-rich phase is exchanged for another phase also should be seen critical. Unless the replacement phase is especially

developed or at least tested for its suitability in hydrogen processing, the recyclability is likely impaired as it relies on rare earths reacting with hydrogen.

A last consideration in recycling-friendly magnet production is the size and shape of the magnets. Larger magnets with balanced proportions have less surface exposed to oxidation or other contaminants and are less likely to break during demounting, whereas they can be deemed favorable.

With this, the analysis of the design parameters concludes, leading to the development of a design for recycling guideline meant to offer easily understandable, tailored information to machine- and magnet designers based on this analysis.

6.2 Design for Recycling Guideline

While some aspects regarding what magnet and motor design is suitable are basically universal, many others are very much conditional, relying on other design feature combinations and on the foreseen recycling method.

It is furthermore not easily accessible to designers without in-depth knowledge of the recycling processes, to choose a suitable recycling process. Therefore, for better usability, a design for recycling guideline with machine designers as the target audience should be developed.

6.2.1 Concept and development

The first concept of the guideline was designing a compact guide with few pages and simple explanations which features are suitable and why. This was soon complemented by adding images to the guide. A short excerpt of the first guideline is seen in Figure 6.6

Stack arrangement

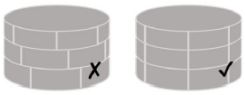
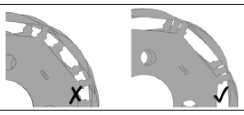
Illustration	*Recommendation*	*Explanation*	*Appl. ES*	*Appl. IS*
	Avoid skewing	Skewing forces disassemblers to either demount all the skewed stacks to push out the magnets or to un-skew on the shaft before doing so. In in situ the removal of the magnet powder may be challenging too.	X	X
	Limit skew levels	The more individual skew levels, the higher the demounting effort, also while for a 2-level skew, all magnets can be reached from the ends, f.e. to damage the coatings, at a 3-level and up skew level, this is not possible anymore.	X	X
	Favor skew angles that allow for large single open areas within the line of sight	This allows easy removal of the hydrogen processed powder without jamming.		X

Figure 6.5: Short excerpt from the first, linear guideline.

However, the guide soon exceeded 40 separate recommendations, while it was challenging to identify which of the recommendations could and should be applied to a specific design. As electric machines usually have to fulfill a variety of closely-defined requirements, few design changes are viable without compromising other functions. Therefore, machine designers can be hesitant to implement any changes. It is therefore advisable to provide few, clear and relevant as well as somewhat prioritized, or weighted recommendations and suggestions for them to work with.

It has to be further understood that it is not necessary to follow all of the recommendations or even the majority, as in many cases, a present design can be adapted with few, targeted changes.

Based on this, it was decided that the provided guideline should be adjusted to the planned or existing design. To do so, the design must be entered into a form and then, recommendations have to be generated based on the entered details. It was decided that the guideline should first only be adapted for IPM and SPM (and with limited applicability for AFM) electric machines for simplicity and that the questionnaire should be branched

based on which machine option is chosen. To generate a guideline from the questionnaire answers, a recommendation algorithm is required, where the recommendations are linked to the entries or entry combinations.

The questionnaire is shown in the following Table 6.2.

Table 6.2: Design for recycling questionnaire.

QID	Question	AID	Answers
1 (all)	How large are your rotor stacks, combined? Diameter + length = X	1A	$X < 200\ mm$
		1B	$200\ mm < X < 500\ mm$
		1C	$X > 500\ mm$
2 (all)	How are your magnets mounted?	2A	Internally mounted (IPM)
		2B	Surface mounted (SPM, limited applicability for AFM)
		2C	Not sure yet
3 (all)	What rotor/stator configuration are you using?	3A	Inner Rotor
		3B	Outer Rotor (or AFM)
		3C	Not sure yet
4 (all)	How are you mounting the stacks on the shaft?	4A	Press fit
		4B	Form fit (serrated/feather key, …)
		4C	Adhesive fixation
		4D	Other
		4E	Not sure yet
5 (IPM)	How are you skewing your rotor?	5A	No skew
		5B	Two levels
		5C	Three or more levels
		5D	Not sure yet
6 (all)	How will the stacks be produced?	6A	Backlack
		6B	Dotted glue
		6C	Interlock, laser welding
		6D	Something else
		6E	Not sure yet
7 (all)	Are you using polymeric features (except coatings and adhesives) in your design?	7A	Yes, larger pieces
		7B	Yes, but only very small pieces
		7C	No
		7D	Not sure yet
8 (IPM)	What magnet layout are you using?	A	Tangential (incl. Surface mounted magnets)
		B	V-shape or other angled designs
		C	Not sure yet
9 (IPM)	Are you layering the magnets tangentially?	A	Yes
		B	No
		C	Not sure yet
10 (IPM)	What magnet proportions are you using?	A	Larger magnets with balanced proportions
		B	Smaller magnets, which are very flat or long
		C	Something in between
		D	Not sure yet

Continuation of Table 6.3: Design for recycling questionnaire

11 (IPM)	Are you layering the magnets axially?	A	Yes, multiple magnets per stack level
		B	Yes, two magnets per stack level, but multiple individual stacks that are separated when the shaft is removed
		C	Yes, one magnet per stack level, but multiple individual stacks that are separated when the shaft is removed
		D	Yes, just two magnets along the whole length of the rotor
		E	No, just one magnet along the whole rotor length
		F	Not sure yet
12 (IPM)	How are your flux barriers designed?	A	Medium to large flux barriers
		B	Small or no flux barriers
		C	Not sure yet
13 (IPM)	How are your magnet pockets shaped?	A	Tight fit
		B	Space behind the magnet
		C	Not sure yet
14 (IPM)	How will you fix the magnets in place?	A	Adhesive unfilled
		B	Adhesive with filler
		C	Inmolding
		D	Press fit
		E	Mechanical, can be easily removed
		F	Not sure yet
15 (SPM)	How are the magnets affixed to the rotor?	A	Adhesive, no bandage
		B	Adhesive-fixed bandage, organic bandage
		C	Mechanically fixed bandage, metallic bandage
		D	Mechanical, bandageless fixation
		E	Something else
		F	Not sure yet
16 (all)	Are you using a passivation or phosphatization, below or instead of a coating?	A	Yes, it is hydrogen permeable
		B	Yes, but it is not hydrogen permeable
		C	Yes, but we are not sure If it is hydrogen permeable
		D	No
		E	Not sure yet
17 (all)	Are you using a coating?	A	Yes, Polymer (Epoxy, Parylene, …)
		B	Yes, Zinc
		C	Yes, Nickel or Nickel with Copper
		D	Yes, a combination of the aforementioned
		E	Something else
		F	None (or just passivation/phosphatization)
		G	Not sure yet
18 (all)	Are you segmenting the magnet?	A	Yes, with unfilled adhesive
		B	Yes, with filled adhesive inorganic filler (e.g. AlOx, glass beads)
		C	Yes, with filled adhesive, organic filler (e.g. fibers, carbon black)
		D	Yes, without adhesive (e.g., unfilled snakeline magnet)
		E	No
		F	Not sure yet

Continuation of Table 6.4: Design for recycling questionnaire

19 (all)	Are your magnets grain boundary diffused?	A	Yes
		B	No
		C	Not sure yet
20 (all)	Are your magnets grain boundary engineered to have a grain boundary phase with low REE?	A	Yes, but it's recyclable via HPMS
		B	Yes, it's not recyclable via HPMS
		C	Yes, we don't know if it's recyclable via HPMS
		D	No
21 (all)	Are you using a low-corrosion alloy?	A	Yes
		B	No
		C	Not sure yet

If the user selected that they plan to do an IPM design, they are shown 21 questions on the design. If they select that they are choosing an SPM (or AFM) design, they are shown 13 questions on the design. For AFM, due to lack of experience with the design, and similarity, the same rules are applied as for OR-SPM. A disclaimer will be added, that in terms of AFM, the guideline is not reliable yet and users will be asked to provide their expertise if they are well versed with designing AFMs. In the following, the recommendation algorithms will be explained.

6.2.2 Recommendation algorithms

When the user has entered the design details, they must be processed into evaluations of features and feature combinations, information about the suitable recycling processes and recommendations to assist the designer in improving the recyclability of the machine design. For this, specific answers and combinations of answers are supposed to trigger a set of rules, which provide the users with a tailored guideline. The details on the algorithm have been moved to the appendix due to being complex and very extensive.

The Table A. 1 in the appendix shows which answer combinations trigger which rule, as well as the text that is provided to the designer. The same is visualized in Figure A. 1. In the appendix, too. There it becomes visually evident, that Design for Recycling is highly interconnected and which aspects are highly interactive with others. Also, it shows that the algorithm is rather complex, which is why it is required to run in the background and should not just be delivered to users to find their own interactions.

To summarize, a total of 127 rules were implemented, which consist of logical triggers using typical AND, OR, AND/OR (transferred into viable logic in JavaScript), or NOT logic and text attached to it. The text for each output is descriptive, whereas illustration is deemed to not be necessary.

6.2.3 Usable guideline

The usable guideline was first designed in MS Excel for the basic logic and then programmed using a template for a conditional logic questionnaire using the free services provided by Figma (figma.com) and adapted using CSS and JavaScript. ChatGPT 5.1 was used for debugging the code and implementing an option to send the answers to get a personal consultation. The website was refined iteratively and in its functional format and is ready to test with industrial partners.

The user inserts information based on the questions shown in Table 6.2 and then gets to a page where it is shown how many neutral, positive and negative recommendations they received. Positive recommendations mean that some chosen features or feature

combinations are a good choice for design for recycling. Negative recommendations mean that changes should be made. Neutral recommendations can either evaluate features as actually neutral or name the conditions under which they could be good or bad. Alternatively, general recommendations are also marked as neutral. The recommendations are then shown below together with checkboxes so the designers can check the options bit for bit. If they find that they still require some additional consultation they can volunteer to send their answers and request a consultation being scheduled.

The output text can be exported as a .txt checklist for the convenience of the designer and discussion with the design team.

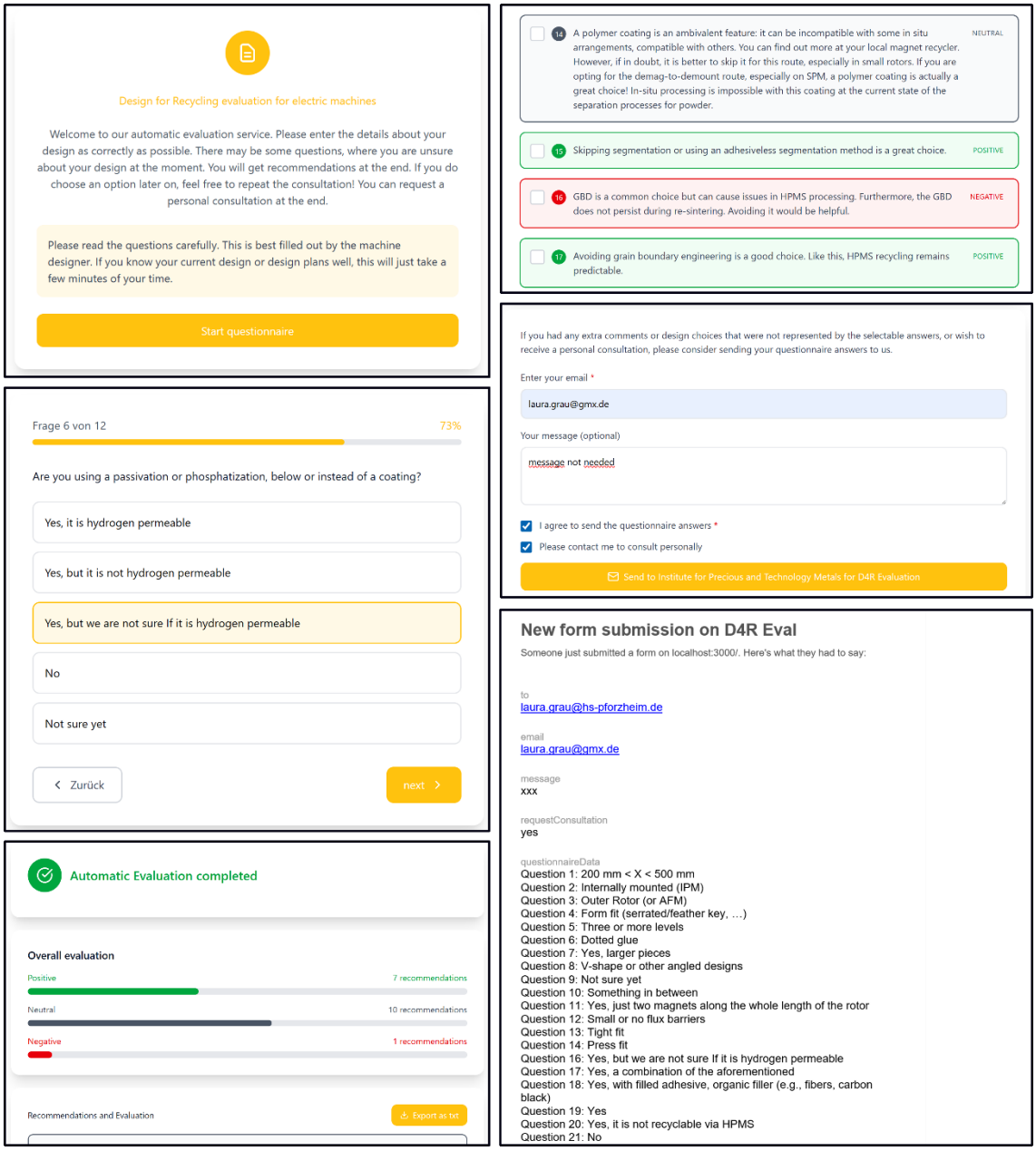


Figure 6.6: Screenshots from the tailored design for recycling guideline based on a questionnaire and a set of logical rules.

6.3 Normative and Legal Implementation Support

Policymaking can be seen as a part of design for recycling, which may sound abstract, but recycling-friendly policies can significantly lower barriers by offering incentives and increasing producer responsibility for the created waste, or by addressing waste collection processes [138].

Some typical measures and the de facto implementation of them in the EU in context of NdFeB recycling from electric machines are explained briefly in the following:

Extended producer responsibility aims to have producers integrate the costs of products throughout their lifecycle into the calculation of the market price and thus, have an incentive to design products that perform well in the end of life [11].

This is for example implemented by the WEEE Directive, where producers are obliged to take back the end of life products or rather, while taking the responsibility, allowing waste management specialists to deal with them [136]. Other means are influencing the collection, like for example informing consumers about the correct disposal (see WEEE symbol and recycling codes for polymers).

It is also typical to mandate the use of a minimum recycled content in products, or a minimum recyclable content of the total mass (like in the 2000 ELV Directive [379]) or for certain critical materials (s. planned new ELV Regulation [380] and CRMA [6]). Possible are also minimum recycling/recyclable quotas [136, 379].

These methods indirectly urge designers to adapt the design for recycling. However, there is also more direct means: For example, the Ecodesign for Sustainable Products Regulation (ESPR) mandates demountability, excludes some non-recycling friendly materials [252].

What can be also regarded as a political tool to support design for recycling approaches is subventioning the procurement of recycled material or also funding research projects which include design for recycling, like the projects SUSMAGPRO or REESilience, which contributed to this dissertation.

Another method is to support the traceability and to supply recycling-relevant information along with the products. This is, for example, done by the means of mandating a digital product passport for various products, a concept developed in context of the ESPR in general [252].

A very targeted approach, however, is the labelling of permanent magnet containing products as per the Article 28 of the Critical Raw Materials Act (CRMA) [6], which will be addressed in more detail in the following section.

6.3.1 Labelling as according to Art. 28 of the Critical Raw Materials Act

Article 28 of the Critical Raw Material [6] requires that a large range of electrical machine containing products are fitted with a label regarding the included permanent magnets.

These products include "magnetic resonance imaging devices, wind energy generators, industrial robots, motor vehicles, light means of transport, cooling generators, heat pumps, electric motors, including where electric motors are integrated in other products, automatic washing machines, tumble driers, microwaves, vacuum cleaners or dishwashers". The to-be-declared permanent magnets are NdFeB, SmCo, AlNiCo and Ferrites.

The label must include if permanent magnets are incorporated and what type of permanent magnets are present.

Also, there should be a data carrier present, which includes a clear identification of the responsible company or person, information on the weight, location, chemical composition,

coatings, adhesives and additives of the magnets and instructions how to safely access and remove the permanent magnets, providing the removal steps, tools and technologies.

The information on the data carrier can also be deposited in a digital product passport and if the magnets are incorporated in an electric motor, the declaration of the demounting can be made for the whole motor instead of every magnet individually.

In preparation of an implementing act, determining the format of the label, as announced in Art. 28.2, the European Commission's Directorate-General Joint Research Centre (DG JRC) and Directorate-General for Internal Market, Industry, Entrepreneurship and SMEs (DG GROW) prompted a standardization activity to support the creation of the Implementing Act.

6.3.2 Development of a technical specification (TS) in support of Art. 28 within CEN/TC 472 "Rare Earth"

The work on this task was executed first in an AdHoc group of CEN/TC 472, later on in a working group due to the long timeframe of starting an official working group being incompatible with the tight timeline. Both groups were operationally led and coordinated by the author of this thesis, with the much-appreciated support and supervision of the Convenor of the Committee at that time and later the Convenor of the founded Working Group 1 (WG1) in CEN/TC 472, Prof. Dr. Carlo Burkhardt and the secretary, Julia Listringhaus.

The groups were composed of a variety of stakeholders from academia, producing industry, policymaking, and waste management. The first AdHoc meeting was held on 07.11.2024 with the group consisting of merely 21 people. The fifth and last AdHoc meeting was held on 24.04.2025, the first and so far, last WG1 meeting to oversee the results of the AdHoc group and accept the draft for processing by the CEN was held on 22.05.2025. At the end of the activities, the overall AdHoc and WG1 members consisted of 74 members.

Due to the short timeline, it was decided, that creating a technical specification (TS) was more suitable as a first step than going for a full standard. Based on the TS, a standard can be created at a later stage.

The work was divided into different tasks. Task 0 served for organizing the compiled knowledge, writing the document and taking care of terms and definitions as well as the document structure and adherence to rules related to creating a TS. Task 1 dealt with defining the physical label specifications. Task 2 dealt with the data carrier, information storage and information accessibility. Task 3 dealt with how the indication of the location and weight of the magnets shall be done. Task 4 dealt with how to declare the magnet composition, Task 5 with the declaration of coatings, adhesives and additives. In Task 6, the format of the demounting instructions was addressed. Task 7 dealt with analyzing related and overlapping legislation, Task 8 dealt with any related normative documents, both to ensure the document does not impose any conflicting obligations, that the TS is as harmonized as possible with the normative and legal environment and that existing documents are used as efficiently as possible. Task 9 further dealt with providing and analyzing surveys for recyclers including general waste management and specific NdFeB recyclers to ensure that interested companies and individuals can participate even if they lack the time to join the meetings and do practical work. Task 10 did the same for producers.

In the following, an overview about the created document is given, however, as the finished standard (Fpr CEN TS-18263: Permanent Magnet Products — Procedure for declaring recycling-relevant information) is not published yet, it cannot be referenced or shown in detail in this work.

In terms of the scope, the original scope from Article 28 is preserved, however, as the TS is meant to be horizontal by nature, it is also recommended to be used on some other typically-magnet containing products like hard disk drives, transducers, loudspeakers, drones, motorized toys, and electrical vertical take-off and landing aircrafts. Both generally from the original scope of Art. 28 and the extended scope from the TS, the labelling recommendations clearly exceed the topic of the dissertation, as also other magnet types and components aside from electric machines are considered. However, it has to be noted that while the TS is highly flexible to apply to any other magnet-containing product, for the application for new permanent magnet materials or niche materials, it would have to be extended or users would have to design their own magnet type-specific information package.

A range of terms and definitions regarding the product groups, magnet types and non-magnetic materials (coatings, adhesives, …), recycling, labelling and determination methods for the magnet composition, are given.

For the physical labelling requirements, first, thresholds are given. Participants, recyclers and producers alike, found it sensible to not label every product, but only such, which contain enough magnet material in a component that disassembly is warranted to facilitate the efforts on both sides. This diverts from the Article 28 but was seen as a factor of utmost importance.

The individual threshold is decided by both the total weight of the product, as larger products can be significantly more complex, as well as the magnet type. The threshold relates to the total mass of magnets within one component. For products under 400 kg, the threshold is 25 g for NdFeB, SmCo and AlNiCo and 250 g for Ferrites. For products over 400 kg, the threshold for Ferrites is kept the same, while the threshold for the other magnets is increased to 60 g. Due to the relative ease of demounting, for NdFeB and SmCo loudspeakers, a threshold of only 3 g is recommended.

The label must be placed either on the surface of the product or in a place where it becomes immediately visible when the product is open or demounted. Further, some components shall be labelled as well to ensure the information is passed to the material recycler. These include electric machines as well, where the label should be placed on the housing. It is further recommended to label the rotor of electric machines as well.

Labelling the individual magnets within components or single magnets below the component threshold do not have to be labelled, it is also not recommended – dismantlers are prompted to pre-sort the magnet holding components and magnets to then forward them to the recyclers. It is further recommended to clarify if any additional sorting criteria are recommended by the material recyclers, like sorting by coatings.

On the label, the magnet type shall be indicated. It is differentiated by magnet base alloy (for example, N for NdFeB) and whether the magnet is a bulk magnet (for example, N1 for sintered NdFeB), for example a sintered or cast magnet, or a composite magnet (for example, N2 for bonded NdFeB), like a polymer bonded magnet. Additionally, for $SmCo_5$ and Sm_2Co_{17} two separate numbers are used as $SmCo_5$ streams must be kept pure to recycle.

If in a product, different magnets are applied, for example, an electric vehicle with several ferrite and NdFeB motors, the product gets the label "M1" for mixed magnets, but the components get labels according to the included magnets. An example is shown in Figure 6.7 for sintered NdFeB.

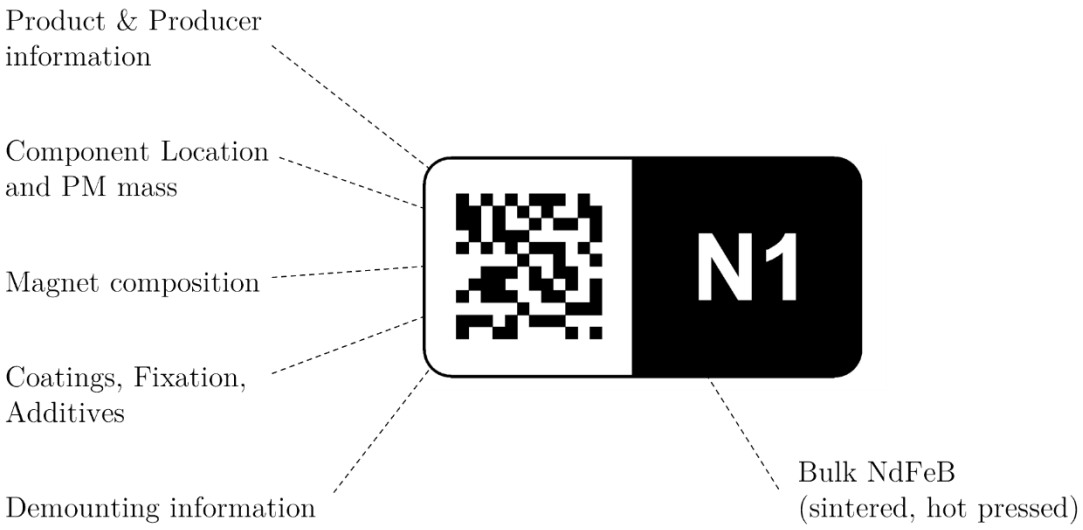
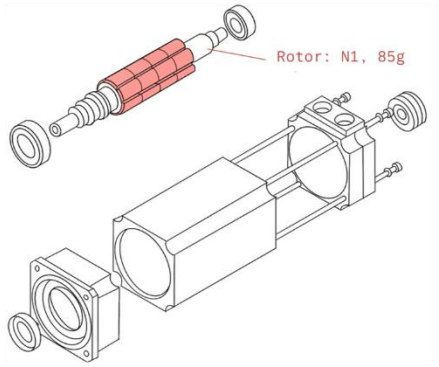


Figure 6.7: Physical labelling format with information on a magnet-containing product.

The data carrier choice is open, but a QR code is recommended. It is required that the code can be read with a mobile phone or tablet. The choice of the labelling method is left to the producer as long as the label is legible and persistent. The data stored must be accessed by scanning the data carrier and it must be linked to a unique identifier. Where the data is stored is left to the producers, however, it must be accessible to stakeholders of the waste management chain. Many producer stakeholders voiced that they would prefer a database managed by the EC. The data must be provided along the value chain; however, the responsible actor is the one placing the product on the market. A third-party service can be used but the information must be kept up to date, safe and tamper-proof. The detailed requirements can be assessed in the standard.

Location and mass



Composition

Co a	(≥0.1 %)
Cr a	(≥0.1 %)
Dy b	(2 - 3 %)
Nd c	(25 - 30 %)
Pr a	(1 – 5 %)

Coatings, adhesives and additives

1. Magnet ID: **R11MN7-1**
2. Coating structure: **2, passivation, epoxy (filled: carbon black)**
3. Hydrogen permeability**: not tested**
4. Fixation method: **adhesive, epoxy, (filled: AlOx)**
5. Additional internal materials: **none**
6. Additional external materials**: End plates, PMMA**
7. Environmental indications: [**Safety data sheet attached**], **formation of carbon monoxide, formaldehyde and phenole possible when heated**

Figure 6.8: Indication of location, composition, and non-magnetic materials on the data carrier, fictious example.

The location of the magnets or magnet-holding components must be indicated along with the mass. This is best done in a schematic plan, where a drawing of the product is provided, where the location of the components is clearly indicated. Simpler drawings are preferable.

The magnet composition shall be declared using declaration tables provided in the annex of the TS. As magnet alloys are intellectual property, it may be difficult to obtain permission from producers outside of Europe to declare the full composition. Furthermore, for short-loop processing, the effects of many elements, for example those used for grain refinement can hardly be quantified well, therefore, this information is rather irrelevant for the recycler. In several panel meetings, tables with the elements relevant for short-loop processing compiled, along with those of safety relevance for long-loop recycling. Long-loop recycling via hydrometallurgy does require full chemical analysis, however, this is often done in bulk with mixed magnets already, whereas this was an acceptable solution for participants from long-loop recycling as well. For each element given in the table, a threshold is given. If the composition has a content of this element above the threshold, it has to be declared using the lowest possible range. The ranges represent the common content ranges for the elements. For example, Dy is declaration exempt below 0.5 %. Range a includes up to 2 %, range b up to 3 %, range c up to 5 % and range d contents over 5 %. For safety critical elements (s. long-loop recycling, includes e.g., Cr), fewer ranges are given just to indicate that there is at least a critical content. It is also allowable to give a full declaration, but this option is meant to alleviate issues for smaller producers with less negotiation power. It is furthermore recommended to declare organic contamination after the production process.

The additional materials are meant to be declared as coatings, internal materials, external materials and fixations. For the coatings, the number of layers and the type of each layer shall be given. The layers include passivations. Along with this, the type of each layer shall be given, however, for organic materials, colloquial names like "epoxy" or "parylene" are allowable. If the material is filled, the filler must be indicated as well.

For the fixation, it shall be given whether the magnets are mechanically fixed directly by the stack, by an additional feature, by a polymer inmolding, or an adhesive. Here again, if filler materials are used, it must be indicated.

"Internal materials" refers to materials within the magnet, like adhesive for segmenting or the bonding polymer. "External materials" refer to polymeric features of the rotor. Again, fillers must be indicated. Furthermore, for all polymeric materials, the safety data sheets of the resins must be provided.

Finally, the demounting should ideally be given by visual cues as well, colloquially referred to within the AdHoc group as the "IKEA principle". While it is only recommended to use a visual plan, not using one entails providing the explanatory text in all EU languages to ensure inclusivity. The demounting sequence, as mentioned, ideally supplied with images, must include the steps, tools and any other required information. Ideally, means for automated disassembly should be given as well.

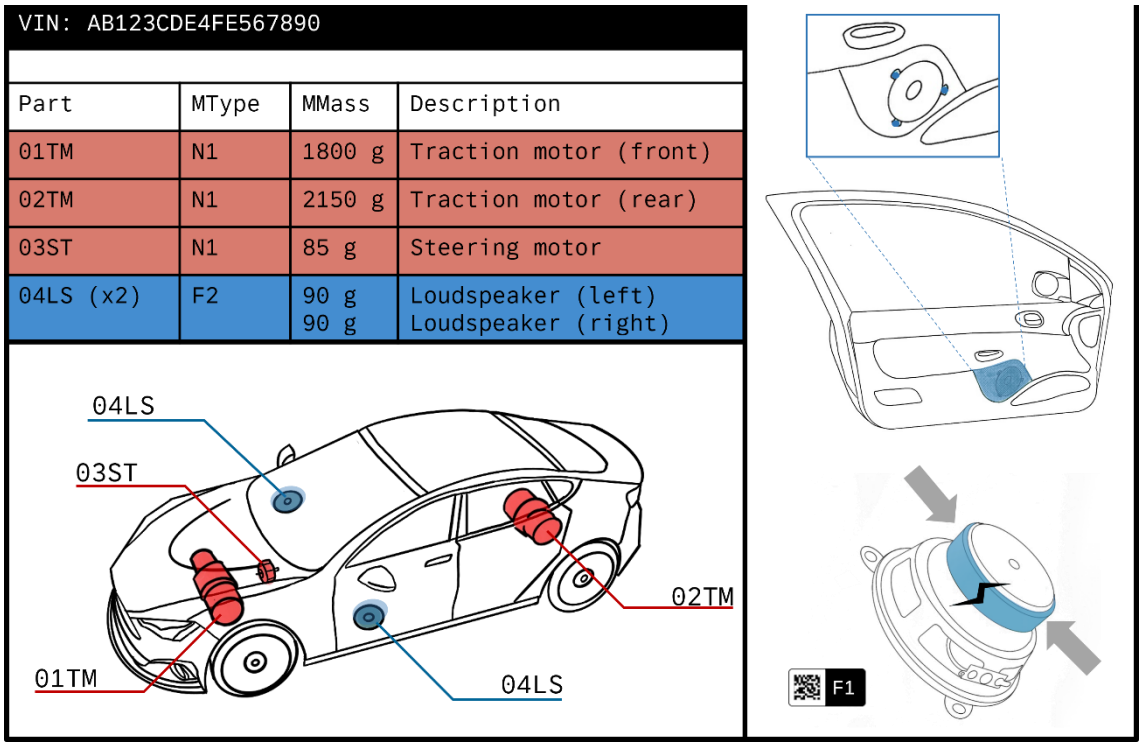


Figure 6.9: Location of permanent magnets within a vehicle and demounting of a magnet holding component, fictious example.

Overall, while the TS diverts partially from what has been foreseen by the CRMA Art. 28, the changes were seen as essential by the working participants to ensure that labelling is most efficiently designed, providing all useful and viable information while keeping the burdens for labelling and analyzing the labels to a minimum.

The document has been finalized in the beginning of June 2025 and entered into voting end of November 2025 after CEN and CCMC editing. The TS will be at least partially adapted or referred to directly in the upcoming implementing act.

In February 2026, the document has been accepted and is currently under editing by CEN to implement editorial comments.

Furthermore, as the technical specification is understood as being horizontal by nature, some other products which were not part of the Article 28 are suggested as further possible applications of it. However, an amendment of Article 28 is foreseen to also include these products as well. This further increases the potential impact of the technical specification.

Limitations to the impact, however, lie within the long timespan until products are labelled, deployed into market and returned to recycling and in products which are not foreseen for the European market.

Chapter 7

Summary

The purpose of this dissertation is to enable higher recycling rates and good quality of recycled materials when recycling NdFeB from electric machines. Contrary to the general understanding of design for recycling being limited to product design, this approach encompasses process design and policy design as well. "Design" in this work is understood as any means to reshape objects, actions and their framework. While this is a rather wide perspective, the work is constrained by using a strongly material-centric recycling approach from one specific type of scrap source, which fortunately is the most abundant one.

In the first chapter, the introduction, the criticality of the rare earths and the application of NdFeB in electric machines is addressed. In the second chapter, the motivation to recycle is explained from three perspectives: The environmental and social issues caused by primary production, the lack of resilience of the supply chain without secondary sources and finally, the so-called balance problem, which shows the imbalance of the elements within the primary ores and the demand of each element. In the third chapter, the strategic approach and any hypotheses are summarized.

Chapter 4 discusses the theoretical foundations and the state of the art and is designed to give the reader a thorough understanding of the topic. It lays the foundation for the practical investigations presented in chapter 5 and the implementation in chapter 6.

In chapter 4.1, permanent magnets are addressed, both in terms of their performance characteristics, applications, production and more in detail, the emergence of magnetic properties in sintered magnets. To understand how the recycling processes can affect the material, it is essential to understand how they come together in the first place, as the strengths of NdFeB arise from a unique interaction of specific elements primarily, and a specific microstructure secondarily. The focus is kept strongly on sintered NdFeB magnets.

In chapter 4.2, the recycling of sintered NdFeB magnets using hydrogen processing of magnetic scrap (HPMS) is addressed, explaining typical pre-processing actions from collection, identification and sorting, disassembly, to demagnetization and separation of the NdFeB magnets from other materials. It is found that the challenges basically start far before the actual disassembly or recycling: Merely identifying what components contain recyclable NdFeB is rather challenging. It becomes clear that most of the effort must be invested in pre-processing, rather than the very straightforward recycling process itself. Then, the path from the HPMS powder to a resintered magnet and the post-processing methods are explained, and finally, short-loop processing via HPMS is set into context and compared to other recycling processes.

Electric machines as a scrap source are analyzed in chapter 4.3. Their function and overall types are explained briefly, to then go into more detail on the ones that contain permanent magnets and to explain why permanent magnets are applied and what the

benefits and disadvantages are. Finally, the application of NdFeB in electric machines is critically discussed, specifically addressing issues which may collide with design for recycling objectives.

In chapter 4.4, the literature-based research is concluded. Firstly, eco-design is addressed as a concept and recycling is set into perspective. The treatment of waste as an anthropogenic mine, the societal factors and responsibilities of industry and policy are discussed. General research on design for recycling, which can also be applied to the specific topic of the work, are reviewed compactly.

Afterwards, published works on design for recycling of electronic products and machines are addressed. Finally, through both literature research and extensive patent research, inventions and developments on the process- and product level are reviewed. However, not all of these are directed specifically to facilitate recycling. The most promising inventions are illustrated and explained, with the intention of inspiring even more recycling friendly developments by the readers.

Overall, design for recycling in general, especially regarding disassembly, which is a topic for several "design for …" methodologies, is well researched. However, research on dealing with permanent magnets or permanent magnets within an electric machine is still highly deficient.

It is identified that the focus of the practical investigations should lay on the processes which are specific for magnet recycling: Demagnetization and separation processes and their influences and the effects of different design choices. The materials and features which are the closest to the permanent magnets, which are coatings and fixation methods, are deemed to be most critical for the success of the recycling process. Furthermore, it is found that the full demounting of especially smaller motors can be economically questionable due to the high effort required. It is therefore sought to find "shortened loop" pre-processing methods to alleviate the effort and minimize manual handling for economic reasons and worker safety.

In chapter 5, the practical investigations are reiterated. In chapter 5.1, the methods for laboratory-scale recycling and analysis of the materials and arrangements are described. The first presented publication in chapter 5.2 deals both with the possibility of treating permanent magnets in situ in an electric machine, meaning the rotor of the machine is not demounted, as well as with the separation of contaminants from the powder. As HPMS turns a magnetized bulk magnet into coarse, demagnetized powder, demounting and demagnetizing could potentially be skipped. However, the resulting powder must be free of critical levels of contaminants before pressing and sintering the new magnet.

It is found that Ni-based coatings can be easily removed, but not with the reported method in literature, where the HPMS powder is sieved before milling, but rather, after milling. If the powder is sieved unmilled, the large HPMS aggregate particle size intersects strongly with the particle size of coating flakes. Polymer resins applied as coatings and adhesives are found to be incompatible, as no viable separation method was found. Zn residues also mill finely and could only be potentially removed chemically. It is further found that sufficient expansion volume must be ensured – either by design or by preparing the rotor with cut-ins. Further, non-hydrogen permeable materials must be broken prior to the treatment to allow the hydrogen to react with the NdFeB magnets.

For this method, ideally, magnets which are merely passivated and affixed mechanically are used. In the best case, this is done with a fixation feature which allows volume expansion of the magnet. To properly protect the magnets from corrosion, a compatible low-contamination housing and cooling strategy would be beneficial.

In the less ideal, but still viable case, Ni-based coatings including Ni-Cu-Ni can be used and the rotor can be prepared minimally to allow hydrogen access and expansion space. Finally, another factor for the success of this method is the powder removal which is

facilitated by avoiding axially layered magnets and skewed designs, especially with large skew angles.

While the design requirements are rather narrow for successful in situ treatment, the process is extremely simplified and viable even for very small internal permanent magnet rotors, tapping a scrap source which would be economically out of reach normally.

The second publication in chapter 5.3 deals both with the effect of thermal demagnetization on the material and the possibility to facilitate demounting via thermal demagnetization.

Adhesive being an exclusion criterium for in situ treatment is now a desired feature: at 400 °C in air atmosphere, most used resins for coating, adhesive fixation and segmentation are combusted almost completely. However, for some segmentation, not enough air reaches the inside between the segments, which necessitates that the segmentation is broken apart while still hot, for example by impacting or tumbling the magnets.

It was found that no significant carbon uptake from polymer coatings and adhesives is happening. This appears to be due to the standard use of passivations or phosphatizations below the coatings. Oxygen uptake is seen in most samples, the least uptake is recorded with Ni-Cu-Ni coatings, the most with uncoated magnets. The oxygen uptake, however, is highly localized and leads to discoloration of the surface. When the discolored surface is removed, for example by sandblasting, the oxygen uptake from the thermal demagnetization can be fully reversed. Long term oxidation from before the demagnetization however reaches deep into the magnet and cannot be reversed this way.

Zn coatings were found to be highly problematic as they infiltrate the grain boundaries during thermal demagnetization, reaching very deep into the material and potentially even carrying in other contaminants. This effect can be mitigated by keeping demagnetization times short. 15 minutes was enough for all smaller rotors and magnets and no issues from infiltration arose. Still, Zn coatings should be avoided in this process as a precaution, just like rotors with large polymer features to avoid an overflow of toxic fumes. Waste gas deduction and treatment are required in most cases. This method works best for SPM rotors, but also for IPM motors with generous amounts of adhesive in the magnet pockets. Some rotor designs making even more use of adhesive fixation, like affixing the shaft with adhesive or using backlack stacking can benefit even more from this method, allowing effortless demounting of all materials.

The findings of both publications do not only apply to the shortened loop methods, but also to general processes applied for demounting, as oftentimes demagnetization is required to reduce demounting forces and promoting safe handling, and since powder purification can be necessary outside of complete in situ treatments. For example, Ni-Cu-Ni coatings are challenging to remove mechanically from the bulk magnet due to the ductility of the material and could be better removed from the powder with less manual handling instead. Looking at both methods, it is clear that the viability for recycling of many design features strongly depends on their compatibility with the recycling process design.

However, both also bring up a question: How much of the specific contaminants are acceptable to remain in the material? Complete separation can be technically impossible or economically challenging to achieve. Therefore, it is important for recyclers to know, what requirements they have to meet.

For this, the third publication presented in Chapter 5.4 combines literature research on the influence of contaminants with practical investigations filling up the gaps. The mechanisms of how contaminants affect the magnetic properties are discussed in detail and their effects are quantified based on data derived from literature and from the complementary experiments. On oxygen and carbon, there is a significant amount of research already, which has been compiled graphically and summarized generally. It is well visible that at around 1000 ppm or 0.1 wt.% of C, the magnetic properties rapidly drop.

For oxygen, the limit is less straightforward to determine, and it is indicated that some oxygen is actually beneficial to recycling. There is no hard limit, but a lower content is preferred. In terms of Ni, the mechanisms of how Ni can be integrated into the material have been researched, however, little information was available in literature.

An investigation with varied Ni addition was conducted, the preliminary experiments testing contents from 0.025 wt.% to 10 wt.%, the main experiments testing 0.1 wt.% to 5 wt.% of Ni addition. The results showed that until around 2500 ppm, or 0.25 wt.%, the addition did not significantly impair the magnetic properties, but that above this, the magnetic properties can be almost completely diminished at higher additions. Another issue here is that Ni enriches in the hard magnetic phase and thus, cannot be removed in short loop recycling and may enrich over several recycling cycles. Cu is generally added to most magnet alloys anyways due to its positive effects on the microstructural development and corrosion resistance, therefore it is not further addressed. In terms of Zn, even extremely low additions of 0.025 wt.% lead to metallization of the furnace tube. As this may entail damage to the sintering equipment, especially the vacuum pump, the experiments were not possible to continue. The effects of Zn on hot-pressed magnets were reported to be positive, but in sintered material, no Zn seems to persist, as it evaporates. This leads to a full loss of Zn as well as a lack of density. Zn should be separated as well as possible and it should be avoided by designers overall.

Overall, the main mechanisms of contamination are a) impairment of densification, b) binding of the active Nd-rich phase which leads to a lack of isolation of the grains, c) the creation of soft magnetic phases like α-Fe and d) loss of magnetization and magnetic anisotropy of the hard magnetic phase by integration of contaminating elements.

Based on the literature research and practical investigations, the implementation of the results are discussed in chapter 6. In chapter 6.1, design recommendations from the integration and labelling of the electric machines down to the processing and alloy of the magnet, are given. However, as mentioned, many design features perform differently depending on the processing, which can be challenging to understand and overview. Therefore, to meet the needs of machine designers who should be able to adapt their designs in a targeted, simple-as-possible way, as recyclability is one of many requirements imposed; a guideline for them was designed. The final guideline is interactive and adapts to the present design features.

The investigations have proven more than once that labelling of the permanent magnet containing products is one of the most important and universally applicable factors of design for recycling. Therefore, the obtained knowledge has also been applied by leading a standardization activity for the purpose of supporting the Implementing Act of the CRMA's Article 28 on labelling.

With this, a well-rounded approach to design for recycling is proposed that offers both short-term solutions by optimizing and simplifying pre-processing and long-term solutions by providing guidance on machine design and supporting the European policy design on labelling of magnet containing products.

Coming back to the hypotheses from chapter 3, not all have proven to be true:

- **Good recyclability is achieved with suitable combinations of product features and process features – the suitable product features strongly depend on the process.**
  ⇒ This hypothesis can be accepted as true to a high extent. The only generalizable "rules" for design for recycling are that labelling is beneficial in general and that designing for non-destructive disassembly is either helpful or not harmful to different recycling processes.

- **Processing of rotors without demagnetizing and disassembling the magnets is viable in most cases. The removal of coating and adhesive residues from powder is viable in most cases.**
  ⇒ This hypothesis cannot be accepted, or rather, only to a very limited degree. While in situ treatment is possible, its viability is highly constrained by very specific design requirements.

- **Thermal demagnetization can be a means to disassemble and demagnetize adhesive-fixed magnets, including segmented magnets.**
  ⇒ This hypothesis can be accepted. Thermal demagnetization in air, being a simple and accessible process is viable for many designs and detrimental effects can be mitigated in most cases.

- **The separability of magnet material and nearby other materials is essential for achieving high quality recycled material. However, for all contaminants, there should be an acceptable limit whereas perfect separation is not necessary.**
  ⇒ This hypothesis can be mostly accepted. Clear limits cannot be given for oxygen contamination, for example. Further, for Zn, while perfect separation may not be achievable, it should either be attempted or means to manage Zn deposition on sintering equipment should be investigated.

- **Suitable labelling and provision of information on the to-be-recycled magnets is superior to testing and the time and money required for labelling and reading the label are well invested.**
  ⇒ This acceptance or rejection of this hypothesis should be postponed. The tendency is to accept it. It is clear that labelling and reading the label is worth it in most cases. If a recycler is already familiar with the product, reading the label may not be necessary. Testing likely takes more time, as the location of the magnets has to be determined, the optimum disassembly sequence must be found and finally, the magnet composition and coating must be determined. However, as a range of novel testing methods for locating the magnets and analyzing their coatings and composition rapidly for example, are currently under development, it would be too early to accept this hypothesis.

Chapter 8

Conclusion and Outlook

For this work, "design for recycling" had to be reframed, as a typical product-centric design strategy was not a viable approach. However, through choosing the most abundant source of end of life NdFeB and focusing on the most efficient and ecological recycling method which is currently available at industrial scale and not duplicating work on general rules for design for recycling, it was possible to work out a rather comprehensive solution.

Providing product design recommendations, or the arguably most important factor of product design, which is labelling of the products and components, is highly valuable and should not be neglected. However, these measures only take effect after the products have been developed, produced, sold, used and then arrived at a waste management facility. This can take up to 20 years or even more depending on the product type. Therefore, process recommendations have also been given.

Guidance on product design is provided by a tailored, adaptive design for recycling guideline. The guideline is currently in an easy-to-use website format but should be integrated into a framework where data safety can be ensured. Alternatively, an integration into existing computer-aided design (CAD) and product lifecycle management (PLM) tool chains can be considered. The presented tool can be applied for the most common permanent magnet driven electric machine types, and some information can be transferred to others, which show constructive similarities. For example, transferring some recommendations from outer rotor surface mounted permanent magnet rotors to axial flux machines as in both cases, the magnets can be secured by adhesive only and very easily removed, may be viable. However, this transfer of recommendations also has its limitations, and should only be done by an expert who is familiar with both machine types and their specific characteristics. As both product architecture and recycling processes are expected to advance in a short timeframe, it is vital to adapt the tool as required. Furthermore, the tool should be “future-proofed” as much as possible, as already within the timeframe of a lifecycle from design to recycling, such advancements can be expected. However, at the moment, the “future-proofness” cannot be easily anticipated due to a lack of application experience. This issue will hopefully be lessened over time. Furthermore, based on user feedback and demand, other electric machines or even other components could be integrated in future versions. It is also planned for future versions, when the processing routes available at industrial scale are clear that an advanced mode can incorporate this and give the user only the answers applying to the selected route. This may be worked out with recycling providers directly and could be specific to the recyclers.

Policy support regarding the labelling of permanent magnet containing products both aims to inform recyclers about the products and to make designers think about the demounting strategy for their product, which has to be given along with the label. For this, CEN TS-18263 was designed and will be published soon, and the EC already works on implementing the input from academia and industry that was compiled in this work.

In terms of process design, different strategies fit for various machine designs to minimize complex processes like demounting and manual handling have been developed, tested and published. Further methods should be researched and developed, especially regarding the separation of epoxy resin from magnet powder and towards recycling-friendlier, electrically isolative coatings.

Any method used in recycling, including such methods that simplify the process in a technical sense, should be evaluated quantitatively in life cycle-based comparative analyses. As the facilitation of recycling, or on the other hand also the implementation of recycling-friendly product designs is targeted towards improved sustainability, the overall sustainability-related performance should not be neglected. This should be subject to further, specialized studies.

While this combined approach already has potential for widespread effectiveness, it is still necessary to recommend that this topic should be further advanced. This is required both to keep up with developments and to take a closer look at any specific aspects which may turn out to be important for recycling. It cannot be guaranteed that all risks to recycling have been adequately addressed, as electric machine design is manifold and often highly specialized.

The popular image of design for recycling is not ideal and frankly, deservedly so if recyclability is prioritized over other requirements towards robustness and performance. Additionally, an electric machine is a system that usually has to meet a multitude of narrow requirements. Therefore, machine designers are oftentimes appalled by the idea of adding design for recycling to their list of tasks and oppose suggestions for changes from their usual approaches.

The solution lies in moderation: recognizing that there is no one-fits-all solution, understanding that good designs can look completely different for different processing paths, and finding the most viable processing path for the design. Like this, minimal adjustments can have a highly positive impact on recyclability.

Analyzing current products in most cases reveals that design for recycling has not been in the mind of the designers, but that small changes could have greatly simplified the process. Some features, like screws overmolded with rubber, or clips that could be easy to remove if one could just get a tool under it seem thoughtless and are very frustrating when disassembling products. A tiny crevice or bent-up end of the clip or skipping the overmolding are easy to implement changes that could save money in assembly and greatly shorten disassembly time.

It is of utmost importance to normalize and popularize eco-design, including design for recycling, and to educate present and future designers that it can and should be applied in accordance with fulfilling other requirements. Some of the measures of this dissertation already support this, along with different events held to educate people on design for recycling for NdFeB permanent magnets. Such events were conference presentations, cooperative projects with machine designers and workshops for school students, university students and professionals.

In the years of the dissertation, the importance of a resilient supply chain and assuring the supply of critical raw materials have been very present in the public due to political upheaval and wars in different areas of the world along with the implementation of tariffs and tangible effects caused by the shortage of a range of materials like oil, semiconductors and computer chips. When extensive export restriction on a range of rare earths were announced, the future of the European automotive industry temporarily seemed on stake.

These experiences and the associated pressure should deeply motivate us to change our approach, to make better use of secondary raw materials and to accelerate both our research and the scale-up of recycling technologies, which is often held back by administrative burden and lack of experience.

It should be noted at that point again, that closing the gap between the demand and supply of critical materials is not automatically given when recycling is implemented, but any amount of material reduces this gap.

To make this more tangible: Recycling is not expected to cover the entire demand gap until at least 2050 due to the increased demand and the long lifetimes of the major scrap source products, namely electric vehicles and wind turbines; however, it is expected to mitigate the supply deficit to some extent [18]. Leitner and Grandjean [381] modeled when EOL vehicles' motor recycling would be economically viable and assumed that 7.4 million EOL vehicles with an average motor power of 72 kW would be required to operate a commercially feasible recycling plant.

Disregarding whether permanent magnet recycling is factored in representatively enough, it is clear that the volume of the waste streams is decisive to the success of recycling businesses dealing with “novel” scraps, such as NdFeB magnets, which have been rarely recycled to date. This shows that expectations should be kept realistic, but also, that every magnet counts.

Aside from the mentioned issues, positive effects can be seen as well: many research projects are working on rare earth recycling, and eco-design theory is evolving as well. Furthermore, policies targeted at ensuring critical raw material supply, clean industries and eco-design have been implemented, and factories for rare earth recycling are built in several places in Europe and the US. Conventional magnet producers are also using their know-how to produce secondary material.

In the near future, it is important to promote the development of the infrastructure for producing secondary material by lowering administrative burdens and by subventioning the production and procurement of secondary material.

After addressing the requirements regarding product, process and policy design, the societal component of the solution of this problem shall be elaborated briefly:

To keep and accelerate this positive process, it must be ensured that this topic does not fall back into public irrelevance. On a more personal level, sustainable development overall will require openness, adaptability, mental flexibility and optimism from every stakeholder, along with a willingness to collaborate and exchange information.

With these thoughts, this dissertation on design for recycling of NdFeB from electric machines is concluded.

Appendix A

Table A. 1: Recommendation rules for the interactive design for recycling guideline.

RID	Requirements	Output text
1	None	Dear designer, this guideline is meant to offer you options to improve the recyclability of your electric machine. Please do not neglect the other requirements for the sake of "perfect" recyclability, but rather try to find a compromise of a robust, functional machine with good recyclability
2	None	There are different options to disassemble or process the permanent magnets from the electric machine. First of all, full disassembly and demagnetization is possible. In some cases, it is possible to demagnetize and demount in one step, where the magnets are mostly fixed by adhesive. This process is referred to as “demag to demount”. Furthermore, some designs allow HPMS processing of the entire rotor, where the magnets decrepitate to powder and demagnetize within the rotor. However, the requirements for such designs are very narrow.
3	None	Label your rotors and motors. A label is – in any arrangement – the best possible feature to improve recyclability. If you use a closed-loop scenario, you may not need it due to information being available still.
4	None	General rules for facilitating disassembly apply here too – with the difference, that dealing with magnetized components often requires demagnetization. Especially when the motors are large and the magnets have to be demounted. In HPMS, the powder is automatically demagnetized. Therefore, in some cases, demagnetization can be skipped.
5	None	Consider an easy to demount product design. In most electric machines, the bearings fail long before the motor takes damage. The motor or rotor could be used in a remanufactured motor with new bearings or similar. Make sure your bearings are easy to remove. This helps repair, reuse, and recycling!
6	None	For designing a good housing, try to use screws to hold the housing together and try to place them in a way they are visible and accessible from one angle – this greatly facilitates automatized disassembly! A great housing also provides good protectivity by being well sealed and providing good cooling. By this, you can save money on coatings, heavy rare earths, and segmentation, and also improve the recyclability.
7	1A	Smaller rotors are often not viable to demount. Ideally, the rotor is compatible with in situ treatments or at least with “demag to demount”.
8	1B	Medium rotors can be viable for demounting, if the material is highly valuable and/or abundant, but the demounting should be easy and quick to do. Another option is to ensure the compatibility with a shortened-loop process like in situ treatment or “demag to demount”.
9	1C	For larger rotors, demounting is usually viable, while shortened loop methods may be restricted due to the size of the rotor. Here, it is vital to ensure the demounting is economical. For very large machines, like industrial-scale energy generators, more effort can be allocated than for, for example, traction motors.
10	2A	IPM are generally more challenging to recycle than SPM, since the magnets are embedded and challenging to reach.
11	2B	SPM are generally easier to recycle than IPM, as the magnets are well within reach and can be demounted rather easily. They are especially suitable for “demag to demount” processing.
12	2C	If you have the choice, favor using an SPM due to the ease of disassembly and the general suitability for “demag to demount” processing.

Continuation of Table A. 2: Recommendation rules for the interactive design for recycling guideline.

13	3A	Inner rotors are a common choice, but require more solid fixation of the magnets due to the centrifugal forces pointing away from the magnet fixation.
14	2B and 3B	using an outer rotor SPM arrangement or an AFM is an excellent choice for recyclability, as in both cases, the magnet fixation can be designed rather simple, facilitating the demounting. These arrangements are especially suitable for “demag to demount”.
15	((2B and 3C) or (2C and 3B) or (2C and 3C)) and (1A or 1B)	If you have the choice, using an outer rotor SPM arrangement or an AFM is an excellent choice for recyclability, as in both cases, the magnet fixation can be designed rather simple, facilitating the demounting. These arrangements are especially suitable for “demag to demount”.
16	4A and (5B or 5C or 5D and/or (1B or 1C))	A press fit mounted shaft is a common choice for IPM, however, when combined with skewing, where the magnets usually have to be demounted by demounting the stacks, it is not the best choice. Either avoiding skewing or at least opting for a rotor-shaft mounting that is easier to demount would be advisable.
17	4B and (5B or 5C or 5D and/or (1B or 1C))	Skewing often requires full demounting. A form fit used for rotor-shaft connection can facilitate demounting the stacks from the shaft. Please check if this is the case for you. If yes, great choice!
18	4C	Connecting shaft and stacks by adhesive can have some benefits. Adhesive is great if you will thermally demagnetize the rotor – for example for a shortened processing route via “demag to demount”.
19	4D and (5B or 5C or 5D and/or (1B or 1C))	As your chosen rotor-shaft connection is not part of our catalogue, but you are skewing the rotor or considering to skew the rotor, please check the following aspects: Is the connection easy and quickly to demount? Is the method to do so intuitive and can be done with typical equipment including hydraulic presses
20	4E and (5B or 5C or 5D and/or (1B or 1C))	For skewed rotors, as well as medium sized or larger rotors, where demounting is an option, please choose a easy to demount rotor-shaft connection, for example a form fit or use adhesive.
21	5A	No skew is always a great decision. Please keep it that way, if possible!
22	5B	Generally, skewing is detrimental to most processing routes. It would be preferable to avoid it, if possible. Alternatively, it should be aimed for in situ or “demag to demount” treatment, which may still be possible with a 2-level skew, if some requirements are minded.
23	5B and 12A and 13B	Combining a two-level skew with large enough flux barriers and magnet pockets can allow in situ treatment or thermal demagnetization, if the coating and magnet fixation work with it as well.
24	5B and 12B and 13B	Combining a two-level skew with small flux barriers is basically one-off. It would be good to either increase the size of the size of the flux barriers or to avoid skewing. However, as the magnet pockets are slightly larger, it may still be possible to process the rotor in situ or by “demag to demount”, if the coating and magnet fixation work with it as well. If the design cannot be adapted in some way, it can be tested whether the current design does indeed work.
25	5B and 12A and 13A	Combining a two-level skew with little space in the magnet pockets is basically one-off. It would be good to either increase the size of the size of the magnet pockets between the magnets and the shaft) or to avoid skewing. However, as the magnet pockets are slightly larger, it may still be possible to process the rotor in situ or by “demag to demount”, if the coating and magnet fixation work with it as well. If the design cannot be adapted in some way, it can be tested whether the current design does indeed work.
26	5B and 12B and 13A	For a two-level skewed rotor, shortened-loop methods like in situ or “demag to demount” treatment should be enabled. Please increase the size of the magnet pocket volume to allow volume expansion of the magnet or loosening of the parts when burning the adhesive. If this is not possible, please consider avoiding skewing overall,
27	5C	Skewing with three or more levels is generally a rather unsuitable decision.

Continuation of Table A. 3: Recommendation rules for the interactive design for recycling guideline.

28	5C and 12A and 13B and (16A or 16C) and (14D or 14E or 14F) and 17F	When skewing with three of more levels, suitability of in situ treatments is unlikely but may be possible under very narrow requirements, which you may fulfill with the design. Hydrogen permeability must be ensured and the magnets must be affixed fully mechanically. The "line of sight" of the skew levels must be large enough so that the powder can be shaken out.
29	(5B or 5C or 5D) and (1A or 1B)	Skewing overall limits both quick demounting and shortened processing routes, especially for smaller to medium motors.
30	(5B or 5C or 5D) and (1C)	Skewing overall limits both quick demounting and shortened processing routes, however, large rotors may allow full demounting. Please estimate if that would be economically viable for the value of material you are integrating into the rotor.
31	5D	If possible, avoid skewing. There are options to reduce torque ripple and vibrations to a limited degree by other means, like asymmetric flux barriers, stator skew or using a notched rotor.
32	5B or 5C	Please investigate if for your application, skewing could be replaced by another method to mitigate torque ripple, like asymmetric flux barriers, stator skew or using a notched rotor.
33	6A	Backlack has different advantages aside from recyclability and works for a range of different recycling routes. However, the hydrogen permeability can be limited, which is why dotted glue or interlock would be preferable.
34	6B	Dotted glue offers similar advantages to backlack but with better hydrogen permeability. It is a good choice for most recycling route.
35	6C	Interlock is a solid choice for most designs and recycling routes. It offers hydrogen permeability, a lack of introduced contaminants and a cheap method overall. However, in a design fully targeted to “demag to demount”, backlack or dotted glue may be favorable. In an in situ processing situation, the powder may be get jammed in the magnet pockets, as interlocked stacks can be easily deformed. It is advisable to place the interlocking points to avoid this or to use steel sheets with high stability or to ensure there is enough expansion space in the magnet pocket.
36	6D	Since your stack production method Is not in our catalogue, please check for yourself: Is it going to introduce contamination? Does it re-couple the steel sheets strongly, inducing more rotor losses? Do the steel sheets deform and detach easily? Ideally, none of these questions is answered with yes. Then, you likely have a recycling friendly method.
37	6E	For choosing a suitable stacking method, mostly consider the requirements towards the robustness of the rotor and price. However, the stacking method can also have some effect on the recyclability. Interlocking is a solid choice for all recycling methods.
38	6E and 4C and (14A or 14B)	In your case, where the shaft and the magnets are affixed by adhesive, it would be useful to also use backlack or dotted glue for the stack production. Like this, when thermally demagnetizing, the whole arrangement can fall apart.
39	7A	Using large polymer features makes the rotor unsuitable for thermal demagnetization They have to be removed before demagnetization. If you intend to recycle the rotor via in situ HPMS, which would circumvent the need to demagnetize, make sure the features either don't obstruct the hydrogen and to be removed powder or can be removed easily before adding the rotor to the reactor – and please skip any fillers.
40	7B	Small polymer features can still be critical to thermal demagnetization, as they can melt over the rotor. Please omit them or make them easily removable.
41	7C	Not using polymer features is a great choice.
42	7D	You indicated you're not sure whether you will use any extra polymer features on your rotor. If possible – avoid them. An exception is when you are very sure that in situ HPMS processing is possible for your small rotor – then a polymer feature may be a nice end plate which can (please test it) be removed easily before HPMS.
43	8A	Aligning the magnets tangentially to the rotor surface is a great choice.

Continuation of Table A. 4: Recommendation rules for the interactive design for recycling guideline.

44	8B	Please reconsider if it is necessary to place your magnets in an angled design, for example v-shape, a tangential arrangement is better for several processes.
45	8C	You indicated being unsure whether to use tangential or angled magnets relative to the rotor surface: If possible, go with a tangential design, especially for larger rotors, as it can facilitate demagnetization!
46	9A and 16B and/or (17A or 17B or 17C or 17D or 17G)	Please reconsider if you really need several magnet layers, as it is a hindrance to several different processing routes. If you absolutely must layer, please at least use the same magnet alloy for each magnet.
47	9A and (16A or 16C) and 17F	Generally, layering magnets is not a great idea. However, as you are using a hydrogen permeable passivation (or switch to one), it would be viable if the design is otherwise suitable for in situ treatment.
48	9B	Great choice not to layer your magnets. Facilitates basically every different processing route.
49	9C	You indicated being unsure whether to use several tangential layers of magnets in your stack layout. Please avoid it, if possible!
50	10A	Great choice to use larger magnets with balanced dimensions – less surface area and oxidation, easier removal and general processing
51	10B	It would be good to consider using fewer singular magnets and switching to larger magnets with balanced dimensions – less surface area and oxidation, easier removal and general processing
52	10C	Please check if you could use less, larger magnets, if possible.
53	10D	Generally, fewer and larger magnets are preferable both any recycling route.
54	11A	Using multiple magnets in each magnet pocket is generally a rather unfavorable choice. However, better than segmenting them – in that case, it may even be the better choice.
55	11B	Two magnets per stack are a better choice than multiple, but not the best option. It can enable in situ processing of the stacks when they are demounted from the shaft. It can enable in situ processing of the stacks demounted from the shaft, given that a suitable surface protection and magnet fixation are used.
56	11C	One magnet per stack is a good choice despite skewing. It can enable in situ processing of the stacks demounted from the shaft, given that a suitable surface protection and magnet fixation are used.
57	11D	Using just two magnets along the whole axial length of the rotor is a good combination if both magnets can be accessed from the outside with minimal demounting and if in situ compatible coatings and fixations are used.
58	11E	Using just one magnet along the rotor length is optimal in many scenarios.
59	11E and 1C and (14A or 14B or 14C or 14D)	Using just one magnet would be a good choice in many arrangements, however, having a large rotor and the magnets fixed tightly may lead to breaking. Please test if you can press the magnets out of the pockets unless you are aiming for an in situ treatment.
60	11F	You indicated that you are unsure how many magnets to insert per pocket. The answer is easy: The fewer, the better! Unless you are using several magnets to replace segmentation – then it could be beneficial, depending on the recycling process and the coatings
61	12A	Larger flux barriers can be beneficial to in situ or demag to demount processes, maybe even for pressing out the magnets.
62	12A and (14D or 14E or 14F)	Combining large flux barriers with adhesiveless fixation is a good choice for in situ treatments
63	12B or 12C	Please consider larger flux barriers, if possible. It can be decisive, but generally, smaller flux barriers are not a definite criterium for bad recyclability.
64	13A and (14D or 14E or 14F)	Tight magnet pockets ensure robustness of the magnet fixation. If they are used to circumvent adhesive fixation, this is a very good decision – however, please consider if it is possible to extend the magnet pockets partially if you plan a design that is otherwise well-suited for in situ treatments.

Continuation of Table A. 5: Recommendation rules for the interactive design for recycling guideline.

65	13A	Tight magnet pockets can be problematic for demounting, in situ and “demag to demount”. Please consider extending them at least partially (for example, behind the magnet over, while still holding all edges firmly in place)
66	13B	Great choice to leave enough expansion space in the magnet pockets and, but please don’t fill them with excessive amounts of adhesive. However, if your overall design is well-suited for “demag to demount” treatment, combining large magnet pockets and adhesive fixation are a good choice.
67	13C	You indicated that you are unsure how to dimension magnet pockets. Generally, larger pockets are better, but the positioning of the magnets has to be ensured still. Please don't fill them with too much glue, though.
68	14A	Adhesive fixation of magnets is an ambivalent feature: Adhesive fixation can be incompatible with some in situ arrangements, compatible with others. You can find out more at your local magnet recycler. However, if in doubt, it is better to skip it for this route, especially in small rotors. If you are going for the other shortened processing route, trying to demount the magnets with demagnetization, especially on SPM, adhesive is actually a great choice!
69	14B	Adhesive fixation of magnets is an ambivalent feature: Adhesive fixation can be incompatible with some in situ arrangements, compatible with others. You can find out more at your local magnet recycler. However, if in doubt, it is better to skip it for this route, especially in small rotors. If you are going for the other shortened processing route, trying to demount the magnets with demagnetization, especially on SPM, adhesive is actually a great choice! However, in all cases, fillers are a bad choice. If you absolutely cannot omit using fillers, please use organic ones that disintegrate below 400 °C.
70	14C	Avoid using inmolding for magnet fixation. Inmolding is detrimental to any process and makes the rotor virtually unrecyclable as soon as there is a solid amount of polymer. Please consider another method.
71	14D	Fixing magnets in place via a press fit is generally a rather good choice. If possible, try to make their removal easy. In case you are trying to demount magnets via thermal demagnetization, you may want to consider switching to adhesive fixture without fillers.
72	14E	A mechanical, easy to remove magnet fixture is the perfect choice!
73	14F	You indicated that you are unsure what magnet fixation to use. A mechanical, easy-to-remove magnet fixture is a great choice for every arrangement. If this isn't possible, preferably go with a press fit of some kind. If you want to use a shortened processing route using thermal demagnetization to remove the magnets, glue without fillers is a nice choice as well. Avoid fillers and absolutely avoid inmolding.
74	15A	Great choice for demounting via thermal demagnetization (shortened processing route)
75	15B	A bandage that can be simply cut off easily would be preferable
76	15C	Good choice. The only better choice would be no bandage
77	15D	Perfect choice to fix the magnets in place without adhesive and a bandage, however, they will not fall off with “demag to demount”, but are perfectly suitable for in situ treatment if you would use a suitable magnet coating
78	3B and (15B or 15C)	You indicated you’re using an OR-SPM or AFM – why do you need an additional bandage? Please reconsider if you really need it to save costs and facilitate recycling!
79	15D and (16A or 16 C or 16D) and (17F or 17G)	Affixing the magnets on an SPM without adhesive using a hydrogen permeable passivation only is a brilliant choice for easy in situ processing.
80	15D and 17C or (17F and 16B) or 17G	Affixing the magnets on an SPM without adhesive using an in situ compatible Ni-based coating or a non-hydrogen permeable coating is a great choice. For in situ processing, they merely must be scored to break the coating.
81	15E	You indicated that your method to affix the magnets to the rotor is not listed. Please contact us to elaborate and get a personal evaluation.

Continuation of Table A. 6: Recommendation rules for the interactive design for recycling guideline.

82	15F	You indicated that you are unsure whether to add a bandage to your SPM. In terms of recycling, no bandage is preferable. Please test it thoroughly so the rotor does not explode. If a bandage is needed, a mechanical one would be preferred.
83	16A	Using a hydrogen permeable passivation or similar is an excellent choice in any case, especially if you are not using a coating above it
84	16B or 16C	Good choice to use a passivation or similar. Ideally, it would be hydrogen permeable as well if you're not using a coating!
85	16D	Reconsider using a passivation for better protectiveness during use and end of life, please!
86	16C	Using a passivation would be highly recommended. Ideally, a hydrogen permeable one if no coating is added afterwards.
87	17A	A polymer coating is an ambivalent feature: it can be incompatible with some in situ arrangements, compatible with others. You can find out more at your local magnet recycler. However, if in doubt, it is better to skip it for this route, especially in small rotors. If you are opting for the “demag to demount” route, especially on SPM, a polymer coating is actually a great choice! In situ processing is impossible with this coating at the current state of the separation processes for powder.
88	17B	A zinc coating should be avoided, if possible. It is potentially detrimental to all recycling routes. In situ processing is impossible with this coating at the current state of the separation processes for powder. “demag to demount” and thermal demagnetization and demounting both can lead to excessive Zn uptake into the material. It can be potentially dealt with during recycling, provided it is clearly recognized so the process can be adjusted. If this is not noticed or the processes are unsuitable, there is a significant risk of damaging the sintering equipment of the recycler.
89	17C	Ni based coatings are well suited due to their protectiveness and performance during different shortened processing methods! They can be removed even from powder.
90	17D	A multilayered coating should be avoided. An exception can be one with Ni-Cu-Ni or Ni inside and another layer on top. An absolute no-go should be a Zn-Epoxy multilayer.
91	17E	For evaluation of a special coating, please reach out. The coating is one of the most important factors for recyclability, so please make sure it is compatible.
92	17F	Good choice, if only a passivation is used. Especially, when it is hydrogen permeable. However, make sure the magnets do not excessively corrode during their lifetime – a protective housing can help.
93	17G	You indicated being unsure about the best choice of coating. Ideally, go with a good passivation only. When good protectiveness is needed, Ni-based coatings are a great option. Polymer coatings can be good if used with a shortened processing method like demag to demount.
94	18A	Please reconsider skipping segmentation or going for an uncoated, adhesiveless snakeline magnet.
95	18B	Please reconsider skipping segmentation or at least, don't use filler. If fillers absolutely cannot be avoided, use organic ones that disintegrate below 400 °C.
96	18C	Please reconsider skipping segmentation or at least avoiding fillers.
97	18D or 18E	Skipping segmentation or using an adhesiveless segmentation method is a great choice.
98	18F	Skipping segmentation or using an adhesiveless segmentation method would be a great choice. If it cannot be avoided, please use filler and opt for either no coating or a polymer coating.
99	19A or 19C	GBD is a common choice but can cause issues in HPMS processing. Furthermore, the GBD does not persist during resintering. Avoiding it would be helpful.
100	19B	Great choice to use non-GBD magnets. They can be recycled well and their properties can be restored better.

Continuation of Table A. 7: Recommendation rules for the interactive design for recycling guideline.

101	20A	Choosing a recyclable, grain boundary modified magnet is a good choice. However, make sure it performs well in HPMS.
102	20B	Choosing a non-recyclable grain boundary modified NdFeB magnet makes the entire machine non-recyclable via HPMS. Avoid it or consult long-loop recyclers to check what they can do. Clearly declare it as well, as it could ruin batches of recycled material.
103	20C	Please avoid grain boundary engineering unless you can test if the magnets remain recyclable via HPMS.
104	20D	Avoiding grain boundary engineering is a good choice. Like this, HPMS recycling remains predictable.
105	20E	Preferably, avoid grain boundary engineering. If impossible, test and ensure it is compatible with all relevant recycling treatments.
106	21A	Great choice to use a low corrosion alloy! This may allow not using a coating.
107	21B or 21C	A low corrosion alloy would be a better choice – you may even be able to omit the coating. Carefully consider with a specialist.
108	2A and 3B	Please reconsider if you could surface mounted magnets on your outer rotor – in many cases, the magnets will be held safely enough by it and will be much easier to demount or process otherwise.
109	4A and (5B or 5C) and (17A or 17B or 17 C or 17D or 17 E) and/or 16B	Your rotor is unsuitable for in situ processing due to using a press fit shaft, multiple, skewed layers of magnets and impermeable coatings. This route is currently off limits, which is very unfortunate if you have a small rotor that also can't be demounted via demagnetizing.
110	1A and (17B or 17C or 17D or 17E)	Please reconsider your coating choices – ideally, use a passivation only, no coating. Your current design is likely too small to demount, off limits for in situ processing – your only chance would be a shortened processing route via thermal demagnetization which also demounts the magnets – only if you use epoxy coated magnets, adhesive fixture and the right geometry to get the magnets out.
111	1A and 17C and (14D or 14E) and 9B	Your rotor can be processed by cutting the magnets from the side of the rotor and then in situ processing them, provided, there is enough expansion space (s. magnet pocket and flux barrier geometry). The Ni-based coating can be removed from the powder.
112	1A and 17C and (14D or 14E) and (11D or 11E)	Your rotor can be processed by scoring the magnets from the open ends of the rotor and then in situ processing them, provided, there is enough expansion space (s. magnet pocket and flux barrier geometry). The Ni-based coating can be removed from the powder.
113	1A and 17C and (14D or 14E) and (11A or 11B or 11C)	If you made all magnets accessible from the outside for scoring the coating by avoiding layering the magnets axially, you could aim for recycling via in situ processing – provided, there is enough expansion space (s. magnet pocket and flux barrier geometry). The Ni-based coating can be removed from the powder.
114	1A and 17C and (14D or 14E) and 9A	If you made all magnets accessible from the outside by cutting the magnets from the sides of the rotor by avoiding layering the magnets tangentially, you could aim for recycling via in situ processing – provided, there is enough expansion space (s. magnet pocket and flux barrier geometry). The Ni-based coating can be removed from the powder
115	2B and (15A or 15B)	Great combination of features. When you thermally demagnetize your rotor and remove potential bandages, your magnets should fall off by themselves.
116	2B and 4C and (15A or 15B)	Great combination of features. When you thermally demagnetize your rotor and remove potential bandages, your magnets should fall off by themselves. Furthermore, your rotor may also dismantle itself so you can cleanly separate the shaft and the stacks!
117	(15A or 15B) and 17B	Combining a Zinc coating with adhesive or another organic coating on top is an unfortunate choice. Consider changing either the coating or the fixation. This combination is unsuitable for all processes. The only chance is for medium to large rotors to use a very short thermal demagnetization time, demounting, and then removing the coatings and outer surface layers.

Continuation of Table A. 8: Recommendation rules for the interactive design for recycling guideline.

118	(14A or 14B) and 12A and/or 13B and 2A and (5A or 5B) not 4C	If you're lucky, the magnets can fall out during thermal demagnetization
119	(14A or 14B) and 12A and/or 13B and 2A and 5D not 4C	If you're lucky, the magnets can fall out during thermal demagnetization – provided you are using a maximum of 2 skew levels.
120	(14A or 14B) and 12A and/or 13B and 2A and (5A or 5B or 5D) and 4C	If you're lucky, the magnets can fall out during thermal demagnetization – provided you are using a maximum of 2 skew levels. In fact, your whole rotor may come apart easily.
121	4C and (5B or 5C or 5D)	Using an adhesive-fixed rotor-shaft connection may greatly facilitate demounting after demagnetization, which makes processing skewed rotors much easier. Well done!
122	1C and 17B	Large rotors usually have to be thermally demagnetized to handle them, especially when they are demounted. Zinc coatings are incompatible with thermal demagnetization unless the treatment is kept very short. Please reconsider your coating choice!
123	17C and (10B or 10C or 10D)	As Ni-Cu-Ni coatings can be quite thick, using them on very small magnets can be a high contamination input factor and make mechanical removal of them even harder. Try to keep the coating thin, or the magnets thick please.
124	(10B or 10C or 10D) and (1B or 1C) and 2A	Using smaller magnets, you may encounter issues when demounting by expressing the magnets from the rotor. Try to use larger magnets or to accommodate their removal by allowing to apply only small axial forces to express them.
125	(10B or 10C or 10D) and (1B or 1C) and 2A and 9A	Using smaller magnets, you may encounter issues when demounting by expressing the magnets from the rotor. Try to use larger magnets or to accommodate their removal by allowing to apply only small axial forces to express them. Furthermore, you will have to separate the stacks before expressing them. This is not a good combination of features.
126	(17B or 17C or 17D or 17E or 17G) and (18A or 18B or 18C)	Please absolutely avoid using segmentation combined with metallic coating or multilayer coatings.
127	1C and 14D	As you have a large rotor, the recycler may demount the magnets. Fixing the magnets with a press fit may hinder that. Consider to facilitate the demounting by reducing the required forces to express the magnets.

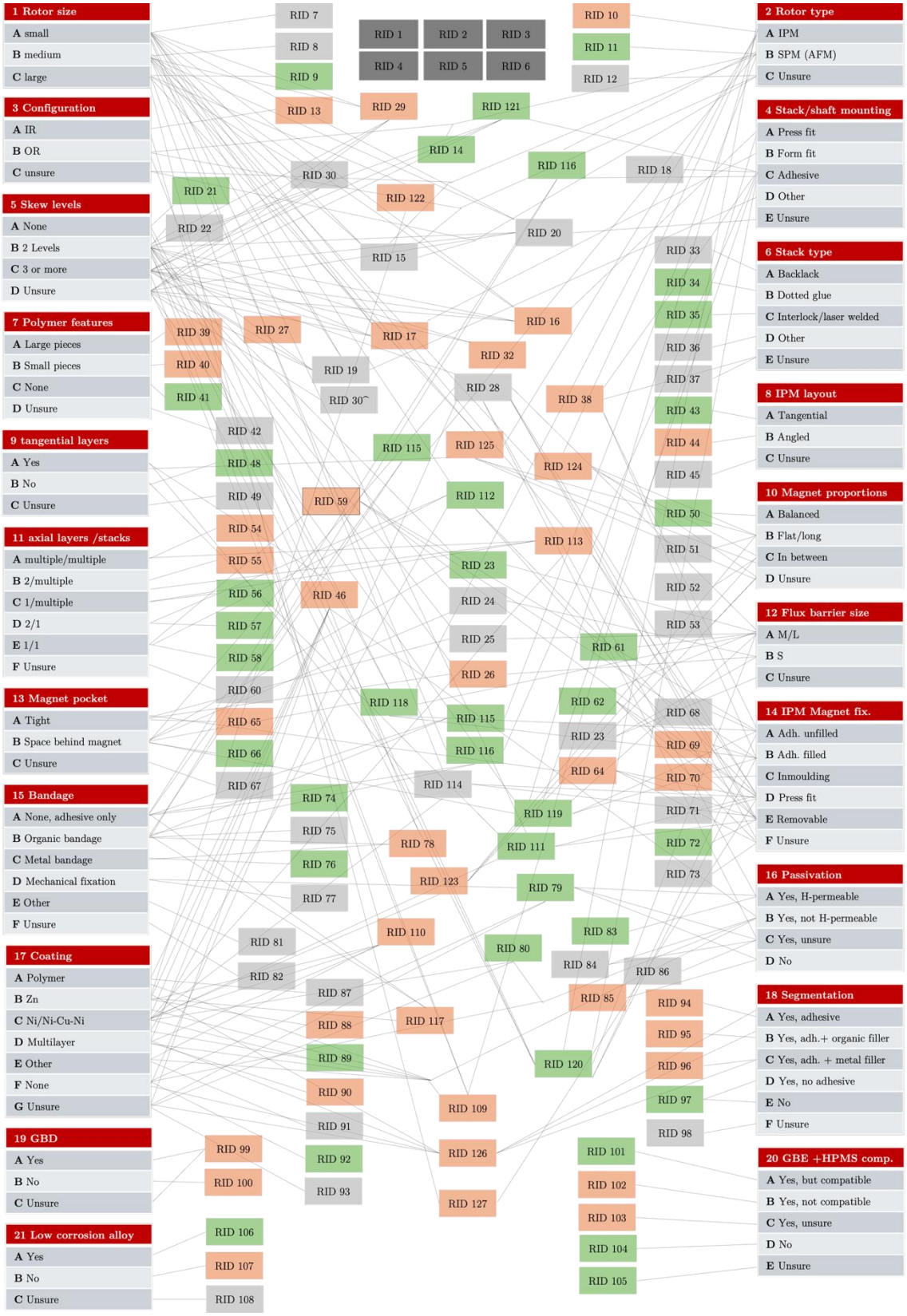


Figure A. 1: Visualization of the questionnaire answers and the recommendation algorithm.

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Biography

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