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A simulation-based exergetic analysis of NdFeB permanent magnet production to understand large systems

ABSTRACT

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Metallurgical simulation and evaluation of the resource efficiency of whole production processes are of key importance for a sound environmental impact assessment. An exergy dissipation analysis is suitable for quantifying the theoretical limits for a process and pinpoint hotspots for improvement along the value chain. The production of NdFeB permanent magnets is evaluated through a simulation-based life cycle assessment and exergetic analysis, comprising 107 unit operations, 361 flows and 209 compounds. This methodology highlights areas with the greatest potential for improvements in terms of technology and environmental impact, shedding light on the true resource efficiency and minimum exergy dissipation for the production of permanent magnets, present in several low-carbon technologies. A maximum exergy efficiency of 60.7% shows that there is a limit for sustainability, which can be improved by technological improvements and recovery of waste streams, showing the inconvenient truth that the resource efficiency will never reach 100%.

Keywords: NdFeB magnet production, resource efficiency, exergetic life cycle assessment, process simulation, geometallurgy, large systems design

1. INTRODUCTION

As we move toward more scarce deposits [1] and complex mineralogy, a clear understanding of particle interconnections and its behavior in a mineral processing and metallurgical plant are of extreme importance for quantification of process and resource efficiency. A geometallurgical approach aims at developing predictive models combining geological and metallurgical data to better understand how the intrinsic characteristics of an orebody affect downstream metallurgical processes [2], where it can be effectively positioned to optimize resource utilization and its recovery [3] and compute the impact in terms of environment, economy, and resource efficiency.

An integrated geometallurgical approach is possible through digitalization techniques, being able to predict metallurgical performance [4], understand material recovery and losses, and aid manufacturers in better designing products through the concept of Design for Resource Efficiency [5]. Digitalization makes it possible to simulate different production scenarios and to identify optimum processes, pinpointing hotspots for improvement and quantifying resource efficiency by benchmarking with best available technologies (BATs) [5–7].

An exergetic analysis of large systems is capable of understanding the resource efficiency and exergy dissipation for each process along the value chain, measuring both quantity and quality of thermal and chemical processes. Combined with digitalization techniques, it provides thermodynamic-validated data to be used for process improvement, offsetting the problem of data availability [8,9], which is a critical issue for traditional sustainability evaluation studies, including life cycle assessment [10–13]. Simulation tools, therefore, differ fundamentally from the more simplified approaches such as material flow analysis and life cycle assessment (LCA).

van Schaik & Reuter [7] state the importance of maximizing resource efficiency by having a deep understanding of the whole value chain, looking at metal recovery and defining optimum product design based on recyclability and resource efficiency. Sciubba [14] correctly states that in a thermodynamic point-of-view, the concept of sustainability is misused, as any open and evolving system is continuously availing itself of exergy supply and entropy is always increasing. Many circular economy experts constantly overlook this problem and oversimplify the concept of circular economy. Markus A. Reuter et al. [15] demonstrated the importance of metallurgical infrastructure in maximizing resource efficiency, while Bartie et al. [16] and Abadias Llamas et al. [17] demonstrated the capabilities of a simulation-based study for large-scale systems in computing the limits for circular economy, having a case study on cadmium telluride (CdTe) photovoltaic technology.

Sintered neodymium-iron-boron magnets have the highest energy density of permanent magnets and are used in multiple applications, including several clean technologies, such as wind turbines, electric vehicles/bikes, air conditioning compressors, among others [18–20]. They support the Sustainable Development Goal by ensuring access to affordable, reliable, sustainable and modern energy for all [21]. However, there is an associated impact for the production of any material, and NdFeB permanent magnets are no exception, as the production process requires large amounts of chemical compounds, electricity and generates radioactive waste [12,13,22]. Hence, the environmental impact and resource efficiency of NdFeB permanent magnets production should be further investigated.

This paper aims at evaluating the environmental impact and resource efficiency for production of NdFeB permanent magnets (rare earth magnets, hereinafter referred to as REM) from a monazite-rich deposit in Brazil, performing an exergetic evaluation in addition to the usual energy analysis of the production process at the unit operation level, from ore to magnet. Figure 1 demonstrates the importance of not only tracking the energy use throughout the whole value chain in large systems but also understanding the exergy and residues dissipation as they build up after each unit operation. There must be a clear strategy and methodology for the identification and minimization of process inefficiencies. This study is the first to thermodynamically understand the complete process chain for REM production, performing a sound simulation-based exergetic analysis coupled with LCA, pinpointing the most critical unit operations in the process and providing information on how to minimize exergy dissipation for large production systems, which cannot be achieved through traditional life cycle assessment alone.

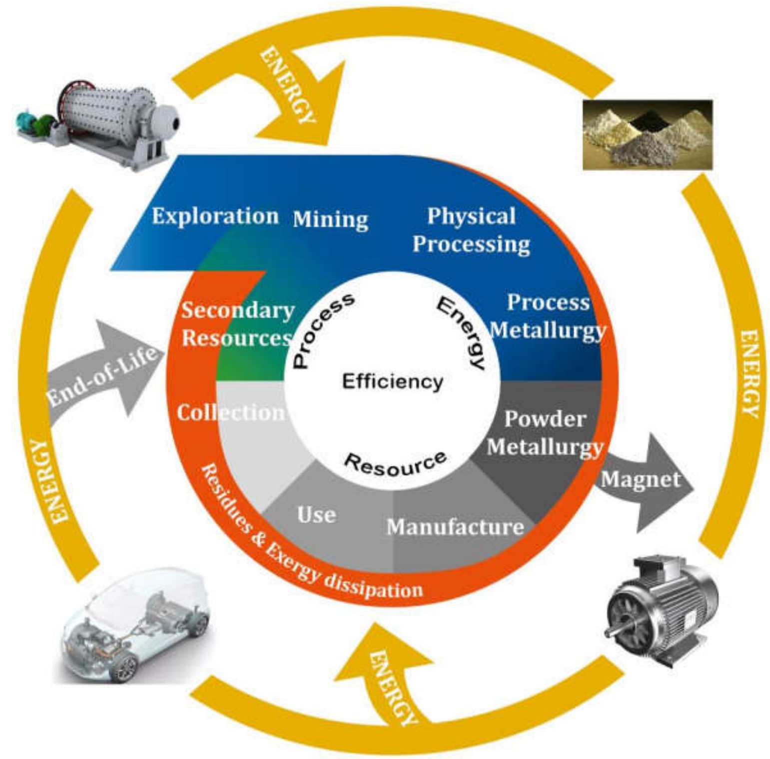

Figure 1. The interconnection of process, energy, and resource efficiency approach for integrated systems assessment, focusing on Nd-Fe-B permanent magnet production, energy requirements, and exergy dissipation. Adapted from Abadias Llamas et al. [17]

2. DESIGN OF A SIMULATION MODEL FOR NdFeB MAGNET PRODUCTION AND RECYCLING

A simulation-based life cycle assessment and exergetic analysis, including the usual energy flow, has been made considering the whole value chain for REM production from a monazite-rich deposit in Brazil, following the simulation-based approach for LCA [6,7]. The methodology makes use of simulation tools to define the optimal route for processes based on energy, resource, and exergy efficiencies.

HSC Chemistry 9.9 [23], a process design software, is linked to GaBi 8.7 [24], an environmental assessment impact calculation software, through an LCA Evaluation tool built in HSC Sim, to validate existing data of metallurgical processes in a scientific and rigorous way by considering thermodynamic and mass balance for all elements entering and leaving the production process [25]. The link connects life cycle inventory data from HSC Chemistry to GaBi. Therefore, the allocation of the environmental impact of upstream materials and energy in the process can be related to the final product itself, regardless of their unit of measure (tph for materials, kWh for energy systems). A critical evaluation of resource efficiency by exergy analysis is performed, in addition to traditional life cycle assessment, by looking at how much exergy has been transformed into actual products. Through digitalization, benchmark processes can be designed and used for comparison of different technologies, having a baseline for comparison and guaranteeing the same system boundaries are used for every study.

2.1. Integrated flowsheet for magnet production assessment: from rock to powder metallurgy

Following literature information on the Brazilian deposit, the main rare-earths-bearing mineral is monazite, at a composition of 4% in the ore [26], with rare earth reserves of 14.20 Mt at 3.02% total rare earth oxides [27]. The distribution of neodymium and praseodymium oxide in the mineral sums up to 20%. Physical separation occurs via low-intensity magnetic separation to remove magnetite, a de-sliming circuit with three stages to remove particles under 5μm and selective flotation of pyrochlore at acidic pH [26,28]. The simulation model was built considering processing parameters available in the literature, such as flotation kinetics studied by Oliveira et al. [29]. The concentrate yields a product containing around 60% Nb_2O_5, while the rare earths report to the tailings and are refined by magnetic separation and flotation to produce a rare earths-rich concentrate.

Hydrometallurgical processes [30] are used to refine the rare earths-concentrate and yield two possible sub-products: mixed rare earth double sulfates ($REE_2(SO_4)_3.Na_2SO_4.nH_2O$) and mixed rare earth hydroxides ($REE(OH)_3$), as described by Krishnamurthy & Gupta (2015). Individual rare-earths are separated by dissolving the hydroxides in hydrochloric acid, followed by a series of mixer-settlers to separate between different rare earth oxides (REOs), precipitation and calcination, yielding four rare earth oxide products: lanthanum oxide, cerium oxide, neodymium + praseodymium oxide and other heavier REOs.

The relatively low melting point of many of the rare earth metals is an advantage [31], so Didymium oxide (a mix of neodymium and praseodymium oxide), is reduced by molten salt electrolysis. Pure liquid Nd + Pr metal is produced by dissolving and electrolyzing the oxide into fluoride-based molten salt - $(Nd, Pr)F_3$-LiF. The didymium metal is further processes by powder metallurgy for manufacturing of sintered NdFeB magnets, following strip casting in inert atmosphere to create a master alloy by pouring alloying metals (Nd, Pr, Dy, Fe, FeB, Fe, Co, Cu, Ga) into an induction furnace [32]. The molten alloy is quenched by centrifugal sputtering [33] to obtain magnet flakes and prevent free Fe formation. The magnet flakes undergo size reduction via hydrogen decrepitation, as hydrogen reacts with the Nd-rich phase forming a hydride, which expands in volume and breaks the alloy along the NdFeB grain boundaries. Further size reduction is performed by jet milling to obtain particles of narrow size distribution of about 5-6 μm [34]. To obtain anisotropic magnetic structures, the powder magnet particles need to be aligned through an external field, followed by pressing to keep the alignment. The pressed powder is sintered under vacuum, which improves the magnet density and further remove gaseous impurities, then annealed to improve magnetic properties, such as coercivity [35]. Final steps are cutting and slicing into the magnet's final shape and electroplating, via the Watts nickel plating solution. Krishnamurthy & Gupta [31] provides an overview of the whole production process for REMs and the study can be complemented by [36,37].

It is important to mention that the simulation model was designed to match the production process and quality specifications for premium magnets for high-temperature applications, having a composition of 30% didymium metal (75% Nd + 25% Pr), 66% Fe, 0.97% Dy, 0.91% B, 1.8% Co, 0.15% Cu and 0.17% Ga.

The system boundary used for digitalization of the complete value chain for magnet production is presented in Figure 2. It has been subdivided into seven sub-processes for designing the integrated flowsheets in the simulation model:

- Mining and mineral processing: from the Nb-rich ore to rare earth concentrate (monazite recovery);
- Acid roasting, leaching, and double sulfate separation: to obtain precipitated rare-earth double sulfate ($Na_2SO_4.REE_2(SO_4)_3.nH_2O$);
- Rare earth hydroxide precipitation and Ce(IV) separation: to prepare the feed for solvent extraction, producing $REE(OH)_3$ and remove Ce^{4+} by thermal oxidation;
- Rare earth oxides separation: performed via solvent extraction, precipitation, and calcination to obtain four products (medium and heavy rare earth oxides (from Sm), didymium oxide (Nd+Pr oxide), cerium oxide and lanthanum oxide);
- Molten salt electrolysis: didymium oxide reduction to liquid Pr + Nd metal.
- Alloy design and powder metallurgy: to obtain the finished coated magnet
- Magnet recycling: performed via hydrogen decrepitation (as described in Section 3.3).

The recycling process has only been used for exergy analysis and quantification of limits for circularity. Life cycle assessment has been made from rock to magnet, having a comparable product system to other literature studies [10,12,13,22,38].

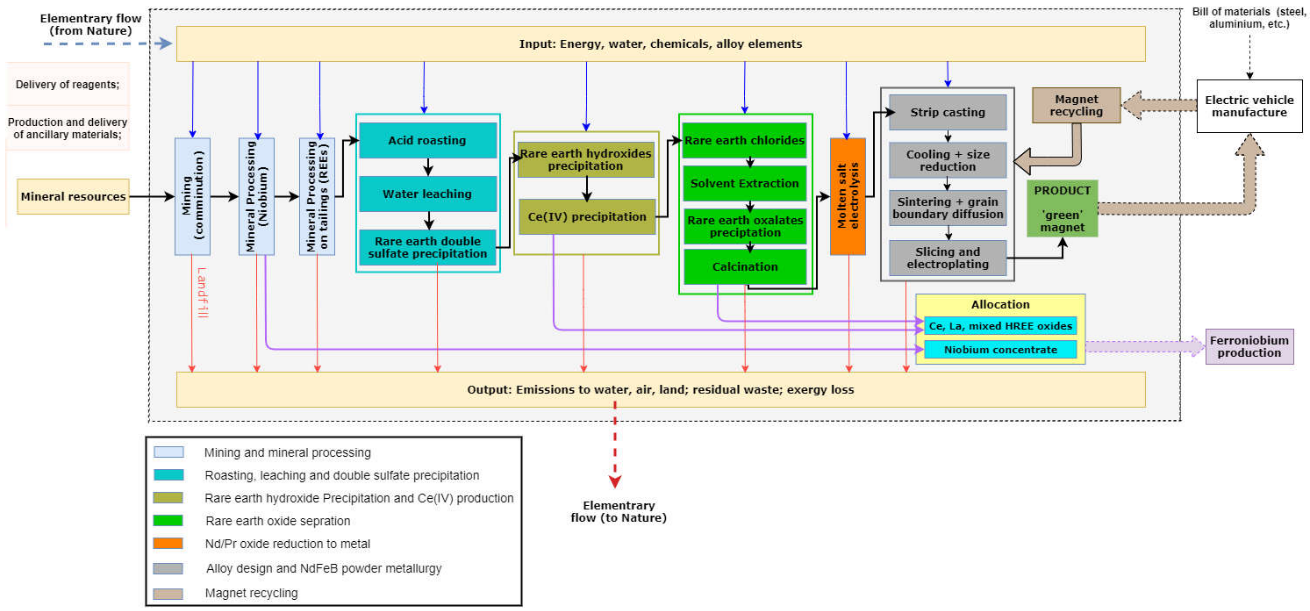

Figure 2. System boundaries for life cycle assessment and exergy analysis on REM production

The complete simulation model comprises 107 unit models, 372 streams and 7 interconnected flowsheet tabs depicted in Figure 3, considering the product and waste flows, in solid, liquid and gaseous forms. Losses are quantified and tracked throughout the whole process, as well as a consistent material and energy flow at the unit operation level is obtained. A screenshot of one out of 7 interconnected flowsheets is presented in Figure 3, exemplifying the simulation for alloy design and powder metallurgy. The remaining screenshots are available in the Supplementary Materials (Figures S.1-S.7). Figure 3 describes the input and output flows for each unit operation, displaying the exergy flow for each stream. On the right-hand side, it is possible to visualize the mass and energy flow for the Strip Casting unit, with its total exergy dissipation. This analysis can be visualized for each unit operation using the Thermoeconomics Calculator tool built in HSC Sim.

The methodology provides a clear understanding of process, energy and resource efficiency for different scenarios, identifying unit processes where most irreversible losses occur, such that optimization strategies can be developed aiming at exergy dissipation minimization.

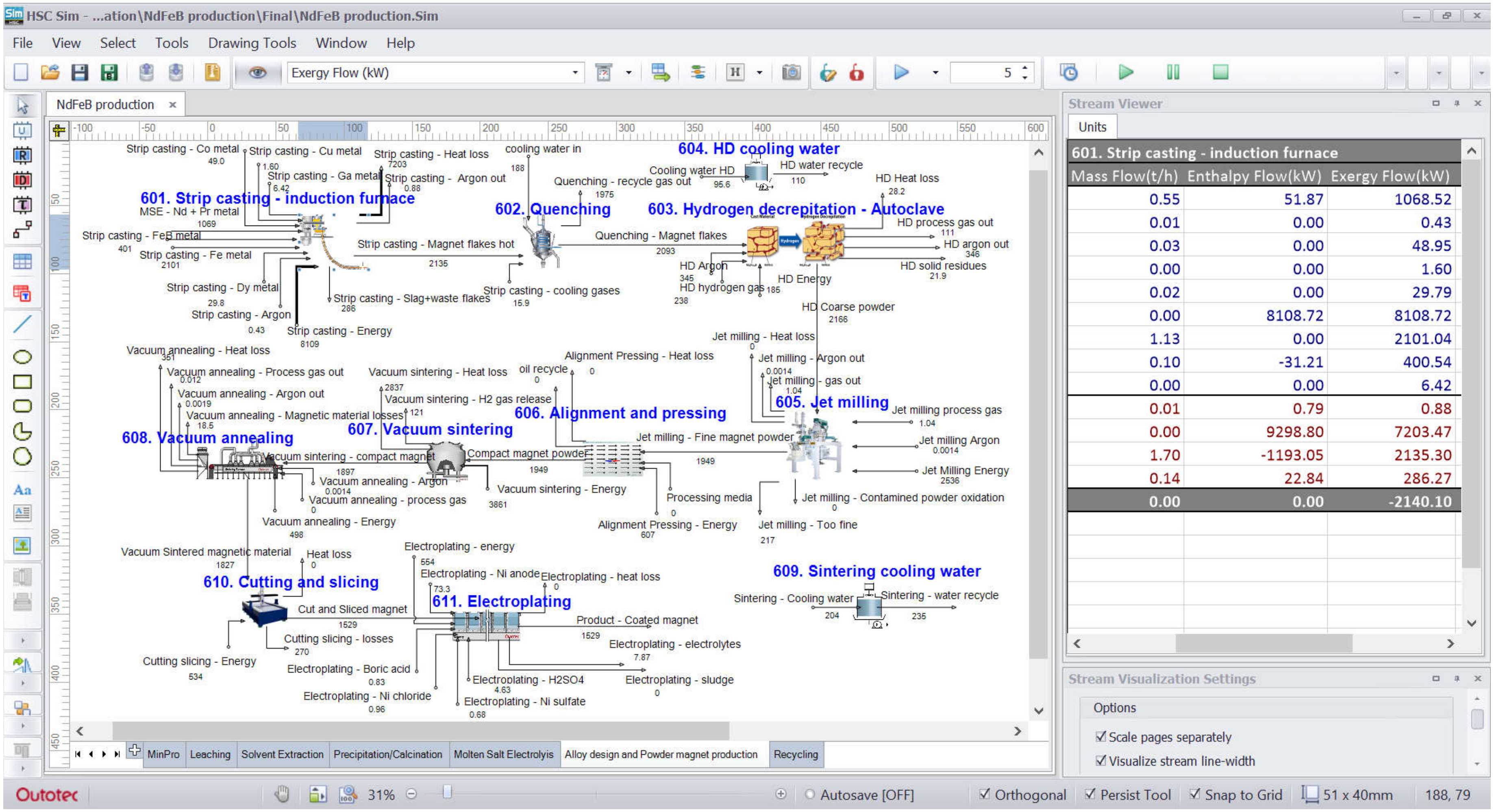

Figure 3. Powder metallurgy flowsheet in HSC Sim. The complete simulation model is comprised of 7 interconnected flowsheets (mineral processing, concentrate cracking, rare earth oxide separation, precipitation/calcination, molten salt electrolysis, alloy design/powder metallurgy, and recycling), 107 unit models, 372 streams, 209 compounds, and 35 elements

2.2. Life cycle assessment and exergetic evaluation of permanent magnet production

Life cycle assessment is carried out according to the best practices and framework described by ISO 14040 [39] and the guidelines on ISO 14044 [40]. It shall clearly state the reasons for carrying out the study, as well as be comparative to other studies, describing the relevant physical flows entering and leaving the system boundary under consideration (elementary flows). It is designed to have a functional unit of 1 kg of NdFeB permanent magnet, suitable for high-temperature applications, such as permanent magnet synchronous motors for electric vehicle applications. The environmental impact analysis is intended to be used as a comparative study to existing literature, to understand the resource potential of magnets produced outside from China and have a more reliable dataset available for magnet production.

Even though Ecoinvent version 3 [41–43] was released in September 2019, the dataset for rare-earths and permanent magnet production remains largely outdated, as the period for data collection on permanent magnet production was from 1995 to 2002 [44]. In addition, the Ecoinvent dataset for magnet production is based on a proxy from aluminum production, which introduces significant deviation. Moreover, the Ecoinvent database does not account for specific metals required for high-temperature applications, such as dysprosium or holmium, cobalt, gallium, and copper.

Therefore, there is a clear need for updating the database for magnet production with information on the best available technologies. A simulation-based LCA and exergy analysis generates reliable data, tracking every energy and mass flow coming in and out of the product system, along with their associated emissions. This is a crucial step regarding permanent magnet production, as data availability is a key issue in uncertainties during LCA, as consistently reported by authors [10–13]. The use of simulation models to create a digital twin of the process, as introduced by Reuter et al. [5] is a great initiative aiming at reducing uncertainties in data collection due to data scarcity, guaranteeing a consistent life cycle inventory and system boundaries.

In this study, HSC Sim 9.9 is used as simulation tool for generating a consistent mass and energy balance, embracing the first and second laws of thermodynamics. The list of material and energy flows coming in and out of the product system is exported to GaBi 8.7 [24], which is used to connect background processes, such as chemical reagents production, electricity generation, and other metals production, to the main REM production process. A screenshot of the interconnection of processes in GaBi is presented in Figure S.8, in the Supplementary Materials.

For exergy analysis, the study considers the resource efficiency and limits for circularity. It measures both quantity and quality of thermal and chemical processes, resulting from thermodynamic imperfections (irreversibilities) generated to obtain a certain product [45]. The total exergy of a stream is obtained from the sum of its physical and chemical exergy, where the physical exergy of a substance is calculated based on the difference in temperature and pressure in relation to its thermodynamic equilibrium state [46], whereas chemical exergy of a substance defines its thermodynamic quality or the exergy of a substance or element at ambient temperature and pressure [47]. In addition, exergy has the advantage of measuring resources in energy units (i.e. kJ or kWh), meaning that material and energy flows can be analyzed simultaneously and resource efficiency of different processes calculated.

The Thermoeconomics Calculator tool developed and implemented in HSC Sim by Abadías Llamas et al. [47] was used in this paper for the calculation of exergy flow cost and irreversibilities in each process step and quantification of resource efficiency. The tool analyses internal exergy losses, resultant from irreversibilities of the process or system being studied, as well as external losses, which appear when waste is rejected to the environment [45]. Internal exergy losses represent the inevitable exergy dissipation occurring in a process, defining the maximum exergy efficiency of a process, whereas external exergy losses can be minimized by reprocessing waste streams.

3. EVALUATION OF LARGE SYSTEMS – CASE ON PERMANENT MAGNET PRODUCTION

3.1. Is life cycle assessment sufficient for a consistent hotspot analysis?

Several authors [10,12,13,22,38,48] have analyzed the production process from ore to magnet, but none of them have performed a thorough simulation model to account for data availability and perform mass balancing of flows. The majority of life cycle inventory data is acquired based on Ecoinvent, other literature studies or discussions with experts. However, these studies only track the product, not necessarily accounting for the waste and all the inevitable losses occurring at every stage along the value chain. In this study, the data for the development of the digital twin was obtained from the actual production process. Furthermore, a simulation-based analysis guarantees that unknown data is calculated through a consistent mass and energy balance, following the first and second laws of thermodynamics, creating a robust inventory of flows entering and leaving the system boundary at the unit operation level.

In this study, the results for life cycle impact assessment (LCIA) are processed in GaBi and are based on a simulation model of the complete value chain, being split into six sub-processes, as described in the system boundary presented in section 2.2. The calculated environmental impact for each LCA category following the CML 2015 methodology is available in Table S.1, in the Supplementary Materials, and the environmental burden share for each impact category is better visualized in Figure 4.

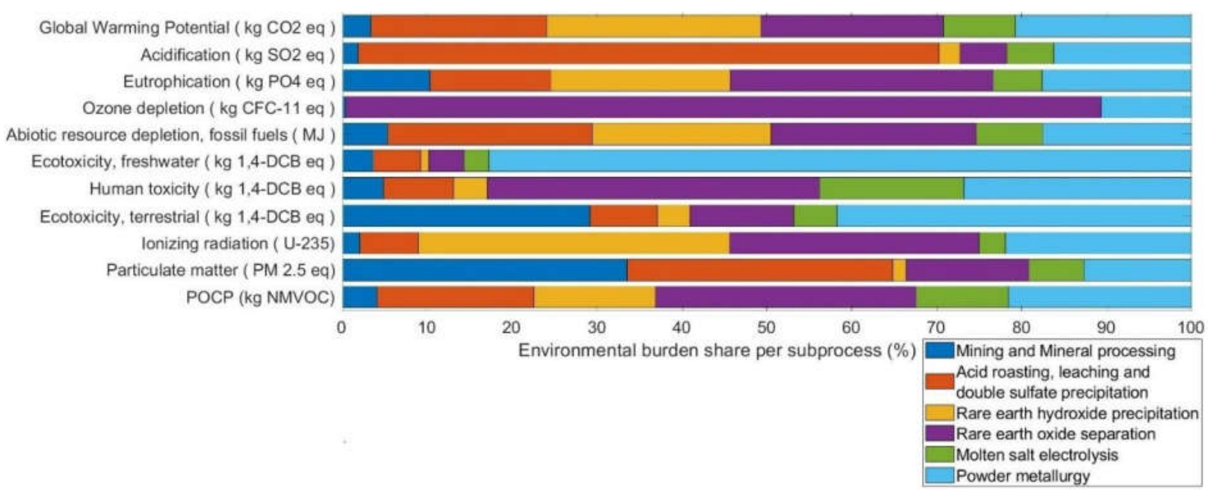

Figure 4. NdFeB permanent magnet production - environmental burden share by sub-process

To understand the main contributors of environmental impact for each LCIA category and which actions could be taken to reduce the impact for each category, the reader is referred to Figure A. 1, in the Appendix. It is important to observe that the electricity grid mix is a critical factor for all of the impact categories, being among the three highest contributors for every category, and generating 27% of the total CO2 emissions. This result is in accordance with previous studies [10,38,49]. For each of the six sub-processes, an overview of the most critical issues is presented:

- Mining and mineral processing: high energy consumption during comminution, together with particulate matter caused by dust generation (impact in particulate matter) and the use of flotation reagents (impact in terrestrial ecotoxicity);
- Acid roasting, leaching, and double sulfate precipitation: high energy and sulfuric acid consumption, with its consequent release of $SO2_{(g)}$, causing most of the impact in acidification potential;
- Rare earth hydroxide precipitation: the consumption of sodium hydroxide generates significant impact in ionizing radiation, GWP, eutrophication and abiotic resources depletion for fossil fuels;
- Rare earth oxides separation: the solvent extraction process consumes hydrochloric acid and organic chemicals, generating most of impact in ozone depletion and human toxicity, together with significant impact in POCP (photochemical ozone creation potential), ionizing radiation and eutrophication. These results are in line with [10];

- Molten salt electrolysis: mostly dependent on energy consumption and its generation;
- Powder metallurgy: high energy consumption and elements required for NdFeB alloy composition (mostly Co, Fe, Ni, and Dy), generating significant impact in ecotoxicity (freshwater and terrestrial), in accordance to [12].

Despite the fact that the sludge stream after leaching and double sulfates precipitation contains radioactive elements, GaBi does not address it as a very significant contribution to ionizing radiation. In addition, the majority of thorium and uranium in the deposit is found within pyrochlore, the main niobium-carrier mineral, so there are pre-existing infrastructure and technology for disposing of radioactive slag/sludge implemented onsite.

It is important to mention that there is also an inherent social impact caused by the exploration of cobalt, used in magnets for high-temperature applications at 1.8% composition by weight. The social impact, however, was not accounted for in this study.

Overall, the most critical processes within the value chain for REM production are powder metallurgy, having high energy consumption (33.28 kWh/kg magnet), and rare earth oxides separation via solvent extraction, having high consumption of hydrochloric acid (7.84 kg HCl per kg of magnet) and organic chemicals. A sensitivity analysis was performed in the shape of a tornado plot, examining the effect of a ± 10% variation over each impact category. Figure A. 2, in the Appendix, presents the sensitivity results for global warming potential (GWP), acidification potential and ionizing radiation, showing the importance of electricity, sodium hydroxide and hydrochloric acid to the environmental impact for REM production.

However, there is an intrinsic difference in tracking energy consumption and exergy dissipation, as exergy deals with the quality of energy used in the process, determining the irreversibilities associated with it. For global warming potential, for instance, traditional LCA shows that energy consumption is a critical aspect, and it points out which processes consume most energy along the process. However, it is not capable of determining the irreversibilities generated in each process, how to change the operation of any reactor in the flowsheet or describe which processes have the highest potential for improvement in terms of exergy efficiency. Therefore, in order to truly optimize a process, it is necessary to understand which process is dissipating most exergy rather than only considering and fixating on the process that is consuming most energy. Therefore, a more complete analysis of the system can be made that includes both the first and second laws of thermodynamics.

3.2. Exergetic analysis of REM production

The integrated simulation model comprises exergy flow, measured in kWh/h, for every stream in the flowsheet, demonstrating state-of-the-art results for resource efficiency that goes beyond life cycle assessment. Exergy dissipation (as represented in Figure 1, Section 1) is a measure of thermodynamic quality loss, representing the occurrence of irreversible processes as equilibrium is reached in a system. This exergy cost encompasses both material and energy flows and brings them to the same energy unit (i.e. kJ or kWh), and it is used to understand how to minimize exergy dissipation by looking at individual processes and reactors. Figure 5 presents the actual and maximum exergy efficiency for the REM production, representing the potential resource efficiency improvement for the process. It is important to mention that the maximum achievable exergy efficiency is related to the process designed in the simulation model and, as new technologies are developed, these limits can be shifted.

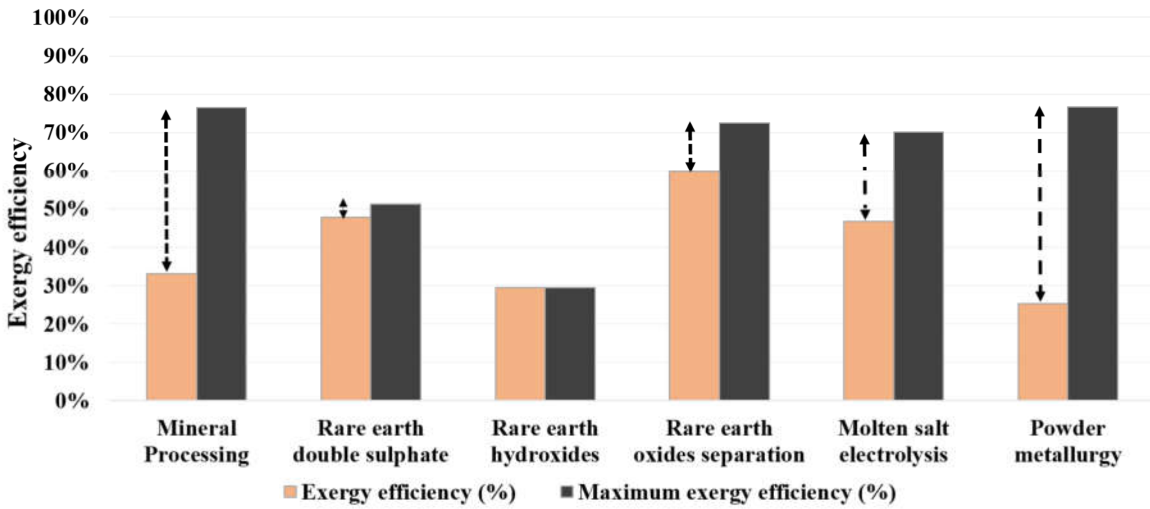

Figure 5. Current exergy efficiency and potential resource efficiency improvement for the system. The maximum exergy efficiency represents a theoretical state where all waste streams are used as useful products

From Figure 5, it is important to understand how to bring the current exergy efficiency of a process to its maximum achievable exergy efficiency, by minimizing exergy dissipation. Systems for heat loss recovery were not implemented in this simulation model, representing the biggest exergy savings if excess heat is converted to useful energy. For mining and mineral processing, there is a potential to reprocess the tailings, which contains iron oxides (magnetite, goethite), barite, gorceixite, ilmenite, and silicates. The overall exergy efficiency for the production of REMs is 60.7%. The thermodynamic quality of resources input to the process is degraded, meaning that entropy is always created (exergy is destroyed), as stated by the second law of thermodynamics.

Not only energy inefficiencies are relevant in a resource efficiency study, but also the thermodynamic dissipation of chemical compounds plays a major role in the total irreversibility created. Note that there is a small gap between the maximum and actual exergy efficiency for the rare earth hydroxide sub-process, meaning that there is little potential for improvement unless there is a change in technology. The irreversibility created in this process is mostly generated due to thermodynamic dissipation of higher exergy content chemical compounds (rare earth double sulfates + sodium hydroxide) being converted into lower exergy content products (rare earth hydroxides + sodium sulfate) through an exothermic reaction [50]:

$$REE_2(SO_4)_3.Na_2SO_4.nH_2O + 6NaOH = 2REE(OH)_3 + 4Na_2SO_4 + nH_2O \quad (1)$$

This reaction dissipates 2.27 MW of exergy, being characterized as internal exergy losses due to chemical exergy losses, which are not reversible.

The analysis shows that exergy dissipation during acid roasting and double rare earth precipitation (9.07 MW) is even higher than powder metallurgy (5.88 MW) (more details in Figure 6) and that an alternative technology should be investigated due to its low theoretical maximum exergy efficiency (Figure 5) and small gap between actual versus maximum exergy efficiency (untapped exergy efficiency). Additionally, exergy analysis shows that there is a high potential for improvement by tackling the rare earths concentrate grade, thus optimizing the acid roasting process. However, when estimating and analyzing immediate improvements, the untapped exergy efficiency might be a more valuable metric than the magnitude of exergy dissipation, as it shows the potential for improvement without major changes in the process chain.

3.3. Exergetic analysis on the circularity of permanent magnets

The exergy dissipation analysis is performed over the whole value chain for magnet production, including the manufacture of permanent magnet synchronous motors (PMSM) for electric vehicle applications, obtained from [48,51–53], in which magnets represent 1.67% of the total mass and steel represents over 70% [48]. The recycling of collected end-use magnets (see system boundaries in Figure 2, section 2.2), was performed via hydrogen decrepitation, according to publications by Zakotnik et al. [54] and Walton et al. [55]. This specific recycling method was selected for analysis because it introduces the recycled material back into the system further downstream. It is based on a commercial-scale recycling process, starting from harvesting (semi-automated dismantling) to separate the magnets from the end-of-life vehicle. The magnet with some impurities is heated up to 400ºC for demagnetization, and the magnets can more easily be separated from steel. The magnet surface is cleaned for Ni removal by abrasive jet and immersion in a hot bath containing diluted nitric acid, followed by hydrogen decrepitation, degassing and jet milling to reach 4µm. Nickel coating peels away from the surface of the magnet as thin sheets. The recovery of REEs during recycling is over 90% and the total amount of virgin rare-earth material (fresh Nd and Pr) used in the process is less than 5%, reducing the energy consumption and mining impacts of this activity [38,54]. The collection infrastructure is not accounted for in this study, as the aim is to understand the exergy efficiency of a recycling route for magnets.

The total exergy flow through the entire system is presented in the Sankey diagram in Figure 6, showing the amount of resources entering (fuels) each subsystem, of which some are transformed into final or intermediate products and inevitably, some resources are irreversibly lost. The exergy flows are given in megawatts (MW) and the results correspond to a flowsheet producing 1.5 t/h of NdFeB permanent magnets.

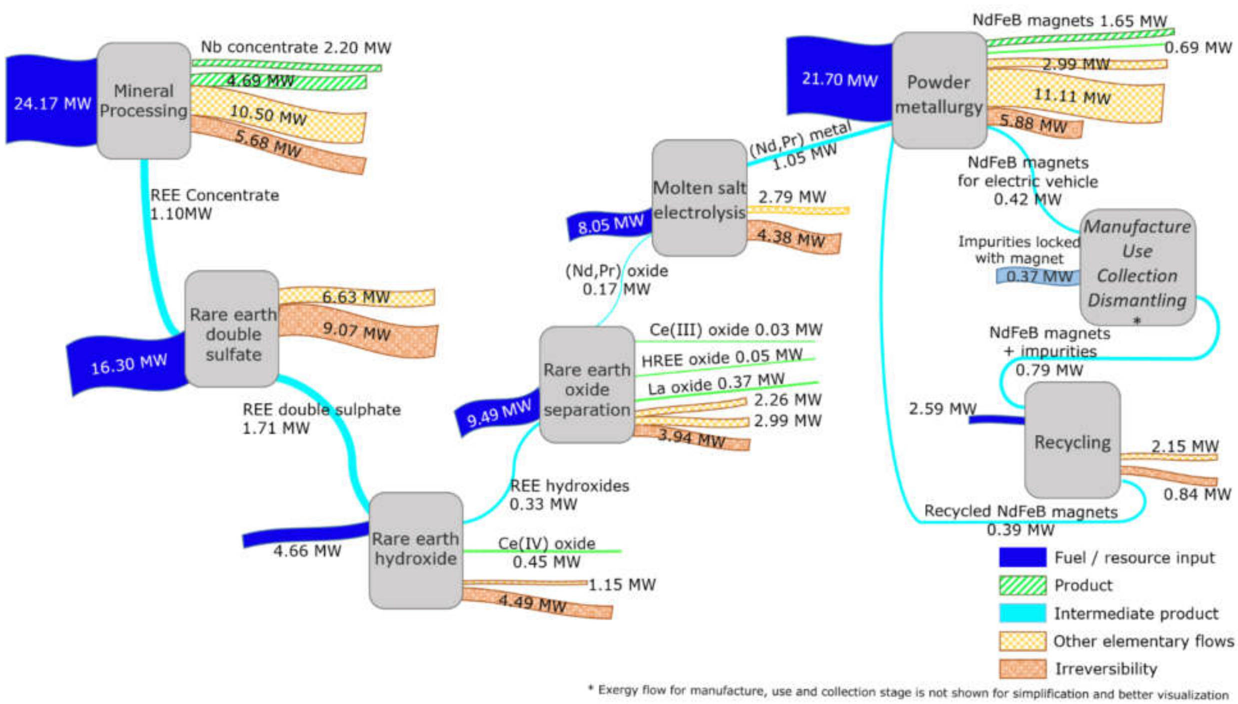

Figure 6. Sankey diagram of exergy flows for the metallurgical production of REMs, use phase for electric vehicle motors and their recycling: a summary of the results of all the interconnected flowsheets subdivided into the show processing groups

The Sankey diagram shows that the recovery processes come at the price of inevitable exergy dissipation or resource consumption, regardless of whether the product comes from primary or secondary resources. Still, the exergy efficiency for magnet recycling is 75.4%, being significantly higher than the maximum achievable exergy efficiency for primary production (60.7%).

Considering the whole value chain, the unit operations with the highest exergy dissipation (top five) and their magnitude are listed:

1. Acid roasting (exergy dissipation of 7.15 MWh/h), as high temperatures for roasting are required (~210 ºC for monazite [31]), as well as excess sulfuric acid consumption, leading to a very resource-intensive process;

2. Molten Salt Electrolysis (exergy dissipation of 4.10 MWh/h), as a very energy-intensive process at high temperatures (~1050 ºC) for melting rare earth oxides;
3. Jet milling (exergy dissipation of 2.63 MWh/h), used for size reduction of NdFeB magnet flakes to obtain a narrow size distribution of small particles (4-6 µm [34,56]);
4. Conversion from rare earth double sulfates to rare earth hydroxides (exergy dissipation of 2.27 MW, as discussed in section 3.2);
5. Strip casting (exergy dissipation of 2.14 MW), due to high temperatures to melt alloying elements.

The capability of a simulation-based exergetic analysis for evaluating large systems and being able to look at the unit operation level is of great value, pinpointing which processes deserve immediate attention. The exergy dissipation analysis of large systems is a powerful indicator for quantifying the consumption of resources, in terms of quantity and quality.

3.4. Sensitivity analyses

3.4.1. Electricity grid mix dependency

It was observed that electricity consumption and its production are extremely relevant to the environmental impact in the life cycle analysis of permanent magnets. Therefore, it is important to understand the impact of different electricity production technologies over a product's life cycle. Figure 7 shows the variation in global warming potential, acidification potential, particulate matter and ionizing radiation as the electricity production is simulated for different countries and electricity grid mix.

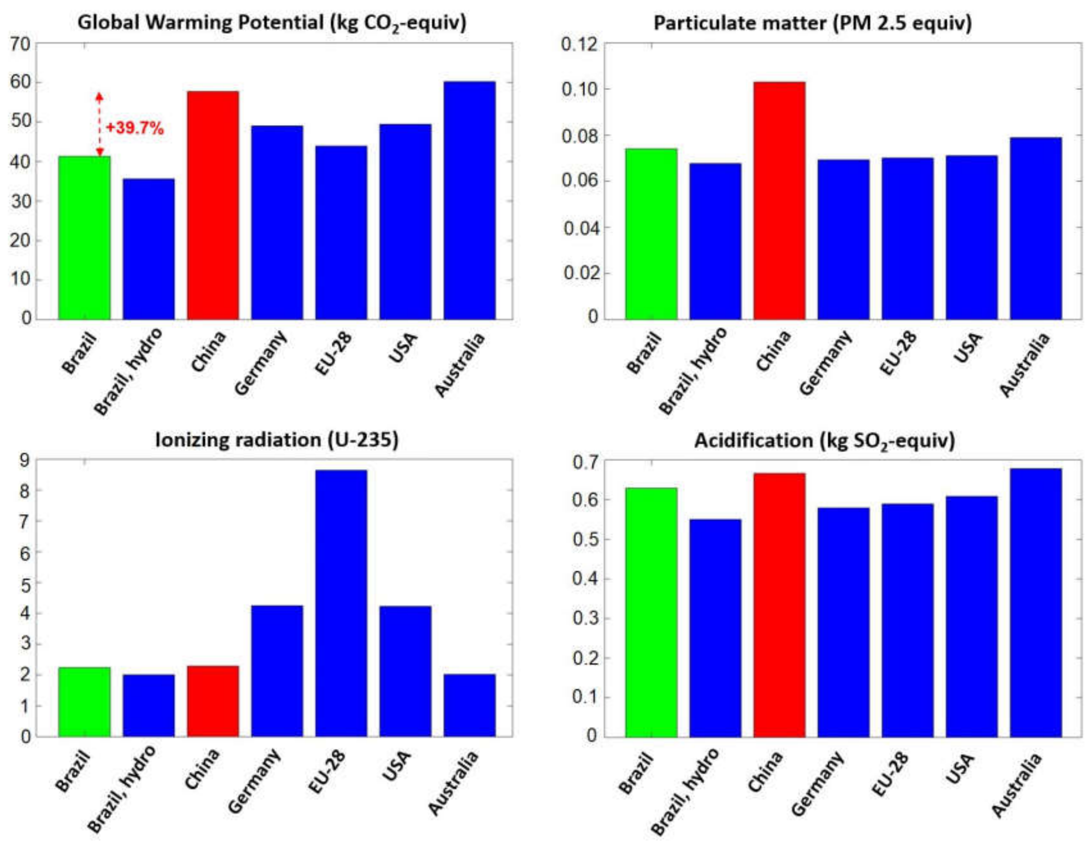

Figure 7. Environmental impact dependency on electricity grid mix (based on different nations and electricity generation technologies): the link mainly to the energy consumption and thus the first law of thermodynamics

The data regarding the electricity grid mix used in this simulation is gathered directly from GaBi. An overview of the current electricity generation technology for each country (Brazil, China, Germany, United States, Australia, and the European Union, EU-28) is given on Figure S.9, in the Supplementary Materials, obtained from the International Energy

Agency - IEA (2019), regarding years 2017 and 2018. The Fraunhofer ISE (Institute for Solar Energy Systems) has made public an updated version for the electricity generation share for Germany in 2019 (available on https://www.energy-charts.de/energy_pie.htm).

Whereas Brazil has 62.93% of its electricity produced by hydroelectric energy, other countries rely largely on non-renewable and carbon-intensive sources. China relies 67.90% on coal-fired power plants, the USA relies 62.41% on coal and natural gas and Germany has 50.25% of its electricity produced by coal and natural gas. Considering every member of the European Union (EU-28), 25.15% of electricity is produced from nuclear power plants and 41.62% is produced from coal and natural gas. Therefore, global warming potential for production in Brazil is 39.7% lower than in China, considering the same production process and system boundaries. The ionizing radiation for production in Brazil is also among the lowest, as the share of nuclear power plants in the Brazilian grid mix is not significant.

Previous studies [38] have stated that electricity generation in a hard coal power plant has the highest impact on the production of magnets originating from rare earth deposits in China. These results are in line with what has been observed in the simulation study, as electricity source is the most sensitive parameter on the environmental impact for the production of REMs (see also Figure A. 2, in the Appendix). In addition, these results are in line with other studies [13,22]. Other important parameters that differ for each geographical location include transport distances, material supply, waste treatment, environmental regulations [13] and ore composition. The latter is of critical importance for the determination of the environmental impact and exergy efficiency for the production of magnets.

3.4.2. Ore composition and mineralogy of REE-carrier

A geometallurgical approach aims at understanding the impact of ore variability over downstream processes (physical and chemical separation processes), being able to calculate resource efficiency as the ore/mineralogy changes. At Bayan Obo, in China, rare earth elements (REEs) are found within bastnäsite, a carbonate-fluoride rare-earth mineral, and monazite, a phosphate mineral, at a ratio of 65:35, respectively [13,58], whereas at Mount Weld, Australia, and at the Brazilian deposit modeled in this study, the ore contains monazite [26]. The presence of monazite as the main rare-earths-carrier mineral generates no CO2(g) and HF(g) emissions during acid roasting, which is a relevant issue whenever the ore contains bastnäsite, a carbonate-fluoride rare earth mineral. Sprecher et al. [12] state that the generation of HF(g) represents the highest impact over normalized environmental impact. These results show the importance of having a geometallurgical understanding of a product system, that incorporates the effects of mineralogy on downstream physical and chemical processes. This would be the authors' understanding of geometallurgy i.e. the predictive and simulation-based analysis of Figure 1 in its entirety.

The exergy dissipation analysis presents more industry-relevant information in understanding the impact in terms of process and thermodynamic efficiency as the ore composition changes. To show the impact of varying mineralogy over a production process, a sensitivity analysis was made considering that the main rare earths-carrier minerals as either bastnäsite, monazite or a combination of both (Figure 8).

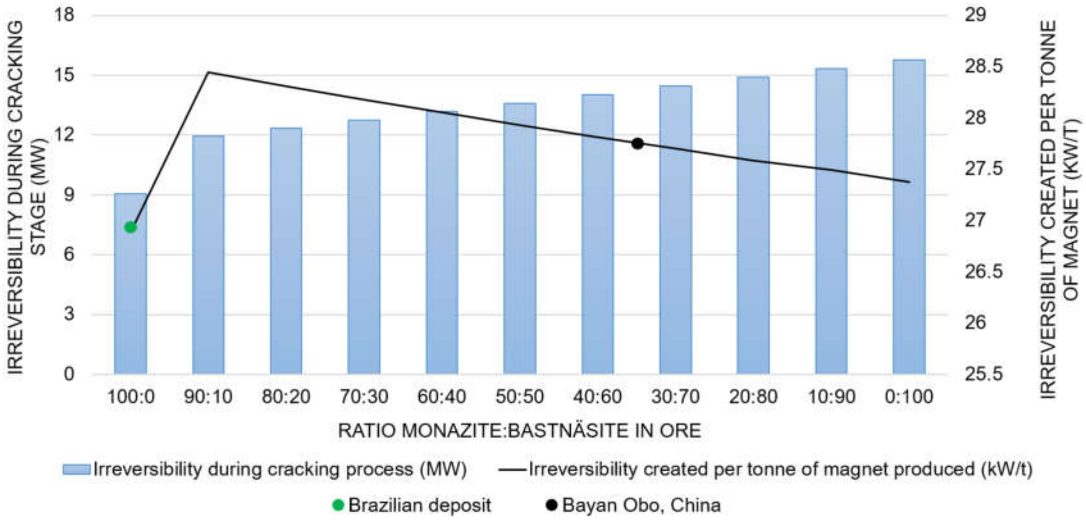

Figure 8. Irreversibility variation due to ore composition: main REE-carrier as carbonate-fluoride vs phosphate mineral: a true analysis of the system under the geometallurgy paradigm

There is higher exergy dissipation in the cracking stage as the share of bastnäsite in the ore increases, mostly due to higher temperatures needed for dissolution and higher acid consumption. However, looking at large systems and considering the exergy dissipation per tonne of permanent magnet produced (purple line in Figure 8), it can be observed that this relationship is not increasing linearly with bastnäsite content. As bastnäsite is introduced into the system, requiring higher temperatures for cracking, there is a sharp increase in exergy dissipation per tonne of magnet. As the ratio of bastnäsite increases, there is an improvement in the exergy efficiency per tonne of magnet, which can be explained by the fact that the content of rare earth elements in bastnäsite is higher than in monazite, leading to an increase in magnet throughput, considering the same amount of ore. Nevertheless, the highest exergy efficiency is related to a scenario with 100% monazite as REE-carrier in the ore, as it currently occurs at the Brazilian deposit.

Figure 8 depicts that there is a relevant impact associated with the intrinsic ore characteristics of each deposit. Therefore, there is a need for properly quantifying and understanding the mineralogy and texture of the ore and how it affects the downstream processes in terms of exergy dissipation/efficiency. The importance of automated mineralogy in the future will thus only increase as key to understanding the true losses from the system [59] while highlighting the effect of the mineralogy on the resource efficiency of the system incorporating all interconnected elements as visualized by the Metal Wheel [15]. There is a potential for improvement during acid roasting, as it is a highly exergy dissipative unit process (18.5% of total irreversibility creation), and this potential for improvement should be tackled.

4. CONCLUSIONS

This paper shows that simulation of large production systems is possible, which incorporates a thermodynamic evaluation of their resource efficiency, both in terms of the first and second laws. Projects can be evaluated prior to their implementation, by understanding the impact of different technologies, different ore composition or energy sources, even though some steps may not be proven technology yet, such as the selected decrepitation process for the magnets.

It demonstrates one of the benefits of digitalizing complete production chains in a single platform, having the same inventory list and system boundaries, consistently tracking material recovery and losses. In addition, simulation can handle the task of updating inventory databases for processes and products' life cycle, which is a critical issue for performing sound life cycle assessments for which little or no information exists, besides being capable of evaluating specific production scenarios, in contrast to traditional LCA databases.

The minimum exergy dissipation for the production of REMs was calculated, considering the consumption of metals, energy, water, and chemical compounds. For any product system, it is important to understand that there are technological limits, which must be accounted for and used to set realistic sustainability goals defined according to the laws of thermodynamics. For REM production from the Araxá deposit in Brazil, the limit for exergy efficiency has been calculated as 60.73%. Recycled magnets have greater exergy efficiency (75.2%) than magnets produced from primary resources, showing the importance of improving collection infrastructure and having manufactures think carefully about product design to support improvements in dismantling and separation of magnets from scrap.

From life cycle assessment results, it was demonstrated the importance of carefully considering which metals to input in a product. The addition of cobalt to magnets generates an impact on social and environmental levels, besides being considered as a critical raw material by the European Union [60]. Therefore, manufacturers should have a clear strategy for product design to assist in reaching higher resource efficiencies during recycling processes.

For magnet production, acid roasting has been identified as a source of significant exergy dissipation, meaning that chemical reagents consumption should be reduced. One alternative is to further decrease impurities content on rare earth concentrate using physical separation methods, thus improving the REO grade and reducing the input feed for acid roasting. This solution would bring benefits in terms of environmental impact, resource consumption, and economics.

Electricity consumption is a major source of environmental impact on magnet production and inevitably for many other critical materials. Policymakers should foster renewable energy transition, improving the sustainability of products manufactured in every country. Brazil has 78.99% of its electricity generation coming from renewable sources, whereas China has only 25.55% of renewable energy in their electricity grid mix (data for the year 2017, extracted from International Energy Agency - IEA (2019), summarized on Figure S.9, in the Supplementary Material). This presents an advantage of magnets produced in Brazil over those produced in China in terms of environmental impact.

In addition, every mineral deposit is unique and it is of extreme importance to take the ore characteristics and mineralogy into account when analyzing the resource efficiency of a product system. Geometallurgy builds the bridge from mineral characterization and its impact on downstream processes, being crucial to quantifying the exergy dissipation throughout the process. The ore characteristics at the Brazilian deposit leads to lower energy consumption during comminution and higher exergy efficiency during acid roasting, when compared to Chinese production from Bayan Obo.

Overall, it has been demonstrated that simulation of integrated flowsheets is a powerful tool to assist in the selection of optimum technologies, looking not only for environmental impact assessment but also on resource consumption. Thermodynamic efficiency limits can be quantified for large-scale industries and the minimum exergy dissipation established. Despite being considered critical for many green technologies, there is an inherent impact caused by the production of permanent magnets. The exergy dissipation throughout the process (Figure 1), i.e. from resource body to product and recycling can be reduced by shifting towards cleaner energy sources and developing new technologies to further recover waste streams and minimize losses.

The illustrated detailed simulation-based approach should become the default to understanding the circular economy and its true resource efficiency based on the first and second laws of thermodynamics and the chemistry in the complete processing chain.

DECLARATION OF INTEREST

The content disclosed in this paper has no conflict of interest.

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APPENDIX

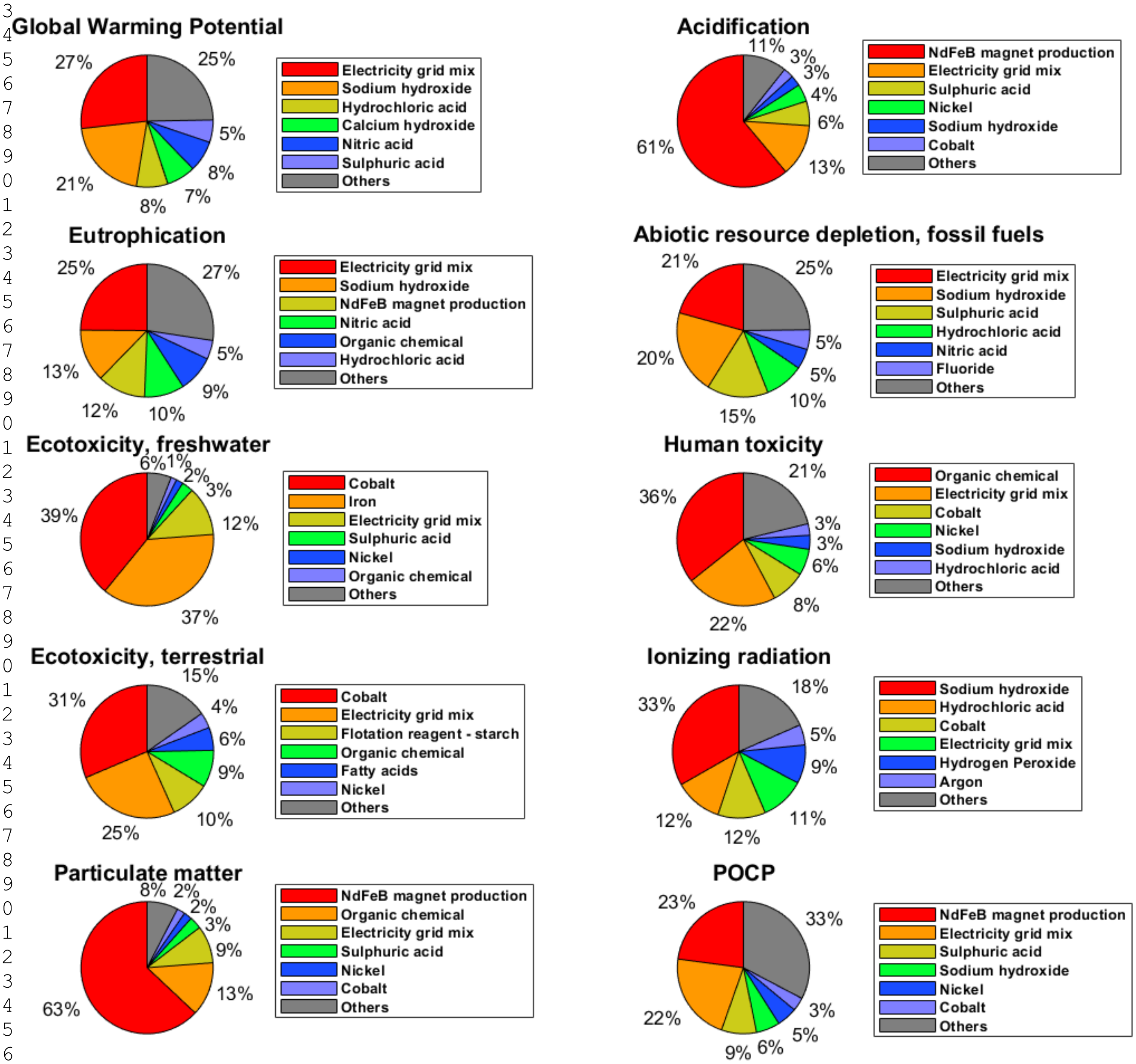

Figure A. 1. Contribution of processes and resources used (material and energy) to various impact categories (six major sources presented)

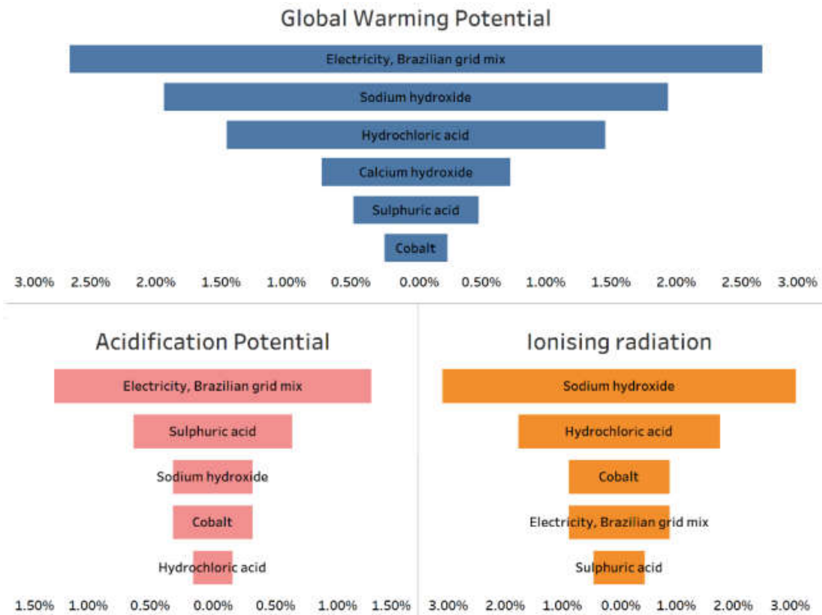

Figure A. 2. Sensitivity analysis for REM production on global warming potential, acidification potential and ionizing radiation