

Article

Sustainable Strategies for the Exploitation of End-of-Life Permanent Magnets

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Abstract: The growing production of green technologies (such as electric vehicles and systems for renewable electricity production, e.g., wind turbine) is increasing the rare earth element (REE) demands. These metals are considered critical for Europe for their economic relevance and the supply risk. The end-of-life permanent magnets are considered a potential secondary resource of REEs thanks to their content of neodymium (Nd), praseodymium (Pr) or dysprosium (Dy). The scientific literature reports many techniques for permanent magnet recovery. This work used a life cycle assessment (LCA) to identify the most sustainable choice, suggesting the possible improvements to reduce the environmental load. Three different processes are considered: two hydrometallurgical treatments (the first one with HCl and the other one with solid-state chlorination), and a pyrometallurgical technique. The present paper aims to push the stakeholders towards the implementation of sustainable processes for end-of-life permanent magnet exploitation at industrial scale.

Keywords: permanent magnet; rare earth oxides; recycling; circular economy; environmental sustainability; life cycle assessment



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1. Introduction

The objective of the European Union is to achieve the climate neutrality. Between 1990 and 2018, the European greenhouse emissions are reduced by 23% despite an industrial growth of 61%. The aim is to reduce the greenhouse emissions by 60% by 2050 [1]. More than 75% of the European greenhouse gas emissions are due to the energy production and extraction/processing of materials, fuel and food [1,2]. The main ways to reduce these sector impacts are the increase of energy production by renewable resources (mainly offshore wind) and the use of hybrid and electric cars to reduce the fossil fuel dependence, with the reduction of 90% of transport emissions within 2050 [1].

The world market of hybrid/electric cars and offshore wind turbines is predicted to reach approximatively 150 million vehicles and 2500 GW, respectively, within 2050 [3]. These green technologies require the use of permanent magnets, such as ferrite, alnico, samarium-cobalt and neodymium-iron-boron (Nd-Fe-B) [4]. Permanent magnets generate electric and mechanical energy by a magnetic field, without the need of electric power [3]. Nd-Fe-B magnets are considered as the strongest and the best permanent magnet available since 1984 to today [5–7]. These magnets are the third generation of permanent magnets and they can produce a maximum electricity power of 512 kJ/m³ [5,6,8]. The main problem of these magnets is the low resistance to high temperatures (Curie temperature stands around 312 °C). To increase both the temperature stability and the corrosion resistant, cobalt (Co), praseodymium (Pr) and dysprosium (Dy) are integrated within the material [4,6]. It is estimated that we will need around 150,000 tons and 1–2 million tons of permanent magnets to cover the future demand of electric vehicles and wind turbines, respectively [3]. Nd-Fe-B magnets are composed of 30 wt% of rare earth elements (REEs) and around 1300 tons (16% of the REE produced) were used for wind turbines in 2010 [5,9]. The REE production is considered critical for European Union, since around 90% of these elements is mined in

China, that produces around 105,000 tons/year of REE. The unique properties of REE make them irreplaceable [10,11]. The production of one ton of REE generates around 1000 m³ of waste gas (containing CO₂, SO₂ and HF), 75 m³ of acid wastewater and 1 ton of radioactive waste (mainly thorium and uranium) [12,13]. For this reason, companies and countries start to study and develop programs for the exploitation, substitution and recycling of these elements. It is estimated that, from 2030, the end-of-life permanent magnets will be the main REE resource, implementing the circular economy pillars [5,10,14]. Nd-Fe-B permanent magnets have a life cycle of 2–30 years, based on the specific application, shorter for electronic devices (e.g., hard disk drives) longer for wind turbines [5]. These permanent magnets could be easily dismantled, refurbished and reused, but, in this way, they show lower performance than the virgin material [9]. For this reason, several researches study the REE recovery from Nd-Fe-B permanent magnet using mainly pyro- or hydrometallurgical processes, but none is applied on industrial scale in Europe, yet [5,10]. The main pyrometallurgy techniques are chlorination [15–17] or sulfation roasting [18], liquid metal extraction with Mg [19], Ag [20] or Cu [21], molten extract with chloride [22], fluoride salts [23], B₂O₃ [24] or CaO-SiO₂-Al₂O₃ [25]. These approaches require high temperature (between 700 and 1500 °C) and the main disadvantages are: the low product purity due to the presence of organic materials (e.g., adhesive or machining oil), different composition of the metal alloys, generated from the starting permanent magnet, and the loss of REE due to their affinity for oxygen [5,9]. On the other hand, hydrometallurgical approaches require lower temperature than pyrometallurgy, but they need the use of chemical agents that produce acid wastewater [4,11,26,27]. The main chemicals used for the REE recovery from permanent magnet are sulphuric [28–32] and hydrochloric acid [9,32–35].

In this work, three different processes are studied by the life cycle assessment approach to identify the best choice for the REE recovery and recycling of permanent magnets. Two hydrometallurgical treatments, with different resources of chloride ions (the first one using HCl, the second one a solid-state chlorination), and a pyrometallurgical process are considered. This work aims to push and lead the stakeholders towards the implementation of sustainable processes for the permanent magnet exploitation, following the European Union guidelines for the circular economy.

2. Materials and Methods

2.1. Process Description

The present section describes the innovative processes considered for the environmental assessment. The description is combined with the energy and mass balance, referred to a functional unit of 1000 kg of Nd-Fe-B permanent magnets (Table 1). The selected processes (hydrometallurgical, pyrometallurgical and solid-state chlorination treatments) ensure a selective recovery of REE with a purity of the final product higher than 95%.

2.1.1. Hydrometallurgical Process

Figure 1 shows the block diagram of the hydrometallurgical process [9]. After their preliminary demagnetisation at 310 °C for 1 h, the Nd-Fe-B permanent magnets are grinded up to a particle size smaller than 150 µm. Before the chemical leaching with 0.5 M HCl solution, the particle substrates are roasted at 810 °C for 6 h to convert iron to Fe₂O₃ that is less prone to the acid leaching, with minimum Fe dissolution. The REE (Nd, Pr and Dy) leaching is carried out under these conditions: roasted magnet powder concentration of 10% (w/v), at 95 °C for 5 h, with a resulting efficiency of 98% for REE with a low concentration of Fe ions 0.014 g/L. Therefore, the leaching solution is filtered and the Fe₂O₃ residue is recovered with a purity of 93%. On the other hand, the resulting leaching solution is treated with oxalic acid for the REE precipitation. The recovered REE oxalates are roasted at 800 °C for 2 h to convert oxalates in oxides. The final recovery efficiency and purity are higher than 98%. The resulting liquor is used for the further preparation of leaching solution.

Table 1. Energy and mass balance of the three considered treatment for the end-of-life permanent magnet exploitation.

Process	Step	Input	Output
Hydrometallurgical process	Demagnetisation	58 kWh	1000 kg permanent magnet
	Crushing and grinding	10 kWh	1000 kg magnet powder (<150 µm)
	Oxidation	369 kWh	1365 kg oxidized magnet powder
	Leaching	13,600 kg water 250 kg HCl 164 kWh	1080 kg solid residue (Fe ₂ O ₃ 93%) 13,100 kg leaching solution
	Precipitation	257 kg oxalic acid 41 kWh	662 kg REE oxalates 12,700 kg solution residue
	Roasting	94 kWh	304 kg REE oxides
Pyrometallurgical process	Demagnetisation	58 kWh	1000 kg permanent magnet
	Crushing and grinding	10 kWh	1000 kg magnet powder (<150 µm)
	Oxidation	434 kWh	1338 kg oxidized magnet powder
	Reduction	6688 kWh 0.15 kg Ar	1070 kg oxidized magnet powder + Fe
	Magnet separation	1 kWh	615 kg Fe 455 kg REE oxides
Solid-state chlorination	Demagnetisation	66 kWh	1000 kg permanent magnet
	Crushing and grinding	100 kWh	1000 kg magnet powder (<100 µm)
	Mixing	1500 kg NH ₄ Cl	2500 kg magnet powder + NH ₄ Cl
	Solid-state chlorination	1740 kWh 116 kg N ₂	480 kg NH ₃ 1990 kg magnet powder + NH ₄ Cl
	Ammonia production	1910 kg water	1910 kg ammonia solution
	Buffer leaching	21,000 kg water 630 kg C ₂ H ₄ O ₂ 30 kg NaCH ₃ COO · 2H ₂ O	316 kg magnet residue need a second solid-state chlorination 23,040 kg leaching solution
	REE solvent extraction/precipitation	2259 kg DEPFA 1581 kg n-heptane 2032 kg H ₃ PO ₄ 5646 kg water	463 kg REE phosphate 23,150 kg solution residue
	Fe recovery	4630 kg oxalic acid	1535 kg Fe oxalate 20,000 kg solution residue

2.1.2. Pyrometallurgical Process

The pyrometallurgical process includes a preliminary demagnetisation at 310 °C for 1 h (comparable with that described by Kumari et al. (2018)) and the permanent magnets grinding with a final particle size lower than 150 µm (Figure 2) [36]. The resulting Nd-Fe-B magnet powder is oxidised at 1000 °C for 2 h to convert metals to the mixture of the oxides (Nd₂O₃, Pr₂O₃, La₂O₃, Fe₂O₃, Al₂O₃ and B₂O₃). Thereafter, the metal oxides are heated at 1400–1550 °C for 7 h to reduce Fe₂O₃ to iron by carbon, under Ar atmosphere (200 mL/min). The resulting iron is removed by magnetic separation. The final REE oxide purity is higher than 95%, and the main impurities are Al₂O₃ and B₂O₃.

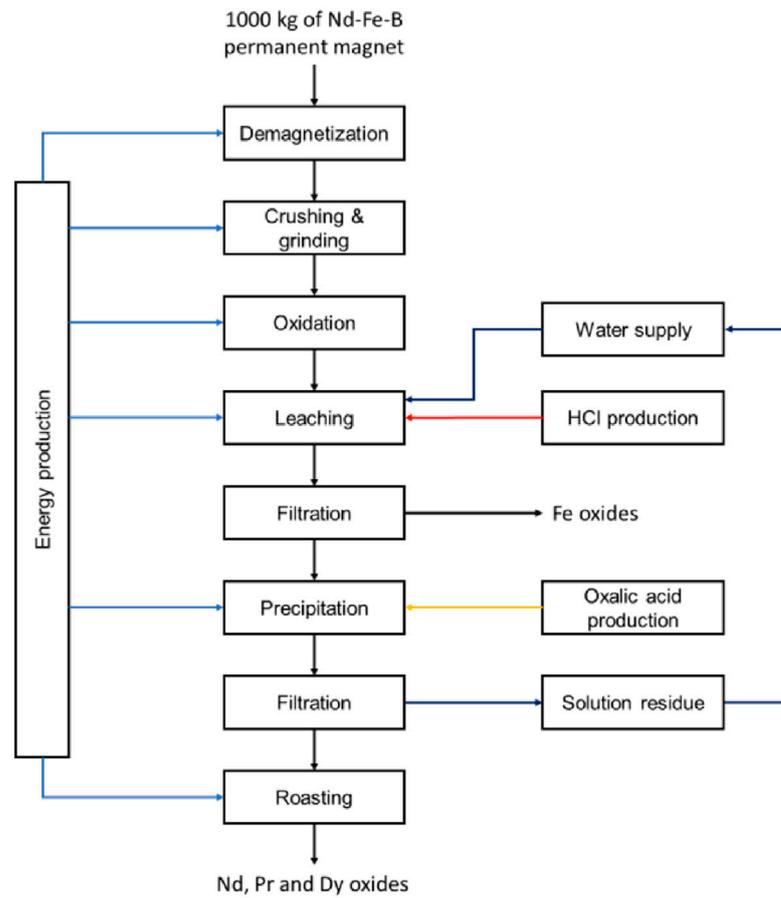


Figure 1. Block diagram of the hydrometallurgical process [9].

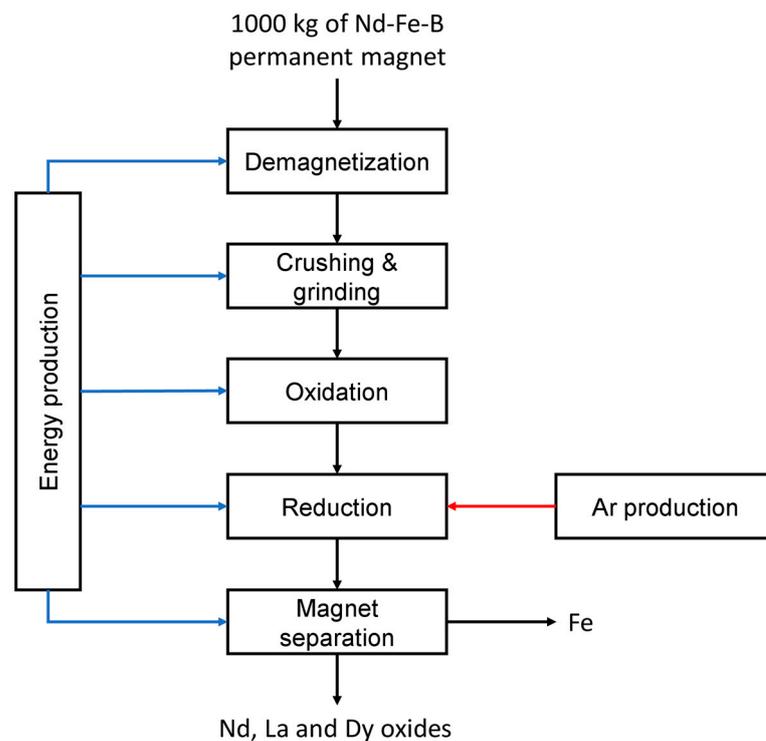


Figure 2. Block diagram of pyrometallurgical process (adapted from Bian et al. (2015) [36]).

2.1.3. Solid-State Chlorination Process

The third process is the solid-state chlorination [10]. This process has some advantages compared to the common hydrometallurgical processes: the low reagent request, the low economic cost and the avoided production of acid wastewater. As showed in Figure 3, the Nd-Fe-B permanent magnets are firstly demagnetised at 350 °C for 2 h and then ground to particle size lower than 100 µm. After these pre-treatments, the magnet powder is mixed with NH₄Cl (1:3 w/w, magnet:NH₄Cl). The solid-state chlorination is carried out at 285 °C for 2.5 h. At this temperature NH₄Cl is decomposed following the Equation (1), and the gaseous HCl reacts with metals to form metal chloride (Equations (2) and (3)):

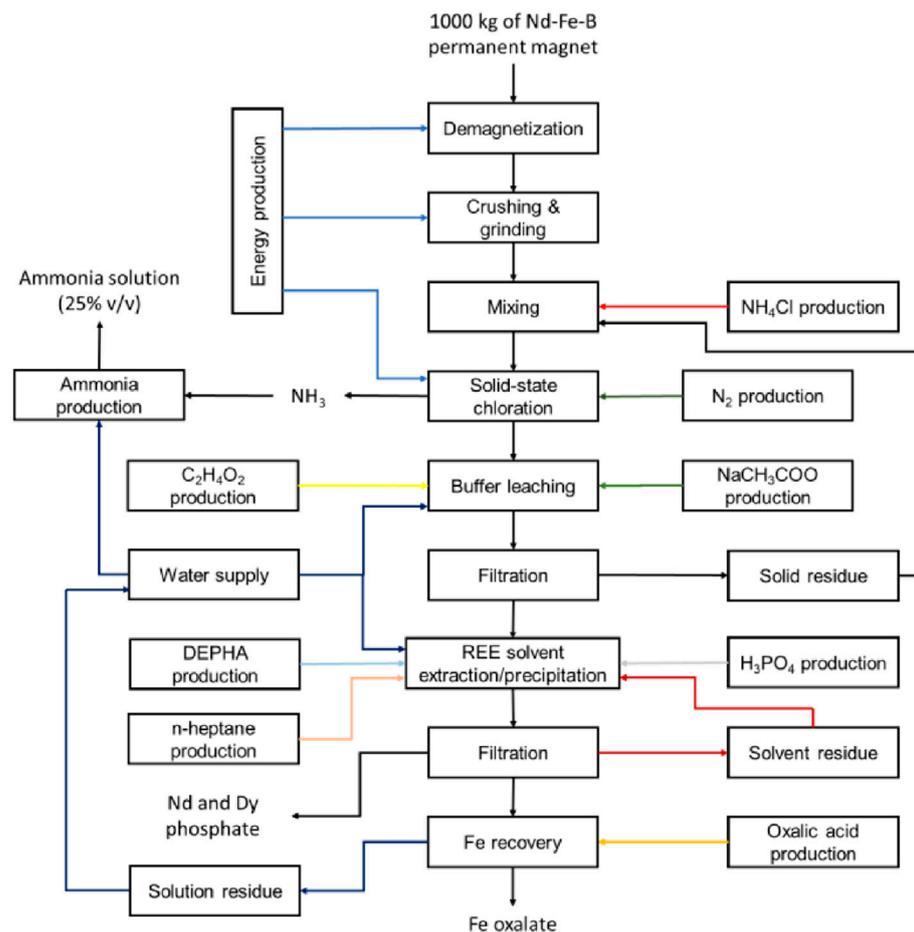
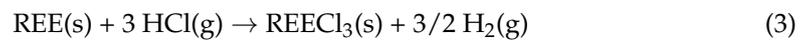
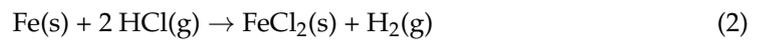
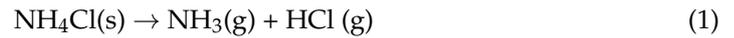


Figure 3. Block diagram of solid-state chlorination process [10,11].

The unreacted NH₄Cl is recovered and reused for further solid-state chlorination. Thereafter, the permanent magnets are treated with a buffer solution (21.02 g of buffer solution/g of permanent magnet). The buffer solution is composed of 0.5 M acetic acid and 0.01 M NaCH₃COO · 3H₂O solution at pH 3. The leaching is carried out at room temperature for 2.5 h. This process allows to achieve a leaching efficiency around 93% and 84% for Fe and REE, respectively. The remaining 16% of the REE in the solid residue can be recovered with a second solid-state chlorination. The main disadvantage is the co-leaching of iron, solved by a solvent extraction, following the method described by Amato et al. (2019), considered for the present sustainability analysis. More in detail, the recovery step

is carried out with an organic solvent bis-(2-ethylhexyl)phosphoric acid (DEHPA), diluted in n-heptane, at a concentration of 50% *v/v*. The solvent solution is treated with a H₃PO₄ solution for the selective REE precipitation with a recovery efficiency of 98%. The 95% of both the solvent and the aqueous solution can be recirculated. Thereafter, iron is recovered from buffer solution by precipitation with oxalic acid [11,37].

2.2. Life Cycle Assessment

The life cycle assessment (LCA) takes into account the energy and mass balance of the three reported processes for the Nd-Fe-B permanent magnet exploitation (from gate to gate) (Table 1), and it is carried out in agreement with the LCA ISO standard 14040 and 14044:2006 [38,39]. The selected functional unit for the assessment is 1000 kg of end-of-life permanent magnet. The software used for the data collection is thinkstep Gabi 9.5. The method selected for the analysis is EF 3.0, including all the environmental categories, recommended models at midpoint, together with their indicators, units and sources [40,41]. The process considered for the present assessment are developed at laboratory scale; some sensible assumptions are made to hypothesise the further scale-up [42–44]. The considered assumptions are reported below:

- Industrial machineries are considered to calculate the energy demand for the mechanical/physical treatments (e.g., demagnetisation, grinding, heating) [42];
- The filtration and mixing energy demand are considered negligible due to the lower value than the other processes (<0.002 kWh/kg end-of-life permanent magnet) [45,46];
- The by-products (Fe₂O₃ or Fe oxalate) are considered as environmental credits since they could be used in different processes, e.g., for environmental remediation strategies [47] or as anode in the lithium-ions batteries [48], respectively;
- The 95% recirculation of the organic solvent for REE extraction is considered [11];
- The NH₃ gas, generated during the solid-state chlorination, is recovered and used to produce an ammonia solution with a final concentration of 25% (*v/v*) and a purity higher than 99% [10];
- The solution resulting from the recovery step is used to prepare further leaching solutions [9,11];
- Considering the absence of Nd, Pr and Dy oxides in the reference database, their environmental burdens have been estimated by the allocation of La₂O₃ impact, on the market price basis [49];
- The oxalic acid production process, considered for the analysis is that described by Santoro et al. (1999), using milk whey as carbon source [50].

3. Results

Figure 4 reports the results of classification and characterisation phases of LCA. Overall, it is evident that the high recovery efficiency of the three options represents a very important benefit from the environmental point of view, with an advantage of the solid-state chlorination. Indeed, the environmental credit of REE balances and overcomes the process burdens, in most considered categories. The main advantage is connected to Nd oxide, mainly for the highest recovered quantities, compared to the other REE. A contribution between 14% and 19% is assigned to Pr oxide in both hydrometallurgical and pyrometallurgical options. On the other hand, in solid-state chlorination process, the environmental credit is equally distributed between Nd and Dy oxides. The choice of environmental impact allocation on the economic price explains relevant effect of Dy oxide, even if its recovered quantity is around six times lower than Nd oxide. Indeed, the price of these REE, at January 2020, reports a value of 251.11 USD/kg for Dy oxide vs. 41.76 USD/kg for Nd oxide [49].

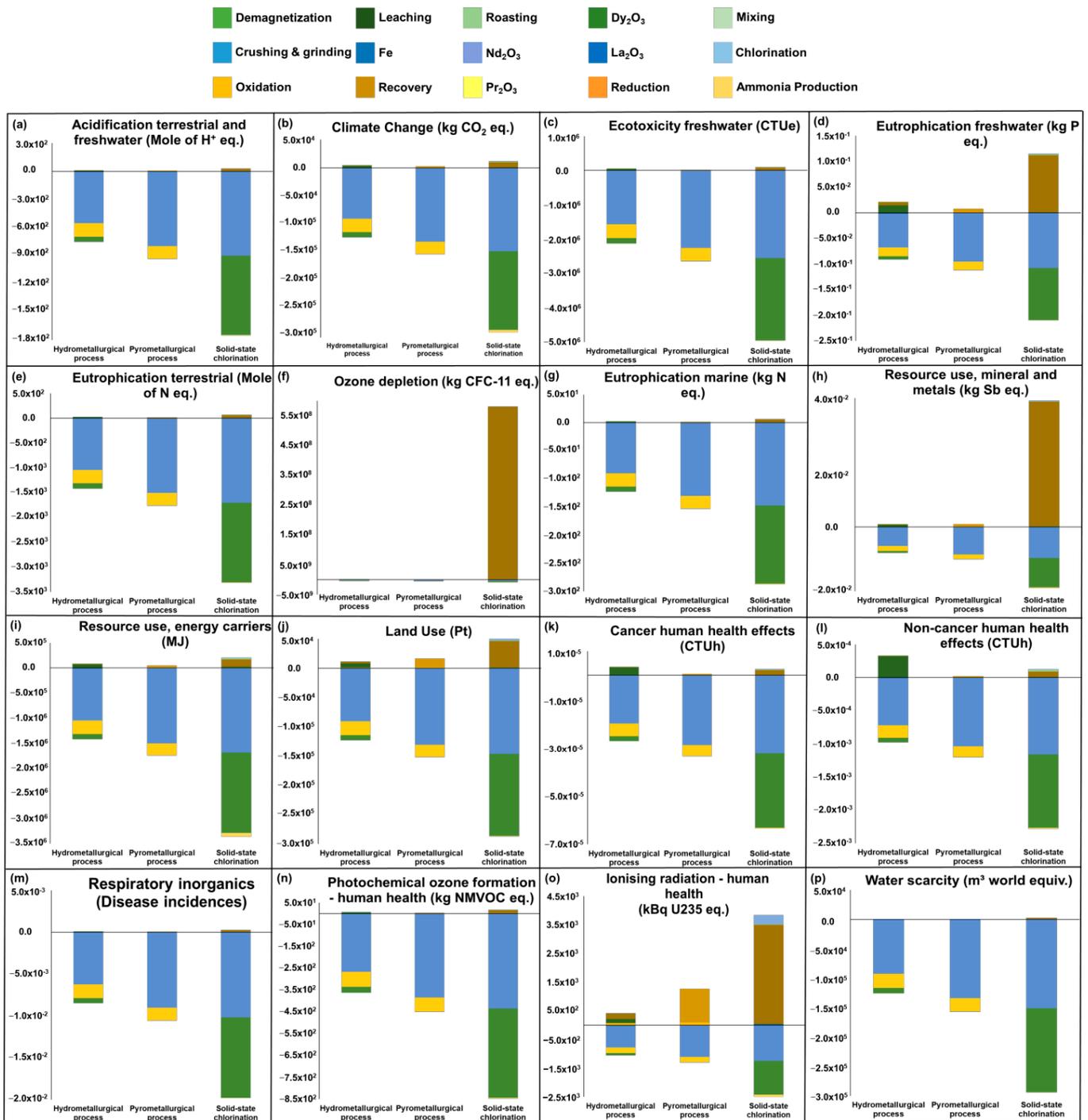


Figure 4. Results of classification and characterisation steps of LCA, the figures from a–p represent the considered impact categories (functional unit: 1000 kg of Nd-Fe-B permanent magnets).

A separate discussion is reserved to the categories of resource use, mineral and metals and ozone depletion (Figure 4f,h), which identify the solid-state chlorination as the worst option. The burden of recovery step on the ozone depletion is due to the use of n-heptane, in the third option. On the other hand, the phosphoric acid affects the category of resource use, mineral and metals, due to the mining of phosphate rock used for its production [51].

The classification and characterisation steps supply essential information to define the most sustainable process for the permanent magnet exploitation. Nevertheless, Figure 5 shows the most complete overview thanks to the estimation of the environmental perfor-

mance index (EPI) able to include all the impact categories. This value is expressed as person equivalent (p.e.), which represents the number of average citizens that generates the same impact in one year [52]. As reported in Figure 5, the hydrometallurgical process causes the lowest environmental burden. Nevertheless, the greatest credit achieved by solid state chlorination makes this option the most environmentally sustainable choice. Overall, the negative value of EPI, confirms the benefit of the permanent magnet recycling, compared to the primary production of REE.

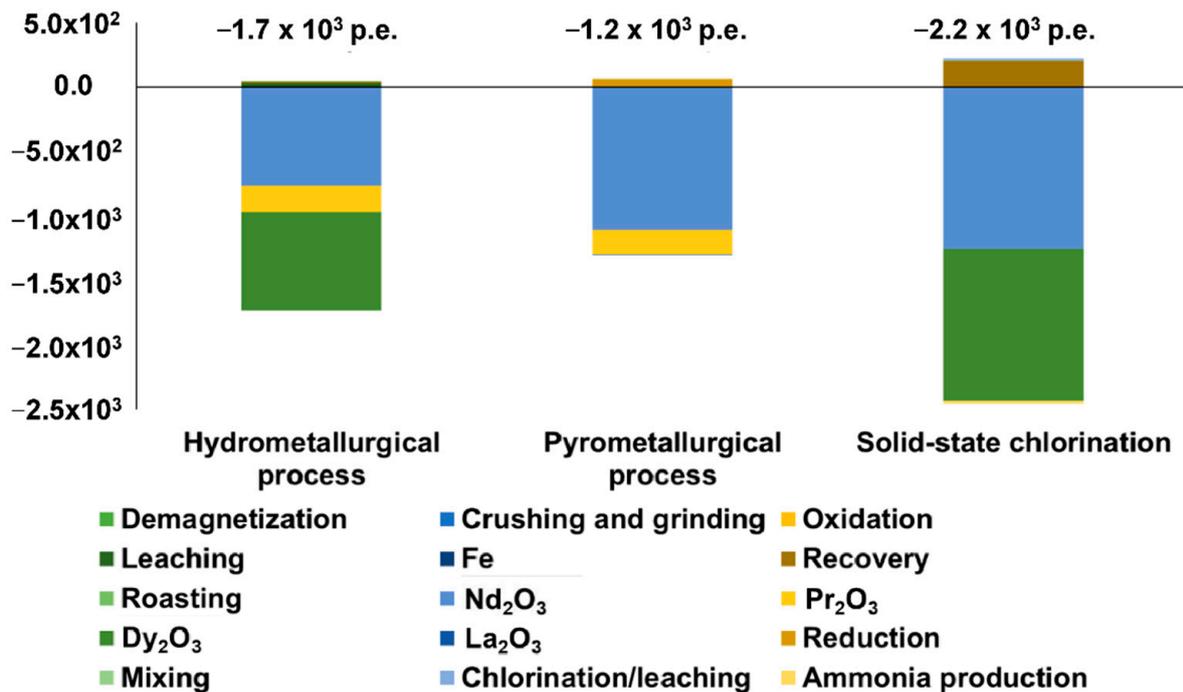


Figure 5. Normalisation and weighing results of the three treatments: effect of the different step expressed as p.e. (functional unit: 1000 kg of Nd-Fe-B permanent magnets).

To better compare the impacts of the three analysed processes, Figure 6 shows the effects on the different impact categories, dividing the most affected by the process (Figure 6a) and the most favored by REE recovered (Figure 6b). It is evident that the ionising radiation-human health represents the main issue. This effect is explained by the electricity demand necessary for the synthesis of oxalic acid used in the recovery step in both hydrometallurgical and solid-state chlorination processes. Furthermore, the high energy request of pyrometallurgical technique (mainly due to the reduction phase) explains the contribution greater than 70% of this impact category on the whole impact. The effect of energy supply on the ionizing radiation-human health category is due to the European electricity grid mix considered for the analysis (supplied by Gabi database), particularly to the radionuclides (potentially toxic for humans) resulting from both the nuclear energy production, and the mineral oil and gas extraction, used as energy carriers [46,53]. As reported in Figure 6b, the benefit of REE recovery is mainly highlighted in the categories of climate change and resource use, and energy carriers. The highest credit resulting from the solid-state chlorination balances its greatest burden making this option the most sustainable and the pyrometallurgical process the worst choice.

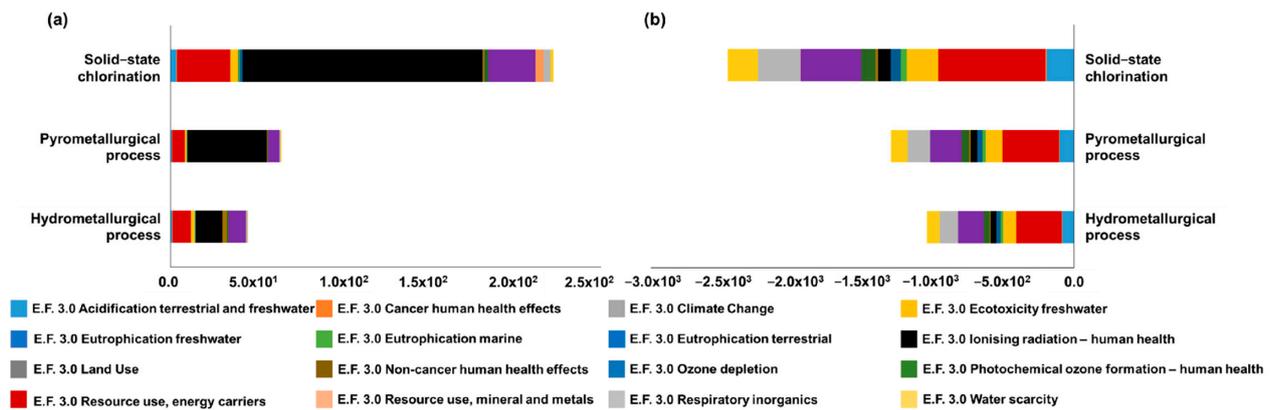


Figure 6. Normalisation and weighing results: (a) the main affected impact categories by processes and (b) the main saved impact categories by REE recovery, expressed as p.e. (functional unit: 1000 kg of Nd-Fe-B permanent magnets).

4. Sensitivity Analysis

The choice of the allocation method based on the economic price of REE oxides could cause an uncertainty on the whole result, due to the fluctuation of the REE prices. For this reason, the assessment of EPI has been repeated estimating Nd, Pr and Dy credits by the allocation of La₂O₃ impact, on the market price in 2009, reported in Table 2.

Table 2. Market price of REE oxides, comparison between 2009 [54] and 2020 [49].

Reference Year	La Oxide	Pr Oxide	Nd Oxide	Dy Oxide
	USD/kg			
2009	6.25	14.0	14.0	100
2020	1.69	48.4	41.8	251

As reported in Figure 7, the exploitation of permanent magnets as secondary resource of REE oxides results an environmentally sustainable choice, irrespective of the specific metal prices. Indeed, the assessed EPI show negative value despite the lower prices in 2009 than 2020. Nevertheless, the effect of market fluctuation is evident and significantly affects the balance between burden and benefit. The sensitivity analysis shows that the most relevant effect is the variation of Dy price, which cancels the advantage of the solid-state chlorination, in favor of the hydrometallurgical option. Therefore, the simple design and the lowest burden resulting from this process make this option the most environmentally sustainable, despite the lowest quantity of recovered REE, irrespective of the market price.

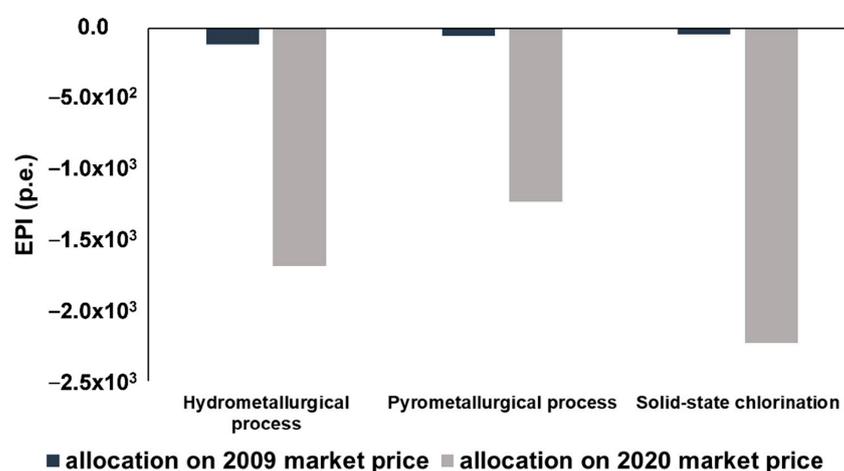


Figure 7. Comparison between the EPI resulting from the allocation on 2009 and 2020 REE oxide prices.

5. Conclusions

The identification of the most sustainable choice for the exploitation of permanent magnets represents a hot topic and it is set to become ever more relevant considering the increasing applications of this technology, and the growing REE demand. The environmental sustainability analysis proved the benefit of recycling treatments, compared to the primary production of metals, irrespective of the chosen approach. Nevertheless, the LCA showed that the economic aspects affect the final choice. More in detail, high economic value of the REE pushes towards the highest recovery processes (but greater environmental burden), that is, the solid-state chlorination. On the other hand, the simple design of the hydrometallurgical treatment ensures high environmental sustainability levels, irrespective of the market fluctuation. The present paper represents an excellent example of implementation of LCA as support tool for the definition of circular economy strategies.

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