



# SIMULATION AND ECONOMICAL ANALYSIS OF HYDRO-ND PROCESS FOR THE RECOVERY OF RARE EARTH FROM END-OF-LIFE PERMANENT MAGNETS: NEW-RE AND INSPIREE PROJECTS

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#### **Article Info:**

Received: 18 April 2024 Revised: 3 June 2024 Accepted: 12 July 2024 Available online: 30 September 2024

**Keywords:** Rare earth elements Recycling Economic Analysis Permanent Magnets **Hydrometallurgy** Circular Economy

#### **ABSTRACT**

The growing demand for rare earth elements (REEs) and sustainable development issues have made the REEs recovery from permanent magnets (PMs) attract the attention of many researchers in the last decade. The NEW-RE and INSPIREE projects have been introduced to evaluate the recovery of REE elements from permanent magnets on a pilot and industrial scale. In this research, the economic aspect of the mentioned projects was performed using SuperPro software. The main aim of the work is to highlight the critical aspects of the process for a targeted optimization. The results showed that the CAPEX and OPEX for treating 3,600 tons/year of permanent magnets are 9 and 110 million euros, respectively, and an EBITDA of 4.8 million euros can be achieved (payback period less than two years). In the economic model, the cost of spent PMs was considered equal to 50% of its REEs value (14,251 €/ton). Also, it was found that the main OPEX costs are raw materials (65%) and energy (16%), respectively. It needs to be pointed out that the maximum price of permanent magnets can be 15,230 €/ton (BEP).

#### **1. INTRODUCTION**

The increase in sensitivity towards issues such as safeguarding resources and the environment is increasingly pushing towards adopting circular economy models. Thanks to this phenomenon and the strong profitability of recovering critical raw materials from end-of-life devices, numerous companies have been created to recycle industrial waste and electrical and electronic equipment.

The application of permanent magnets containing rare earth elements (REEs) has increased dramatically in various electrical and electronic devices due to their special and unique properties. The supply chain (China, Australia, and Russia are the primary world producers (Kapustka et al., 2020)) and their increasing consumption (the consumption is expected to be more than 350,000 tons per year in 2035 (Yang et al., 2017) have led to the classification of several REEs such as Nd, Pr, Dy, and Y as the most critical raw materials by the European Union (Eyl-Mazzega et al., 2020). In addition, depositing end-of-life electronic equipment causes loss of mineral resources and conse-

merous devices containing REPMs are starting to return as waste from their respective markets: wind turbine generators, HDDs, electric motors, and acoustic devices. As a result, there is a need to have more and more operational plants to manage these waste flows correctly, providing adequate solutions to avoid the loss of critical raw materials. Although hydrogen decrepitation (HD) is the most widely used method of REEs recovering from PMs, this method has some drawbacks, such as huge waste production, and is not applicable for treating all PMs types. In light of these

considerations, as well as the advantages of hydrometallurgical compared to pyrometallurgical processes (less energy required, removal of gas pollution, and environmentally friendly) (Romano et al., 2023 (a)), hydrometallurgical methods can be considered one of the most promising technologies in this field.

quently increases mining activities. Thus, recovery of rare earth elements (REEs) from end-of-life permanent magnets (PMs) is essential from sustainable development, economic, and environmental points of view. To date, nu-



Detritus / Volume 28 - 2024 / pages 141-149 https://doi.org/10.31025/2611-4135/2024.19414 © 2024 Cisa Publisher. Open access article under CC BY-NC-ND license

Various research works on a laboratory scale were carried out to investigate the effect of chemical and operational parameters on dissolving and separation efficiencies. It was found that the demagnetization of PMs is an essential step to increase the selectivity and efficiency of the leaching stage as well as to facilitate the subsequent steps of the recycling process (Kumari et al., 2018; Klemettinenet al., 2021). Previous studies showed that organic (Gergoric et al., 2018; Romano et al., 2023 (b)) and inorganic (Lee et al., 2013; Uysal et al., 2023) acids can effectively dissolve valuable metals of PMs. Also, it was reported that DEHPA is more efficient in separating both light and heavy REEs than Cyanex 272 and Cyanex 572 (Yang et al., 2017; Ni'amet al., 2020).

In 2019, a novel method for the recovery of REEs from end-of-life permanent magnets, based on hydrometallurgical methods, was introduced by Birloaga and Veglio. This method leaches demagnetized and ground spent PMs by the citric acid solution to dissolve REEs, iron, and boron. The REEs are extracted at the solvent extraction, while iron and boron remain in the raffinate solution and are recovered as the byproduct of this process. Also, the loaded organic phase is stripped by phosphoric acid to produce REEs- phosphate and convert to REE-oxalate in the next step. In the end, the REE oxide is achieved by introducing the REE-oxalate to a furnace. According to the appropriate laboratory results of this method, the NEW-RE and IN-SPIREE projects were defined to evaluate the REEs recovery on pilot and industrial scales, respectively.

The main objective of this work is to highlight the critical aspects of the Hydro-Nd process to optimize it in a targeted way. This result is achieved through a detailed simulation that allows material and energy balance realization.

The simulation allows us to analyze the system's material flows and energy consumption, identifying inefficiencies and improvement points. This activity is essential for the scale-up of the process, moving from the laboratory to the industrial scale through the pilot phase. This way, it is possible to intervene precisely in the critical process phases, ensuring a more effective and sustainable optimization and facilitating the transition to large-scale production.

# **2. MATERIALS AND METHODS**

This work aims to create a model to describe the hydrometallurgical process using the SupePro Designer v9.5 process simulator. This simulator is widely used in the hydrometallurgical sector due to the numerous features it offers. The scenario analyzed concerns the continuous treatment of approximately 3600 tons/year of end-of-life PM.

#### **2.1 PM sample**

In order to proceed with the process simulation, it is necessary to identify all the input streams. Among the different streams, the one most influenced by the market is that of permanent magnets. Results obtained from laboratory tests on samples of NdFeB permanent magnets are used as a reference composition for this current. Figure 1 shows the composition of the supply stream.

#### **2.2 Process Description**

The hydrometallurgical process studied is composed of different phases. The permanent magnets are sent to an oven for the thermal demagnetization phase at approximately 400°C. The oven is powered by the hydrogen





produced in the extraction phase, described below. Once demagnetized, the magnets are removed from the rotors and subjected to a grinding process. The granulate is then introduced into the reactor, where the REEs, iron, and boron contained in the demagnetized powders are leached with a citric acid solution. The following reactions are involved in the processs:



 $2Pr + 2C_6H_8O_7 \rightarrow 2C_6H_5Pro_7 + 3H_2$  $(2)$ 

 $2Tb + 2C_6H_8O_7 \rightarrow 2C_6H_5TbO_7 + 3H_2$  (3)  $(4)$ 

 $2Nd + 2C_6H_8O_7 \rightarrow 2C_6H_5NdO_7 + 3H_2$  $(5)$ 

$$
2B + 2C_6H_8O_7 \rightarrow 2C_6H_5BO_7 + 3H_2
$$
\n
$$
2Fe + 2C_6H_8O_7 \rightarrow 2C_6H_6FeO_7 + 2H_2
$$
\n(6)

$$
2Y + 2C_6H_8O_7 \to 2C_6H_5YO_7 + 3H_2
$$
 (7)

In the overall chemical reaction, in addition to the solubilization of REEs (Nd et al.), boron, and iron, hydrogen is developed, extracted from the reactor, and sent to the demagnetization oven. Figure 2 shows the simplified block diagram of the process.

Once the reaction is over, the resulting solution is filtered, obtaining a solid residue rich in nickel and a liquid solution rich in rare earth to be recovered.

The solution is sent to a mixer, where an extraction with solvent (D2EHPA and n-Heptane) takes place. Subsequently, thanks to a settler, the two liquid phases are separated. Aqueous solution rich in iron, boron, and citric acid is first treated with oxalic acid. The reaction between the acid and

the iron ions precipitates iron oxalate, which is separated from the liquid waste and filtered again. The filtered water is recirculated in the system while the waste treated with calcium hydroxide and calcium citrate goes to a boron recovery section. Boron hydroxide is recovered, and the remaining water is sent for reuse.

The other solution that comes out of the settler, rich in rare earth ions, is sent to a mixer for the second extraction with solvent: phosphoric acid is inserted into the processing unit, which causes the rare earth ions to return to the aqueous solution purifying the solvent that is recirculated. The aqueous solution is treated with oxalic acid, obtaining rare earth oxalates, which, once precipitated, are recovered.

# **2.3 Simulation setup**

A model was created on the SuperPro Designer v9.5 software to characterize each flow rate and equipment of the process, starting from the above data. Figure 3 shows the complete simulation flowsheet.

The diagram highlights the process inputs (for example, PMs Powder) in light blue and all the outputs (for example, Hydrogen) in orange. The scheme also includes the process for the recovery of rare earth oxalates and the recovery of iron. The simulation was set directly with the possibility of recycling part of the solutions used (purple dashed lines). All recycles were analyzed in order to minimize reagent consumption.







**FIGURE 3:** Flowsheet of SuperPro Designer simulation.

#### *2.3.1 Equipment setup*

The different equipment was configured based on the results obtained from laboratory-scale experimental tests. Table 1 briefly summarizes the procedures and various operations.

#### *2.3.2 Input streams setup*

The variables of the different flow rates entering the plant were calculated based on the optimizations carried out on a laboratory scale for each of the envisaged unit operations. As regards the leaching section, a citric acid concentration of 1.45 mol/L was considered, with a pulp density of approximately 10% wt./vol. The solvent extraction section used a volume ratio between the two phases (organic and aqueous) equal to 1. The exact ratio was used to prepare the organic phase: the exact volume quantities of D2EHPA and n-Heptane. Regarding the stripping phase, the volumetric ratio of the organic phase/aqueous phase is 2:3. The quantity of phosphoric acid entering this section was chosen to obtain the optimal value of 1.8 mol/L during the operation. In the conversion phase of rare earth phosphates into rare earth oxalates, a solid-liquid ratio of 7% was considered, with an incoming oxalic acid concentration of 1.6 mol/L. Finally, for the precipitation phase of the iron oxalate to recover this element, a quantity of oxalic acid over 80% compared to the stoichiometric quantity obtained from the following reaction was used:

$$
C_6H_6FeO_7 + C_2H_2O_4 = FeC_2O_4 + C_6H_8O_7
$$
 (7)

Equipment	<b>Operation</b>	<b>Procedure</b>	Setting
R-101	Leaching	<b>REACTION</b>	Reaction Progress: 95-99%
FP-101	Filtration	<b>FILTER</b>	Solid fraction removed: 100%
			LOD Cake Dryness: 30%
		CAKE-WASH	Wash stream amount: 30% vol. leaching solution
MS-101	Solvent Extraction	<b>REACTION</b>	REEs Reaction Progress: 95-99%
			Fe Reaction Progress: 30%
MS-102	Stripping	<b>REACTION</b>	Reaction Progress: 100%
R-102	Precipitation	<b>REACTION</b>	Fe Reaction Progress: 100%
R-103	Precipitation	<b>REACTION</b>	REEs Reaction Progress: 100%
FP-102	Filtration	<b>FILTER</b>	Solid fraction removed: 100%
			LOD Cake Dryness: 30%
		CAKE-WASH	Wash stream amount: 30% vol. leaching solution
<b>RVF-101</b>	Filtration	<b>FILTER</b>	Solid fraction removed: 100%
			Cake Porosity - Cake Dryness: 0.4 vol/vol
		CAKE-WASH	Available in Wash-In Stream

**TABLE 1:** Summary of Equipment Setup.

To estimate the volumes of the different equipment it was necessary to enter the different residence times. The residence times used were:

- Leaching Time: 180 min;
- Solvent Extraction: 20 min;
- Stripping: 90 min;
- Oxalates production: 60 min;
- Iron Production: 60 min.

All operations are conducted at room temperature. This aspect allows for a process with low energy consumption. The energy consumption of the process includes the mixing operations in the different reactors and the pumps for moving the different streams (obviously, if the thermal sections of the demagnetization oven and the oven for converting the oxalates into oxides are excluded). Figure 4 shows the summary of INPUT streams.

#### *2.3.3 Economic Analysis*

Table 2 briefly summarizes the calculations and assumptions made for the economic analysis.

## **3. RESULTS**

## **3.1 Material balance**

# *3.1.1 Drain Optimization*

Before analyzing the results of the material and energy balances in detail, an optimization of Drains 1 and 2 was carried out. In this regard, the influence of the split ratio between the two currents coming out of each Drain was evaluated to obtain the lowest possible reagent consumption in the leaching reactor. Figure 5 shows the results of this optimization.

Thanks to purges, significantly reducing the demand for raw materials is possible. In particular, the following is

achieved: a reduction of approximately 50% in the requirement for citric acid and the total removal of the need for dilution; a 90% reduction in the demand for D2EHPA and n-Heptane and a 90% reduction in the demand for sulfuric acid and the water needed to dilute it. Due to the demand for oxalic acid, it is impossible to optimize everything through recirculation; thanks to water in the solution in which the iron citrate is present, dilution is optional.

#### *3.1.2 Output*

The material and energy balances of the proposed method were investigated for comprehensive characterization of all process streams. Figure 6 shows the results of the outputs of the process streams. It needs to be pointed out that the required reactions of this method take place under ambient temperature and pressure conditions, which is important from economic, technical, and safety standpoints.

The final product, REE-oxalate, consists of more than 86% Nd and Pr oxalate. The high concentration of REEs in the REE-oxalate confirms the subsequent industrial applications of this production. On the other hand, due to the high concentration of Ni in leaching residues (78.5%), recovery of nickel from these wastes can be important from economic and environmental aspects.

Although  $\sim$  25% of iron was extracted by DEHPA in the process, the low iron concentration in the REE-oxalate powder (~0.1%) presents a notable advantage. Also, the iron oxalate achieved using this method showed an impressive purity level of 98%, which facilitates its conversion into iron oxide for subsequent industries as a raw material for Nd-FeB permanent magnet production.

Another point of consideration is the low concentration of boron in loaded DEHPA and was remained in the waste solution of the iron recovery stage. To address this,



**FIGURE 4: Summary of INPUT streams.** 

#### **TABLE 2:** Assumptions and calculations of economic analysis.



the application of other methods to recover B needs to be evaluated. it should be noted that recovering B not only increases overall resource efficiency but also minimizes waste generation.

# **3.2 Economic Analysis**

Through the economic analysis, it was possible to identify some of the indices to evaluate the feasibility of the investment necessary for the plant's construction. For the case study of 3600 ton/y of permanent magnets (500 kg/h) with a cost equal to 50% of the intrinsic economic value (IEV) associated with the rare earth content, the following values were calculated: a CAPEX of approximately 9 M€, an OPEX of approximately 110 M€ and revenues worth approximately 115 M€. Based on these values, an EBITDA of approximately 4.8 M€ and a PBP of less than two years



**FIGURE 5:** Optimization of the Drain-to-Feed ratio for the two splits SP-101 and SP-102.





were calculated. Table 3 shows the results of the Business Plan created per ton of PMs. It should be noted that the required amount of OPEX is significantly higher than CAPEX for this process.

# **4. DISCUSSIONS**

 Figure 7 shows a breakdown of the OPEX involved in the process of recovering REEs from spent rare earth permanent magnets. From the economic analysis, it was observed that the costs related to the procurement of raw materials, which constitute 65% of the total OPEX, and the energy required for this process, which constitutes 13% of the OPEX, constitute the main parts of the OPEX portion. It is worth noting that the major share of the cost of raw materials is considered for the use of permanent magnets, the price of which is determined based on the market demand.

**TABLE 3:** Estimated CAPEX, OPEX, and hydrometallurgical process cost for studied scenarios. Values referred per ton of treated PMs.



Furthermore, the findings show that waste storage costs account for 10% of total OPEX. It should be noted that this cost can be reduced by recovering nickel. Thus, the waste volumes and associated storage costs can be reduced.

Based on the current flowsheet, the key to improving the economic efficiency of this process is identifying solutions to procure spent permanent magnets at a lower cost. Addressing this aspect holds promise as the primary avenue for improving the overall cost-effectiveness of the process.

Based on the previous observations, thanks to the model created, a sensitivity analysis was performed to understand the trend of the PBP as the price of the lead magnets varied. The results of this sensitivity analysis are shown in Figure 8.

The PBP varies very little for permanent magnet costs below 14,500 €/ton. However, if this value is exceeded, the PBP grows significantly asymptotically at the Break Event Point (BEP) by approximately 15,230 €/ton. Magnet costs higher than this value led to an unprofitable activity. Consequently, the increase in the price of spent permanent magnets beyond 15,230 euros/ton, without a corresponding adjustment in the selling price of the final product (REE-oxalate) leads to the investigation of alternative methods for the recovery of REEs through hydrometallurgical approaches. Notably, the industrial plant of the INSPIREE project is designed with a flexible circuit that allows for modification of the flow of materials, solutions, and chemicals. Also, to reduce the economic risk of these projects, the recycling of lithium battery cathodes (LFP) and photovoltaic panels was also considered in the design phase. This strategic decision not only expands the scope of the project but also increases its flexibility by diversifying potential revenue streams and reducing dependence on a single source of raw materials.

#### **5. CONCLUSIONS**

The recovery of material and energy from industrial waste and end-of-life products is increasingly a fundamental element for the sustainable development of countries. In particular, recovering rare earths from permanent magnets at the end of their life is of great strategic interest.



**FIGURE 7:** Influence on OPEX of some factors.



**FIGURE 8:** Trend of the PBP with the price of permanent magnets.

In this work, the Hydro-Nd hydrometallurgical process was analyzed from an economic point of view.

By creating a simulation using SuperPro Designer v9.5 software, it was possible to characterize all the process currents and create a model that allows sensitivity analyses to be carried out for different scenarios.

The scenario with an inlet flow rate of 3600 tons/h (500 kg/h) was studied in detail. Thanks to the model created, the process was optimized in terms of reagent consumption thanks to the inclusion of various recirculations. An economic analysis was set up starting from the optimized process.

For the scenario considered, with a magnet price of around 14,251 €/ton, there is a CAPEX of around 9 M€ and an OPEX of around 110 M€. This high OPEX value is due to the high cost of the magnets. However, an equally high revenue of around 115 M€ allows for an EBITDA of around 4.8 M€ and a PBP of less than two years. By evaluating the influence of the price of magnets on the PBP, the BEP was determined to be approximately 15,230 €/ton.

The economic analysis has allowed us to identify the critical points of the Hydro-Nd process for subsequent targeted optimization. It has been seen that the most critical aspect is linked to the cost of raw materials. In this regard, work is being done on a laboratory and pilot scale to optimize this aspect, facilitating the scale-up of the process towards large-scale production.

# **FUNDING**

This study is financially supported by the LIFE22-ENV-IT-INSPIREE project (number: 101113882).

# **ACKNOWLEDGEMENTS**

The authors thank all the NEW-RE and INSPIREE Consortium partners for collaborating in this research activity. The authors would like to thank the University of L'Aquila for their precious support.

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