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Towards a Sustainable Future: Circular Strategies for Critical Raw Materials Recovery from End-of-Life Magnets

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Abstract. The transition to a circular economy is crucial for addressing the environmental and resource challenges posed by traditional linear production models such as resource depletion, environmental degradation, and waste accumulation. In this context, growing interest is devoted to critical raw materials (CRMs), including rare earth elements (REEs), cobalt, and others, which play a vital role in clean energy technologies, advanced electronics and high-performance magnet applications. This study presents an eco-friendly method for the recovery of REEs from end-of-life (EoL) NdFeB permanent magnets. The proposed method involves a hydrometallurgical metal recovery using an aqueous solution of bio-based leaching agents (citric acid), enabling the one-pot material dissolution and selective precipitation of REE-based compounds. This approach demonstrates high recovery efficiency, with ≥ 90 wt.% of neodymium retained in the solid phase, and yields valuable metal-rich fractions that can be potentially repurposed into new advanced applications. By opening the possibility to valorize both REE-rich solids and base metal-containing leachates, the process offers a viable path toward reduced CRMs dependency and aligns with circular economy principles through waste minimization and resource reuse.

Keywords: rare earth elements, circular economy, green chemistry, waste valorization, permanent magnets

1. Introduction

Critical Raw Materials (CRMs) are crucial for future clean and digital technologies. The European Commission has identified 30 key materials, including lithium, cobalt, and rare earth elements, as essential to the EU economy but facing supply risks (Akcil, A. et al., 2021). In recent years, CRMs demand has surged, particularly for magnets, batteries, and other advanced technologies, with rare earth elements experiencing an 11% growth between 2012 and 2020. This rising demand has led to soaring prices, while supply remains a challenge due to environmental and resource limitations. Researchers and industry stakeholders have been pursuing to reduce CRMs consumption and develop sustainable alternatives (Akcil, A. et al., 2021; Serpe et al., 2025).

In this context, the recycling of end-of-life (EoL) technology devices emerges as a promising strategy, yet traditional recovery techniques remain intricate and economically unfeasible, frequently depending on high temperatures and harsh chemicals, which lead to significant environmental consequences. Currently, only a minimal fraction of critical raw materials is recovered, highlighting the need for enhanced recycling technologies (Serpe et al., 2025).

To advance this effort, the GREEn SM project focuses on innovative and environmentally sustainable techniques for the leaching and selective recovery of critical raw materials from EoL high-performance NdFeB permanent magnets (also referred to as "Super Magnets", SM), repurposing them into high-value materials for advanced applications.

Specifically an integrated approach for the separation and recovery of REEs and base metals was established and is here described. After an appropriate thermo-mechanical pre-treatment on spent SM from hard-disk drives, the process involves the use of bio-derivable reagents – namely citric acid and ammonia, respectively, for the first and second chemical recovery stages of the process – and obtains distinct fractions enriched in valuable metals prone to be valorized for a new application.

2. Experimental Section

Test specimens consisted of NdFeB-based permanent magnets were manually dismantled from hard-disk drives (HDDs) of various manufacturers and year of production. The magnets were thermally treated at 200 °C for 1 hour to induce partial demagnetization, followed by mechanical removal of the Ni coating and grinding via hammer milling to reach a particle size of 0.5 - 1 mm. Metal leaching was performed on 1 g of powdered magnet using a 0.3 M citric acid aqueous solution under refluxing conditions and magnetic bar stirring for 24 hours. During the first hour, around 4 mL of H_2O_2 (30%) were added dropwise. The solution quickly turned from colorless to brown and a copious precipitation of a whitish precipitate occurred, identified as $Ln_2(C_2O_4)_3$ (lanthanide oxalates, $C_2O_4^{2-}$ = oxalate ion), as the reaction went off. After centrifugation, the solid REE-rich precipitate was separated from the leachate, which contained mainly base metals. All samples – magnets, leachates, and precipitates – were analyzed for metal content via ICP-OES following acidic digestion with 65% HNO_3 and appropriate dilution.

3. Results and Discussion

Test specimens were HDDs magnets constituted of $Nd_2Fe_{14}B$ alloy (approx. 70 wt.% Fe, 27 wt.% Nd, 1 wt.% B, and 2 wt.% of Co, Dy, Pr, Cu, and others), coated with a thin Ni layer to prevent surface oxidation (Figure 1).

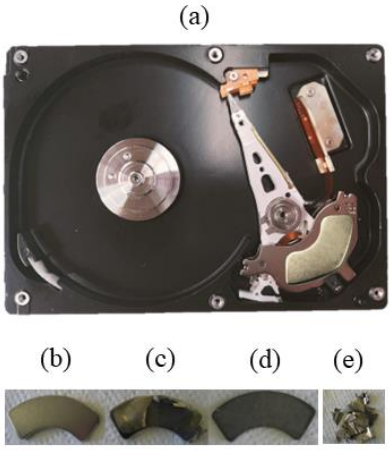


Figure 1. HDD (a); SM as removed by the HDD (b) and after thermal treatment at 200 ° C for 1h with (c) and without (d) Ni coating; removed Ni coating (e).

The Ni coating was removed after a partial thermal demagnetization (see Sect. 2) in advance to enhance metal recovery selectivity and reduce the need for chemicals. To replace strong, oxidizing acids with safer, bio-based alternatives, citric acid was used as a mild, low-cost complexing agent in aqueous solution to dissolve metals contained in the solid mixture. Preliminary mechanical grinding increased the exposed surface area of the alloyed metals with the view to improve the effectiveness of the weak acid. The leaching, performed at refluxing conditions and supported by the addition of a small amount of H_2O_2, provided a liquid phase enriched in base metals (mainly Fe and then Co) and a precipitate of insoluble lanthanide compounds (e.g., Nd, Dy, Pr oxalates, $Ln_2(C_2O_4)_3$). The role of hydrogen peroxide extended beyond oxidation, as it degraded citric acid via the Fenton reaction (in the presence of Fe^{2+}/Fe^{3+}), forming oxalate that enabled highly selective lanthanide precipitation (Sillanpää, M., Shestakova, M., 2017). This process, which proceeds via a radical mechanism, led to oxalate yields of 81-92%, with ICP-OES showing that ≥ 90 wt.% of the initial neodymium was recovered in the solid phase. The Ln-based oxalates recovered from SM treatment was used as a precursor to produce functional luminescent molecular materials for photonic and bioimaging applications. The resulting aqueous leachate, rich in Fe and citric acid and low in residual Nd, was analyzed by ICP-OES and HPLC to determine its metal (Fe ~ 9 g/L) and organic acid content (~ 11 g/L), with the goal of fully valorizing all process streams and optimizing subsequent recovery strategies. Ongoing preliminary experiments demonstrated the potential of this solution to undergo an NH_3-based treatment providing valued Fe_3O_4 particles (as described in the work of Goodarzi *et al.*, 2004) and a boron-rich aqueous solution.

4. Conclusion

The described integrated recycling process offers a selective, low-impact route for recovering REEs from EoL NdFeB magnets. With high lanthanide recovery and potential reuse of both solid and liquid fractions obtained through the leaching, the approach supports circular economy goals and presents a viable strategy to address critical material supply risks through sustainable innovation.

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