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A Simple Supramolecular Approach to Recycling Rare Earth Elements

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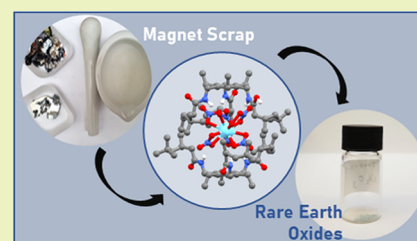
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ABSTRACT: The rapid increase in demand for rare-earth elements reflects their crucial roles in climate critical technologies. However, the lack of simple solutions for the separation of these metals from waste materials and ores represents a significant barrier to sustainable and environmentally benign rare-earth production. We report the application of a supramolecular approach to this challenge, using a triamido-arene receptor to selectively precipitate f-elements through their encapsulation as hexanitratometalates. Single-step, near quantitative recovery of Nd/Pr directly from magnet scrap was observed without the need for pH adjustment or pretreatment of the acidic leach solution. The rare-earth nitrate was rapidly stripped from the host–guest precipitate with water and the receptor recycled for further use. Near quantitative and highly selective uptake of La–Nd and Th from lateritic rare-earth ores was also achieved with no uptake of any non-f-element. These results show that targeting f-element metalates in separations chemistry can deliver exceptional and unique selectivity that may have significant consequences in the sustainable production of the rare-earth elements.

KEYWORDS: *Supramolecular, metal separations, rare earths, lanthanides, anion-recognition*



INTRODUCTION

Driven by their critical roles in low-carbon technologies, the demand for rare-earth elements (REEs) is projected to increase three to 7-fold by 2040.¹ Permanent magnets account for the largest share of REE use and, due to the increasing demand for electric vehicles, wind turbines, and electronics, this share is forecast to grow rapidly over the next ten years.²

As a result of the difficulties associated with the extraction and separation of REEs from primary sources, their production has a significant environmental cost. The global warming potentials (the amount of CO₂ released during the production of 1 kg metal) of Nd and Dy (17.6 and 59.6 kg CO₂ – eq/kg *cf.* 1.5 kg CO₂ – eq/kg for iron) are some of the highest for a bulk-produced metal.³ Recycling REEs from end-of-life (EOL) materials could also provide a solution to the “balance-problem”, avoiding excess production of La and Ce from ores when targeting Nd for permanent magnet manufacture.^{4,5} The modeling of future supply and demand pressures has suggested that, in the long term, recycling from EOL materials could provide as much as 50% of supply, reducing geopolitical supply risks and minimizing environmental impacts of REE production.⁶ However, as less than 1% of REEs are currently recycled from EOL products,^{1,7} there remains a critical need for new and efficient REE separation methods.

Several processes have been developed for the direct recycling of EOL REE-containing magnets through hydrogen processing or mechanical separation routes.^{8–11} These processes minimize some of the costs and waste associated

with metal recovery but require careful dismantling steps and predominantly single-source waste streams.¹² The majority of REE waste is present in the form of shredded material, where, among other reasons, the vast array of other metallic impurities often preclude direct recycling methods. As such, developing methods for the recovery of REEs from these waste streams is critical.

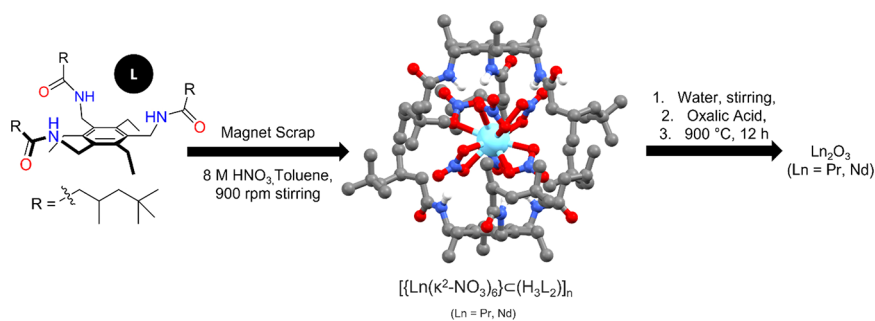
Hydrometallurgical processes have the capacity to purify metals from highly complex and dilute waste streams. Current industrial solvent extraction processes for REE recovery require a large number of separation steps, resulting in high mass intensities and costs.^{13,14} New solvent extraction and selective precipitation processes have been developed which achieve good separation between early and late REEs.^{15–23} However, these processes often require selective leaching steps or the pretreatment of feed solutions to remove high concentrations of non-REE metallic impurities. Furthermore, many of these processes cannot operate directly on highly acidic leach solutions and so require significant dilution or pH adjustment steps. The acids used to leach the metals into

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Scheme 1. Simplified Process for the Recovery of REEs from Magnet Scrap Using Triamido-Arene Receptor, L, to Selectively Precipitate Pr and Nd from the Leach Solution through the Formation of the Supramolecular Complex and the Subsequent Stripping of the Complex, Oxalate Precipitation, and Thermal Annealing to Yield the Oxide Product^a



^aThe complex representation is based on the previously reported Ce-complex structure.²⁵

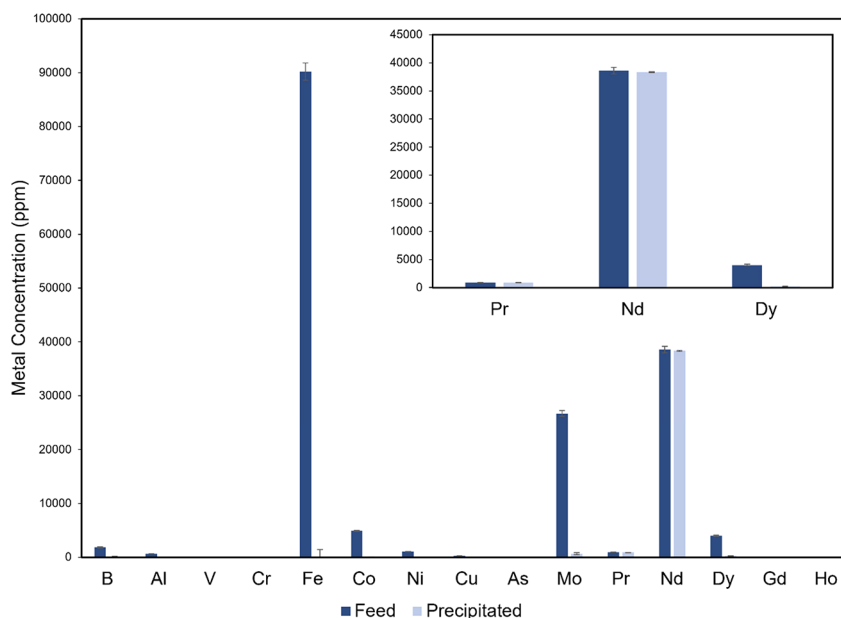


Figure 1. Concentrations of major metals present in the magnet scrap feed solution (dark blue) and precipitated with L (0.31 mmol) after 48 h stirring at 900 rpm (light blue). Inset, expanded view of major REE concentrations.

solution are thus consumed, preventing their reuse and increasing waste.

Recently, we reported the first example of a supramolecular receptor (L, Scheme 1) which, from acidic nitrate media, selectively precipitates early REEs (La–Eu) from metal mixtures through the formation of self-assembled hydrogen-bonded capsules.²⁴ The work represented the first example of a separation process that targeted REE metalates but its scope was limited to the investigation of lanthanide-lanthanide separations from synthetic mixed-metal solutions. As such, the application of this chemistry to the recovery and separation of REEs from real-world secondary sources and primary ores remains unexplored. Herein, we report the first application of supramolecular anion recognition chemistry to the recovery of REEs from real-world magnet scrap and ores and demonstrate its integration into a complete recovery process, from raw material through to the purified rare earth oxide product (Scheme 1).

RESULTS AND DISCUSSION

To replicate the challenging conditions associated with the recovery of REEs metals from waste, a mixture of shredded

hard drives, mobile phone and computer components was manually ground and reacted with 8 M HNO₃ at RT for 24 h. The resulting solution was filtered and analyzed by ICP-MS to determine the initial metal content (Table S1). The metal concentrations in the leach solution are representative of those typically found in this type of scrap.²⁶

One of the critical advantages of metalate anion separation processes is the capability to conduct effective separations without requiring the dilution of the leach solution. Typical cation or metal-salt extraction processes generally suffer from dramatically decreased recovery in these environments due to competition from high anion or proton concentrations. As such, a small portion of the leach solution (2 mL) was contacted with an equal volume of toluene and an approximate 5 times excess of L, relative to the calculated concentration of REEs in solution. The biphasic mixture was stirred for 48 h, resulting in the formation of a precipitate at the organic/aqueous interface (see Figure 2). The raffinate was analyzed to determine metal uptake into the precipitate.

After 48 h, greater than 98% precipitation of Pr and Nd is seen with no measurable precipitation of any other metal in solution, including the late REEs (Gd, Dy, Ho) (Figure 1).

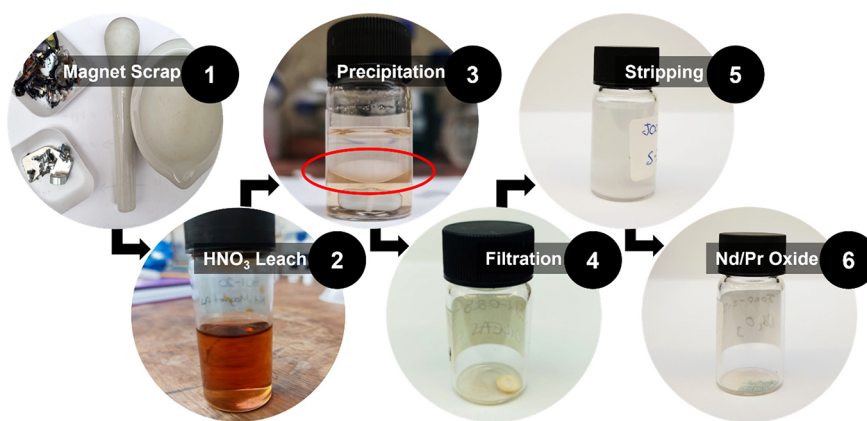


Figure 2. (1) Magnet scrap prior to grinding and leaching, (2) filtered leach solution, (3) biphasic precipitation system showing metal containing precipitate at the organic/aqueous interface, (4) filtered metal-loaded precipitate, (5) suspension of free ligand after stripping of precipitate with water, and (6) Nd/Pr oxide product after oxalate precipitation and calcination.

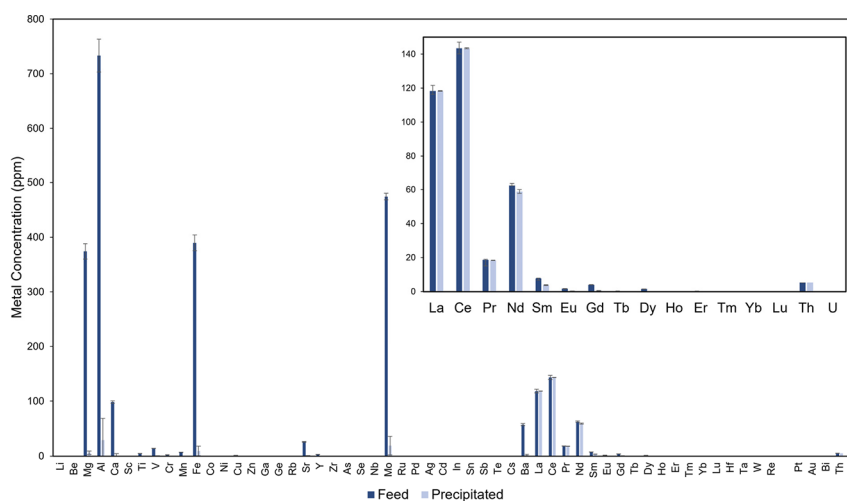


Figure 3. Concentrations of major metals present in ore sample feed solution (dark blue) and precipitated with L (0.06 mmol) after 48 h stirring at 900 rpm (light blue). Inset, expanded view of f-element concentrations.

This therefore represents a single-step, quantitative recovery of the early REEs. The metal-containing precipitate was isolated by filtration and characterized by powder X-ray diffraction (Figure S1). Pawley fitting of the powder diffraction pattern confirmed the precipitate adopts a similar supramolecular capsule structure as the La and Ce complexes previously characterized by single-crystal X-ray diffraction, and closely resembles those of the previously characterized Nd-containing precipitate.²⁴

Washing the isolated precipitate with water results in the quantitative release of the RE nitrate guest from the host receptor. This offers a significant advantage over cation and metal-salt extractants that typically require forcing conditions or pH changes to release the loaded metal. The receptor is straightforwardly recycled by filtration and is directly reused in subsequent precipitations. Over 90% loading and stripping of Nd was maintained during three load-strip cycles (Figure S3). Minor losses between cycles are attributed to solid losses during transfers between vials and filters.

The rapid and complete stripping of the precipitate arises from the unique nature of the supramolecular complex. The encapsulated hexanitratometalates²⁷ are not thermodynamically stable in aqueous conditions,²⁷ and it is only in very high concentrations of nitrate that these species transiently form to

be “trapped” out of solution by the receptor to form a stable host–guest precipitate. Consequently, interruption of the hydrogen bonding interactions in the capsules by addition of water results in the immediate and irreversible release of the metal. As such, it is possible to load REEs from very dilute feed sources and to strip into minimal amounts of water, thereby significantly concentrating the solution without the need for additional energy input or the risk of re-equilibrating the strip solution.

The strip solution was treated with oxalic acid to precipitate the REEs as their oxalate complexes. Filtration, isolation, and calcination of these solids at 900 °C yields the REE oxide product (Figure 2, step 6). Characterization of the oxide by PXRD confirmed the formation of the hexagonal-Ln₂O₃ product (Ln = 98.7% Nd, 2.3% Pr, based on ICP-MS analysis) (Figure S2).

To recover the remaining rare earth element (REE) content, primarily Dy, in the magnet scrap, solvent extraction employing the extensively studied N,N,N',N'-tetraoctyldiglycolamide (TODGA) was explored. Previous research has demonstrated that TODGA is selective for REEs while rejecting non-REEs but exhibits limited selectivity among the REEs at high nitric acid concentrations.^{28,29} Contacting the raffinate from which the early REEs had been precipitated with

a 0.1 M solution of TODGA in toluene results in >98% extraction of the remaining REEs (Gd, Dy, Ho) (Figure S4). After phase separation, the loaded organic phase is stripped through multiple contacts with water. The integration of these two systems presents an opportunity to devise a streamlined flow-sheet comprising just two separation steps for the efficient recovery of both early and late REEs from waste (Figure S5).

To further investigate the applicability of this supramolecular separation approach to industrially relevant feed solutions, a powdered lateritic REE ore sample from Australia was leached by 8 M HNO₃ (RT, 24 h). After filtration, a portion of the leach solution was, without pH adjustment, contacted with an equal volume of toluene and an approximate 5 times excess of the receptor L, relative to the calculated concentration of REEs present. After 48 h stirring, the precipitate was isolated and the raffinate analyzed by ICP-MS to determine metal uptake.

The initial leach solution comprised significant concentrations of Mg, Al, Fe, and Mo, with numerous other metals present in lower or trace concentrations. Despite this highly complex feed solution, near-quantitative (>94%) separation of the early REEs (La–Nd) is observed (Figure 3). In line with previously reported selectivity data for the system, uptake of the trans-Nd elements decreases sharply, with no recovery seen of any lanthanide beyond Gd. Significantly, no uptake of any non-f-element was observed, further highlighting the rigid selectivity imposed by the supramolecular receptor.

Radioactive elements such as Th and U seriously hinder the processing of certain rare-earth bearing ores. As expected, based on our previous investigations into actinide precipitation by L, quantitative uptake of thorium from the ore leach solution is observed along with the early REEs.²⁵ However, our previous work showed that the precipitation of An(IV) nitratometalates as host–guest complexes with the protonated receptor L is dependent on the concentration of nitrate. This feature suggests that separation of early REEs from Th is achievable, potentially through a selective stripping step which, at about 4 M HNO₃ could release the REEs from the host–guest complexes while retaining Th. Further investigation into these challenges is ongoing.

CONCLUSIONS

We have demonstrated the successful application of a supramolecular metalate anion receptor in the recovery of REEs from real-world waste and ores. Quantitative recovery and separation of Nd and Pr directly from concentrated acid leach solutions is seen, without the need for any dilution or pretreatment of the feed, minimizing the amount of nitric acid consumed during across the recovery process. The selectivity for f-elements and robust uptake in the presence of large excesses of non-f-elements, highlights the unique advantage of this metalate anion encapsulation system over traditional approaches. The system affords simple separation by filtration and rapid stripping with water alone, facilitating a significant concentration of the feed with no additional energy input. The receptor can be recovered for reuse in multiple precipitation cycles with minimal losses. The separated metals were successfully processed into their commercial oxide products through conventional oxalate precipitation/calcination steps. However, the ease with which the encapsulated metal can be released opens up the possibility of exploring stripping into nonaqueous media to enable direct electroplating of the

separated metal, thereby eliminating the CO₂-producing calcination step.^{30–32}

ASSOCIATED CONTENT

Data Availability Statement

Quantitative metal analyses and PXRD data are available upon request.

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.4c03063>.

Experimental details, materials, and methods; metal leaching and precipitation data; and powder X-ray diffraction data (PDF)

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Author Contributions

J.G.O'C.-D., B.O.I., and E.E.H. undertook all of the experimental work, analytical characterization, and X-ray crystallography. J.G.O'C.-D., C.A.M., B.T.N., and J.B.L. analyzed the data and wrote the manuscript. C.A.M., B.T.N., and J.B.L. directed the research.

Notes

The authors declare no competing financial interest.

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ABBREVIATIONS

PXRD, powder X-ray diffraction; ICP-MS, inductively coupled plasma mass-spectrometry

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