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ABSTRACT

Coal byproducts could be a promising feedstock to alleviate the supply risk of critical rare earth elements (REEs) due to their abundance and REE content. Herein, we investigated the economic and environmental potential of producing REEs from coal fly ash and lignite through an integrated process of leaching, biosorption, and oxalic precipitation based on experimental data and modeling results. Two microbe immobilization systems (polyethylene glycol diacrylate (PEGDA) microbe beads and Si sol-gels) were examined for their efficiency in immobilizing *Arthrobacter nicotianae* to selectively recover REEs. Techno-economic analysis revealed that North Dakota lignite could be a profitable feedstock when Si sol-gel is used due to its high cell loading and REE adsorption capacity as well as high reuse cycles. Life cycle analysis revealed that Si sol-gel based biosorption could be more environmental friendly than the prevailing REE production in China due to use of less toxic chemicals. However, fly ash sourced from Powder River Basin coals was neither profitable nor environmentally sustainable, primarily due to low solubility of high-value scandium at an economical pulp density (100 g ash/L of acid solution). To further improve the proposed biotechnology, future research could focus on scandium recovery, leaching efficiency at high pulp density, and reuse cycles of the immobilized microbes.

KEYWORDS: Rare earth element, Biosorption, *Arthrobacter nicotianae*, polyethylene glycol diacrylate (PEGDA) microbe bead, Silica sol gel

INTRODUCTION

Rare earth elements (REEs) are essential components of many high-tech electronics and low-carbon technologies including wind turbines, solar panels, and (hybrid) electric vehicles.[1,2] However, the absence of sufficient domestic REE production outside of China leaves many countries dependent on foreign imports and hinders the growth of several renewable energy technologies.[2] For example, the U.S. relies 100% on importation of individual REEs and rare earth metals, creating a vulnerability in the REE supply chain.[3] Because of this limitation, it is crucial to identify diverse sources of REEs and develop novel methods for REE extraction from unconventional sources. For instance, REEs were detected in the deep-sea mud of the Pacific Ocean (at thousands of parts per million (ppm)), the water produced from coalbed methane production ($28.99 \pm 10.79\ \mu g/L$), shale gas produced water (77 to 380 $\mu g/g$ of shales or solid wastes produced by water treatment), and acid mine drainages (a median concentration of 36.1 $\mu g/L$).[4,5] Recently, coal and coal byproducts have been recognized as one of the most promising feedstocks for REEs.[6]

Coal contains REEs at ppm levels.[7] Coal byproducts have an elevated REE concentration compared to coal by approximately an order of magnitude.[7] For instance, fly ash contains 250-800 ppm REEs.[8] As such, coal byproducts are abundant: from the more than 100 million metric tons of coal combustion products generated in the U.S. per year, approximately half are recycled for beneficial reuse applications.[8] The unused fly ash comprises approximately 8,000 tons of REE reserves per year.[8] However, these residuals are generally disposed of in landfills and surface water impoundments. REE recovery from coal byproducts may offer strong environmental and economic potentials compared to traditional sources.

REE recovery from coal byproducts has yet to be established for commercial production. Thus, techno-economic analysis (TEA) is critical for advancing proposed recovery processes. Das et al. (2018)[9] investigated REE extraction from coal ash, using supercritical CO_2 and tributylphosphate (TBP) to produce individual rare earth oxides (REOs). However, the cost was high ($680-$2,545 per kg of REO), and Sc contributed up to 90% of the revenue, highlighting the importance of Sc recovery. Zhang and Honaker (2018)[10] investigated natural leachate of coal coarse refuse pile using sodium hydroxide precipitation followed by re-dissolution, oxalic acid precipitation, and roasting to produce 94% total rare earth oxides (TREOs). Carlson (2018)[11] investigated REE recovery from fly ash, using sodium hydroxide pretreatment, HCl acid digestion, and REE separation by carbon adsorption and column chromatography to produce TREOs. Peterson et al. (2017)[12] also investigated REE recovery from fly ash, using nitric acid leaching and solvent extraction to produce 99% pure individual REOs. None of the above-mentioned studies reported a cost-effective process, highlighting the needs for further technology development and process optimization to lower the costs.

Another research gap is the lack of literature on quantifying environmental impacts associated with REE recovery from coal byproducts. It has been reported that adsorption-based technologies have the potential to minimize toxic chemical usage for REE recovery from low-grade feedstocks.[13,14] For example, a metal organic framework sorbent technology has been developed for REE extraction from geothermal brines, utilizing functionalized magnetic beads with chelating ligands.[15] Activated carbon has also been used for REE adsorption from leachates of different geological samples.[16] Although these technologies aimed to offer environmentally benign processes for REE recovery, the environmental impacts have never been quantified especially for coal byproducts. Therefore, environmental life cycle analysis (LCA) is necessary for closing the knowledge gap.

This work extends the prior research showing that bioengineered microbes can facilitate the selective extraction of REEs from the vast majority of non-REE impurities.[17,18] However, the industrial applicability of microbial biomass for REE recovery is currently limited by the energy and time-intensive centrifugation and/or filtration procedure required to separate biomass from the feedstock leachate. To apply microbial biomass for efficient and scalable rare-earth recovery in a flow-through format, we have developed procedures to immobilize cells at a high-density without compromising their adsorptive characteristics. To support the immobilization of microbes for REE recovery, PEGDA- and Si sol-gel crosslinked nanoparticles[19,20,21] have been developed to embed microbes in the polymer matrix.[22] This work focuses on TEA and LCA of PEGDA and Si sol-gel-based biosorption to evaluate the economic viability and environmental impacts for sustainable development of this biotechnology.

MATERIALS, PROCESSES, AND METHODS

Figure 1 depicts the overall process flow and system boundary of the current study. In brief, coal byproducts undergo pre-processing (crushing and milling) and acid leaching steps to solubilize REEs in preparation for biosorption. *Arthrobacter nicotianae,* is cultured in minimal medium and immobilized by PEGDA-[22] or Si sol-gel-based[19] platforms to selectively absorb REEs in the leachate. Then REEs are recovered from the cell surfaces in a subsequent desorption step, and the immobilized microbes are reused for further adsorption/desorption cycles. Finally, REEs are precipitated and further purified through oxalic acid precipitation and roasting steps to produce 95+% pure TREO.

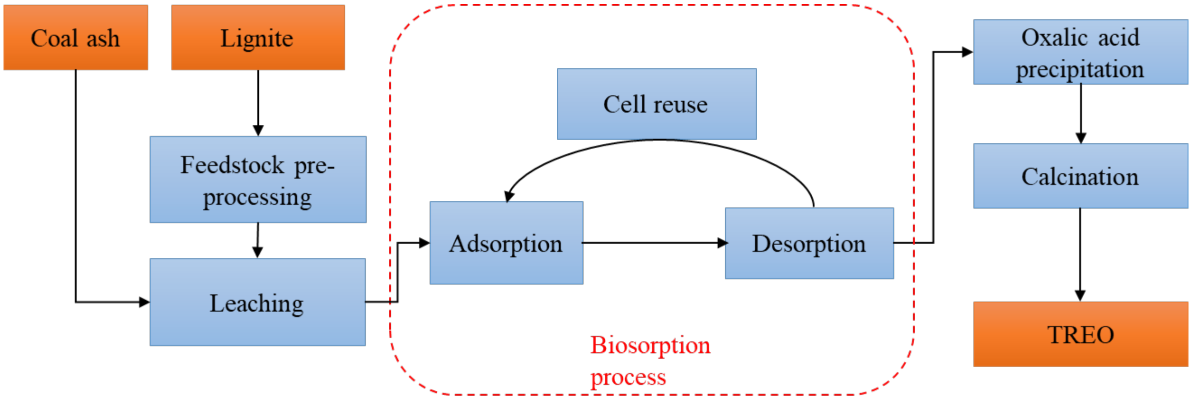

Figure 1. Process flow diagram and system boundary of our analysis.

Feedstock description

Two types of coal byproducts are considered in this research, including fly ash produced from the combustion of Powder River Basin (PRB) coal and the pre-combusted North Dakota (ND) lignite coal. They were selected based on their availability, REE content, and compatibility with biosorption processes (Table 1). PRB offers the largest low-sulfur subbituminous coal in the world and produces nearly 42% of the total coal in the United States.[23] To scale up the biosorption process, we assumed ~1.5% of the PRB coal fly ash is available for REE recovery, amounting to 200,000 tonnes/year.[24] North Dakota has the largest lignite reserve in the world from which 25 billion tonnes of lignite can be economically mined.[25] Similarly, we assumed 200,000 tonnes/year of ND lignite is available for REE recovery, which is a conservative estimate considering the size of the reserves. The same feedstock processing rate allowed us to compare the two feedstocks based on the proposed technology rather than confounding the technology impact with economies of scale resulting from assuming different feedstock processing rates.

Table 1. Feedstock characteristics.

	Feedstock source	
	Fly ash	Lignite
Location	Powder River Basin (PRB)	North Dakota (ND)
Feedstock availability	13 million tonnes/year[24]	25 billion tonnes[25]
Feedstock processing rate assumed in this study	200,000 tonnes/year for both feedstocks	
Mining	None	Yes
REE concentration range in solid phase feedstock*	233-406 ppm[8]	300-600 ppm[26]

*The individual REE content is available in Table S1.

Leaching

The leaching step for the PRB coal fly ash entailed 1 mol/L of HCl, tested in previous work with a range of pulp densities (10 to 125 g ash per L acid) for 4 hours at 85 ˚C.[6,27] The pulp density of 100g ash per L acid was selected (Figure S1) where maximum REE extraction efficiency (more than 80%)[27] was achieved while minimizing processing costs.

The leaching process for the ND lignite was based on prior work that utilized a source from an outcrop of the H-Bed seam in the Harmon-Hanson coal zone in Slope County, North Dakota

(Sample 6A).[25] This sample contained 551 mg kg^{-1} total REE (dry whole coal basis), and was leached in 0.5 M mineral acid based on a leaching procedure described in Laudal et al.[25] The coal to acid solution ratio was 60 g (dry mass) to 125 mL, and leaching lasted for 48 hours to solubilize 82-90% of REE (Figure S2).[25] The high REE extractability at high pulp density in lignite was attributed to REEs being predominately associated with organic complexes.[25]

A major distinguishing factor between leaching PRB fly ash and ND lignite is Sc extractability. Efficient extraction of Sc from PRB coal fly ash requires a much lower pulp density (e.g., 40g/L vs 100g/L leachate for Sc and REE, respectively; Figure S3); This is due to the acid consuming property of PRB, which results in Sc precipitation at elevated pulp densities.[27] Leaching at lower pulp densities results in (1) a higher degree of contamination or co-extraction of competing metals (e.g., aluminum and iron) that interfere with biosorption, and (2) higher leaching costs. On the contrary, Sc extraction from ND lignite was achieved using a pulp density (480g/L), which is much higher than that of PRB fly ash for non-Sc REEs (100g/L). Therefore, Sc extraction was pursued for ND lignite but not for PRB fly ash.

Biosorption

To apply microbial biomass for flow-through REE recovery in fixed-bed columns, we developed two distinct approaches to immobilize bacterial cells in high-density within a porous polymer matrix. In one approach, a bulk emulsification technique via UV-crosslinking was employed to immobilize microbes in polyethylene glycol diacrylate (PEGDA) hydrogel, forming microbe beads. The REE adsorption properties of the microbe beads under flow-through conditions were recently reported.[22] Briefly, the microbe beads exhibited an average diameter of 59 ± 24 μm, an adsorption capacity of 0.5 mg total REE/g of adsorbent when a complex coal leachate solution was used, with 87% REE adsorption capacity retained after 9 consecutive adsorption/desorption cycles. In the other approach, bacterial cells were embedded within a Si sol-gel matrix to form microbe particles.[20] Although this process yielded larger and more irregular-shaped particles relative to the spherical PEGDA microbe beads,[22] it enabled higher adsorption capacity (1 mg total REE/g of adsorbent, 0.37 mg Sc/g) and improved column stability: more than 92% of the adsorption capacity is retained over 12 adsorption/desorption cycles.[19]

A two-stage biosorption/desorption operation scheme has been developed for the sequential extraction of Sc and LN+Y from coal leachates based on prior batch biosorption data (Figure 2).[19] Lanthanides and Y can be extracted with high selectivity from ND lignite and PRB fly ash at pH 5.[28] However, Sc is not soluble at pH 5, precluding a single-step biosorption/desorption process. The high affinity of cell surface functional groups for Sc enables its selective extraction at a lower pH (3) where LN+Y recovery is minimal (<1%).[19] As such, following an acid leaching step to produce a pregnant metal solution, the pH is adjusted to 3 and the leachate is passaged over a microbe resin column where Sc is selectively adsorbed onto the bacterial surfaces. Weakly adsorbing LN+Y and base metals are collected in the flow through. Immediately prior to Sc breakthrough, the inlet feedstock flow is shut down and the microbe resin column is washed with a saline solution (0.9% NaCl) prior to circulating a small volume of citrate solution (10 mM, pH 6) to desorb and recover Sc. The Sc-depleted, Ln+Y pregnant flow through solution is adjusted to pH 5 to precipitate Al and most of Fe impurities and passaged over a larger microbe resin column for selective LN+Y adsorption, while weakly adsorbing alkaline earth and d-block metals are discarded in the flow-through. Immediately prior to REE (La) breakthrough, the microbe resin is rinsed with a saline solution and subjected to a citrate circulation step (5 mM, pH 6) to desorb and concentrate LN+Y. Following a saline rinse to remove residual citrate, both extraction columns can be subjected to subsequent adsorption/desorption cycles.

Our data suggest that both microbe resin types can be reused multiple times with some loss in REE adsorption capability defined by mg of REE recovered per g of biosorbent. As the capacity remained strong after ~12 reuse, we assumed that we can reuse the biosorbents up to 100 times and modeled the REE adsorption capability linearly as shown in Eqs. 1 and 2. Notably, PEGDA-based biosorbents lose REE adsorption capacity much faster than Si sol-gel, yielding a lower REE recovery efficiency per microbe resin over time. The biosorbent is a major cost driver of our proposed technology, so reusing it for multiple cycles is desirable. Sensitivity analysis will follow in a later section of this paper to identify the optimal reuse cycles.

REE adsorption capacity for PEGDA beads (mg/g) = -0.00815 * biosorbent use cycle + 0.50815 (Eq. 1)

REE adsorption capacity for Si sol-gel (mg/g) = -0.0027 * biosorbent use cycle + 1.013 (Eq. 2)

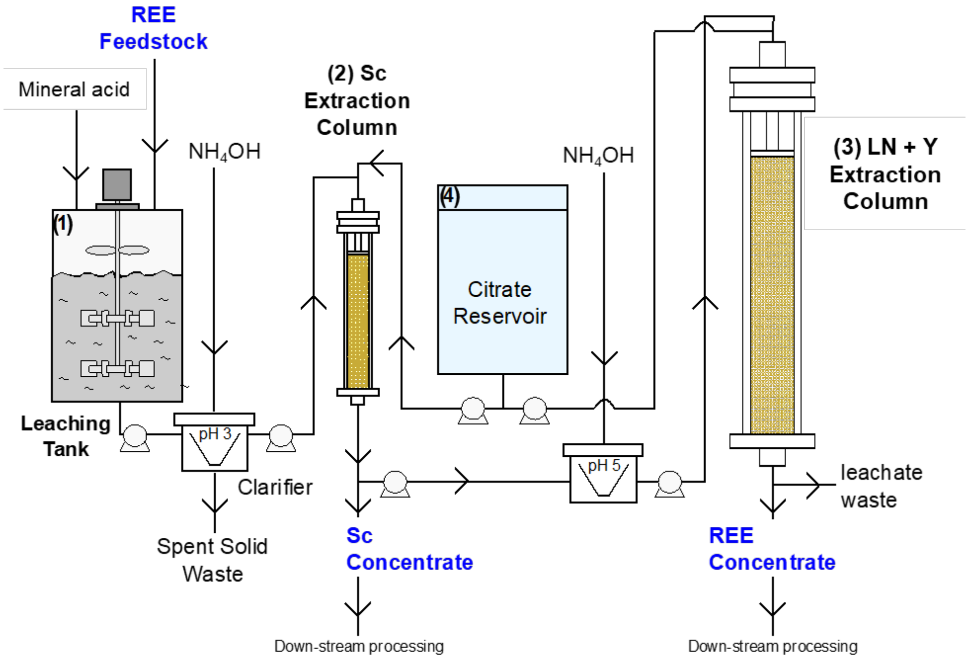

Figure 2. Two stage packed-bed bioreactor design and process flow diagram for sequential Sc and LN + Y recovery from North Dakota lignite. The packed bed columns are loaded with either PEGDA microbe beads or Si sol-gel microbe particles. (1) A coal byproduct leaching tank to generate a pregnant metal solution. (2) A small volume packed-bed bioreactor for Sc recovery. (3)

A larger volume packed-bed bioreactor for LN+Y recovery (LN stands for lanthanides). (4) A citrate reservoir for Sc and LN+Y desorption and biosorbent regeneration. NH_4OH is used for pH adjustment after leaching prior to biosorption when needed. PRB fly ash leachate (pH 4) skips the Sc extraction step and proceeds directly to pH adjustment (to pH 5) and the LN+Y recovery step (3), since soluble Sc is not generated using the current leaching process.

Techno-economic analysis

An industrial-scale adoption of the proposed biosorption technology was modeled based on a hypothetical processing plant that would be in operation for 20 years, and 8,000hours/year. Direct costs consist of (1) material costs associated with leaching, pH adjustment, biosorbent production, bacterial growth media preparation, desorption, and oxalic acid precipitation; (2) utility costs such as water and electricity for leaching, bioreactor, pumps, and roasting; and (3) other costs which include waste management, feedstock collection, operation and maintenance labor, and maintenance materials and supplies. The hourly wage of operating labor was assumed to be \$46.43[12] and the required number of workers was calculated based on Alkhayat et. al.[29] Operating supervision, quality control, maintenance labor and material, and operating supplies were calculated with factors established by Silla.[30] Capital cost includes major equipment cost[31] (i.e., leaching tank, clarifier, bioreactor for growing bacteria, columns for biosorption, sedimentation tanks, oxalic acid precipitation tank, and oven for REE roasting; see Table S4) and related infrastructures (e.g., land, buildings, and piping).[32] Indirect cost includes property taxes, insurance, fringe benefits, and overhead, which were calculated with factors.[30] General cost consists of administrative, marketing, research, and development costs with factors.[30] To account for various risks associated with the actual process implementation (e.g., equipment failure and supply delay) and future market conditions (e.g., cost increase), contingency costs were added to all the cost categories: 10% for the material cost, 100% for the capital cost due to large uncertainties,15% for all other costs.

It should be noted that the REEs prices[3] are currently available for 99+% pure individual REOs, whereas the proposed biosorption process delivers 95+% pure TREO. To estimate the revenue, we discounted the TREO price by 30% from the individual REO prices, similar to the Bear Lodge project.[33] After applying the discount rate, revenue from PRB fly ash was \$16/kg of TREO and that from ND lignite was \$344/kg of TREO.

Net Present Value (NPV) was used to evaluate the economic potential of the proposed REE recovery process. Eq. 3 shows that NPV is calculated by aggregating the future net cash flows (NCF_t) discounted by i (i.e., 8% in this study) over the project life n (i.e., 20 in this study). More TEA assumptions are available in Supporting Information Table S3, and the specific TEA data are available in the Supporting Information (two Excel files).

$$NPV = \sum_{t=0}^{n} \frac{NCF_t}{(1+i)^t} \qquad \text{(Eq. 3)}$$

Life cycle assessment

LCA was conducted to quantify the environmental impacts associated with REE recovery from coal byproducts using the biosorption approach. The LCA system boundary starts with feedstock collection followed by acid leaching, media preparation, biosorption, oxalic acid precipitation, and roasting. The specific material flow for each processing step is available in Figures S4 and S5, with relevant experimental data from Middleton et al,[27] Park et al,[18,22] and Jin et al,[34] and Laudal et al.[6,25]

Table 2 shows the unit processes for recovering 1 kg of mixed REOs from ND lignite, and those from PRB fly ash are shown in Table S6 for brevity. The functional unit was set as 1 kg of REO.

Table 2. Unit processes for recovering 1 kg of mixed REOs from ND lignite.

Unit process	*Process*	*Value*

		PEGDA	Si sol-gel	Unit
Mineral acid {RoW}| production | APOS, U[**]	Leaching	2.51E+02	2.51E+02	kg
Ammonium hydroxide[*]	Leaching	4.78E+00	4.78E+00	kg
Water, completely softened, from decarbonized water, at user {GLO}| market for | APOS, U	Leaching	6.46E+03	6.46E+03	kg
Transport, freight, lorry, unspecified {RoW}| market for transport, freight, lorry, unspecified | APOS, U	Leaching	1.20E+01	1.20E+01	tkm
PEGDA microbe beads/Si-sol gel particles[*]	Microbe bead production	1.13E+02	1.11E+01	kg
Disodium phosphate[*]	Biomass production	1.38E-03	1.38E-03	kg
Monopotassium phosphate[*]	Biomass production	8.40E-01	8.40E-01	kg
Ammonium chloride {GLO}| market for | APOS, U	Biomass production	7.92E-01	7.92E-01	kg
Glucose {GLO}| market for glucose | APOS, U	Biomass production	3.17E+00	3.17E+00	kg
Magnesium sulfate {GLO}| market for | APOS, U	Biomass production	9.51E-02	9.51E-02	kg
Calcium chloride {RoW}| market for calcium chloride | APOS, U	Biomass production	8.80E-02	8.80E-02	kg
Iron sulfate {RoW}| market for iron sulfate | APOS, U	Biomass production	2.41E-03	2.41E-03	kg
EDTA, ethylenediaminetetraacetic acid {GLO}| market for | APOS, U	Biomass production	4.63E-03	4.63E-03	kg
Sodium citrate[*]	Post processing	5.95E-01	5.95E-01	kg
Citric acid {GLO}| market for | APOS, U	Post processing	1.35E+00	1.35E+00	kg
Electricity, medium voltage {GLO}| market group for | APOS, U	Leaching, biomass and microbe bead production	4.77E+01	4.77E+01	kWh
Emission to water				
Wastewater		6.46E+03	6.46E+03	kg

[*] Refer to Tables S7-S12 for more details.
[**] Confidential data not disclosed herein.
Citric acid was used as a proxy for oxalic acid since both can be industrially produced from carbon monoxide.[35]
RoW, rest of the world, GLO, global, APOS, allocation at the point of substitution, and U, unit process.

As the output from biosorption includes various REOs, the individual impact of recovering Nd_2O_3 was estimated using economic allocation. Different feedstocks entail different REE compositions, so direct comparison of LCA results based on mixed REO is not feasible. Nd_2O_3 was chosen because (1) it is a critical material with an ever increasing demand in electric vehicles and wind turbines,[36] and (2) it is well-studied in the LCA literature, allowing a comparison of the environmental performance of the proposed technology with other REE production routes.[37,38,39,40,41,42] Economic allocation was chosen because (1) it is the preferred method when co-products offer significantly different values[43] (e.g., in our case, scandium oxide is sold at $4,689/kg, whereas lanthanum oxide at $2/kg) and (2) revenue is a driving force of value recovery without which the proposed process may not take place and therefore, economic incentives drive environmental impacts.[44] Table S7 shows the specific allocation factors. Since the focus is REE recovery in the U.S., TRACI (Tool for Reduction and Assessment of Chemicals and Other Environmental Impacts) developed by the U.S. Environmental Protection Agency, was used to classify and characterize the environmental impacts.[45]

RESULTS AND DISCUSSION

Techno-economic assessment

The economic feasibility of our recovery process varies significantly depending on the feedstock type and microbe immobilization system (assuming 100 reuse time for both systems as the baseline scenario) (Figure 3). Overall, ND lignite is less costly to process than PRB fly ash, and Si sol-gels are less costly than PEGDA beads per kg of TREO recovered (Table S5).

Recovery cost with PEGDA microbe beads

The total cost to recover 1 kg of TREO is estimated to be $2,853 for PRB fly ash and $1,635 for ND lignite, with the difference primarily attributed to leaching cost. As PRB fly ash requires a lower pulp density compared to ND lignite, the leaching cost is ~12 times higher per ton of feedstock processed. In addition, the REE concentration affects biosorbent costs per ton of feedstock processed (i.e., the more REEs to recover, the more cells and PEGDA beads are required), so ND lignite has a higher PEGDA bead cost than PRB fly ash. The main cost drivers for REE recovery from PRB fly ash are leaching (25%) and PEGDA beads (34%), and that for ND lignite is PEGDA beads (66%), at the annual feedstock processing rate of 200,000 tonnes (Figure 3).

Recovery cost with Si sol-gel microbe particles

The total cost to recover 1 kg of TREO is estimated to be $1,634 for PRB fly ash and $297 for ND lignite. As Si sol-gel beads have a higher cell loading and maintain a higher REE adsorption capacity after multiple reuse cycles compared to PEGDA beads, their overall biosorbent cost is much lower (Figure 3). More specifically, Si sol-gel beads only contribute 5% of the total cost for REE recovery from PRB fly ash (as opposed to 34% when PEGDA beads were used). The major cost drivers for ND lignite are Si sol-gel beads (32%) and leaching (12%).

It should be noted that PEGDA- and Si sol-gel-based microbe immobilization for REE recovery is a technology at early stages of development and significant improvements (e.g., in adsorption capacity) are expected as the technology matures. Therefore, if the technical hurdle of low cell loading is overcome in the future, PEGDA could be also attractive economically.

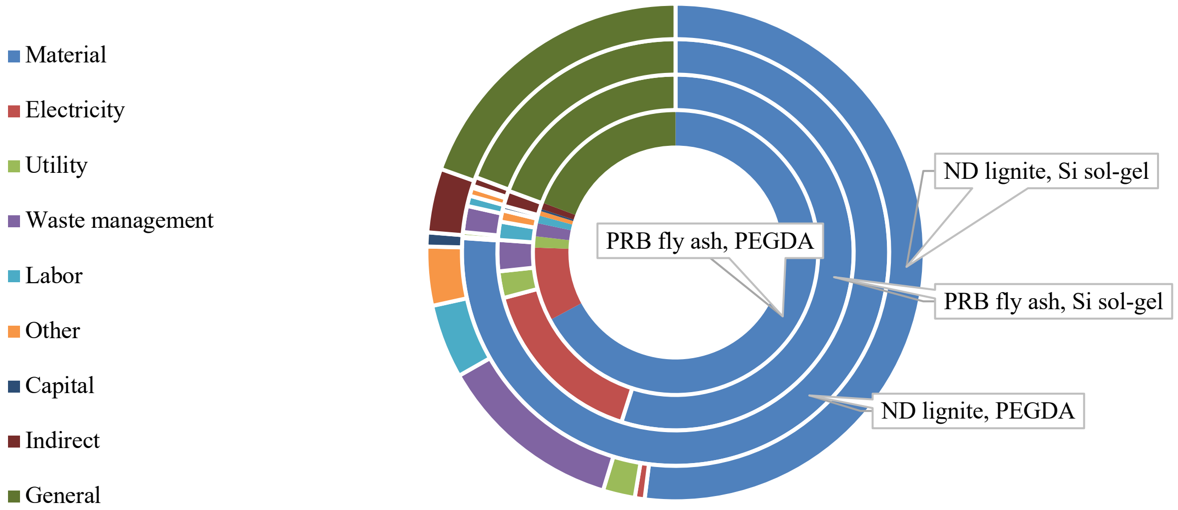

Figure 3. REE recovery cost breakdown from PRB fly ash and ND lignite at an annual feedstock processing rate of 200,000 tonnes.

Revenue from REE recovery

The revenue is estimated to be \$16/kg TREO for PRB fly ash and \$344/kg TREO for ND lignite, with the difference attributed to REE concentration and more significantly Sc extractability. ND lignite samples had higher REE concentration than PRB fly ash (551ppm vs. 337 ppm), yielding higher REE yield with the same feedstock volume. Scandium oxide constitutes about 94% of the total revenue from ND lignite, so the TEA results are sensitive to the scandium oxide price. As the historical scandium oxide price was volatile, we used the average price over the last 9 years (i.e., \$4,689/kg from 2011 to 2019) reported by the USGS Mineral Commodities Summary report to smooth out the price variation.[3,46,47,48,49,50,51,52,53] As a result, the profitability is highly dependent on Sc content in the feedstock, its recovery efficiency, and sales price, highlighting the need to focus on Sc recovery and market dynamics for economic reasons. Due to the inability to recover Sc from PRB fly ash, our procedure outlined is not economically viable for PRB fly ash. However, ND lignite is projected to be a profitable feedstock using the Si sol-gel biosorbent, yielding a net profit of \$47/kg TREO. NPV is estimated to be more than \$28M over 20 years (see Supporting Information Table S3 for more details on the financial assumptions and Table A.5 for the projected cash flow).

Table 3 compares the TEA results of our approach with other literature for REE recovery from coal byproducts. Das et. al[9] was the only study that reported a profitable process, which used coal ash located in Poland with a higher REE concentration (934 ppm) than feedstocks used in this study.

Table 3. Summary of techno-economic analysis on REE recovery from coal byproducts.

Data source	Feedstock	REE content (ppm)	REO output quality	Revenue (\$/kg REO) (B)	Total cost (\$/kg REO) (A)	Profit rate (B-A)/(A)
This paper	PRB fly ash	337	95% TREO	16	PEGDA 2,853 Si-sol gel 1,634	-99% -99%
This paper	ND lignite	551	95% TREO	344	PEGDA 1,635 Si-sol gel 297	-79% 16%
Das et al. (2018) [9]	Coal ash	608-934	Individual REO	577-1,150	680-2,545	-73%~30%
Zhang & Honaker (2018)[10]	Natural leachate of refuse in Kentucky	7	94% TREO	29	34	-15%
Carlson (2018)[11]	Louisville fly ash	480	TREO	33	2,669	-99%
Peterson et al. (2017)[12]	Ohio fly ash	532-558	Individual REO	179	235	-24%

Life cycle assessment

The LCA results for recovering 1kg of Nd_2O_3 from ND lignite after economic allocation are shown in Table 4, and those from PRB fly ash in Table S14. Similar to the TEA results, ND lignite was more preferable than PRB fly ash due to the higher leaching efficiency and successful recovery of high-value Sc. The Si sol-gel sorbent showed lower environmental impacts than the PEGDA sorbent due to the higher adsorption capacity of REEs (and thus less material assumption) and the chemicals involved in the production of the polymer materials (Figure S6, Table S15).

Table 4. Life cycle impacts for recovering 1 kg of neodymium oxide from North Dakota lignite for both PEGDA and Si sol-gel biosorbents. The results are calculated after applying economic allocation, revealing the impacts of Nd_2O_3 only.

Impact category	Unit	PEGDA (A)	Si sol-gel (B)	A/B
Ozone depletion	kg CFC-11 eq	1.38E-05	3.93E-06	3
Global warming	kg CO_2 eq	9.67E+01	1.81E+01	5
Smog	kg O_3 eq	5.74E+00	1.54E+00	4
Acidification	kg SO_2 eq	6.46E-01	3.40E-01	2
Eutrophication	kg N eq	2.52E-01	7.04E-02	3
Carcinogenics	CTUh	3.59E-06	8.47E-07	4
Non carcinogenics	CTUh	1.46E-05	3.90E-06	4
Respiratory effects	kg PM2.5 eq	1.07E-01	3.49E-02	3
Ecotoxicity	CTUe	3.39E+02	8.33E+01	4
Fossil fuel depletion	MJ surplus	2.86E+02	6.32E+01	4

As the next step, the environmental hotspots (i.e., the major materials/energy/emissions that contribute significantly to the environmental impact) were identified for REE recovery from both feedstocks. For REE extraction from PRB fly ash using PEGDA beads, the environmental hotspots were cell immobilization (46% of the total global warming potential), and hydrochloric acid (39%) while hotspots using Si sol-gel were hydrochloric acid (67%) and electricity (22%) used for heated leaching and roasting (Figure S7). For REE+Sc extraction from ND lignite using PEGDA beads, the environmental hotspot was solely from cell immobilization (88%), while hotspots using Si sol-gel beads were cell immobilization (41%), electricity for growing cells and roasting (28%), and mineral acid (11%) (Figure S8). Overall, the impact from leaching acid was 37%-56% lower for

ND lignite compared to PRB fly ash due to the use of a less hazardous acid and lower acid consumption.

The environmental impacts of our approach were compared with two dominant REO production routes in China: mining and REO production from (1) bastnäsite and monazite in Bayan Obo, Inner Mongolia, and (2) ion adsorption clays (IAC) in South China for 92% mixed REO.[37] Figure 4 depicts the life cycle impacts of recovering 1 kg of Nd_2O_3 (mixed with other REOs) from the specified feedstocks. Our proposed REE recovery process has a comparable purity and lower environmental impacts than Chinese production for all the impact categories of TRACI except for fossil fuel depletion. The higher fossil fuel depletion impacts of our process are attributed to (1) the lower REE concentration in the feedstock (i.e., 551 ppm for ND lignite vs. 6% in bastnäsite and monazite for Bayan Obo[37]) and (2) the different leaching methods (i.e., 251 kg of mineral acid required for 1 kg of REO recovery from ND lignite vs. ~3.3kg of hydrochloric acid and ~ 2.7kg of sulfuric acid are required for bastnäsite and monazite in Bayan Obo[37,54] vs. 10.4 kg of ammonium sulfate for IAC in South China[35]). For the same amount of REEs recovered, our process requires more acid to solubilize a higher volume of feedstock compared to REE recovery from ores in Bayan Obo or IAC.

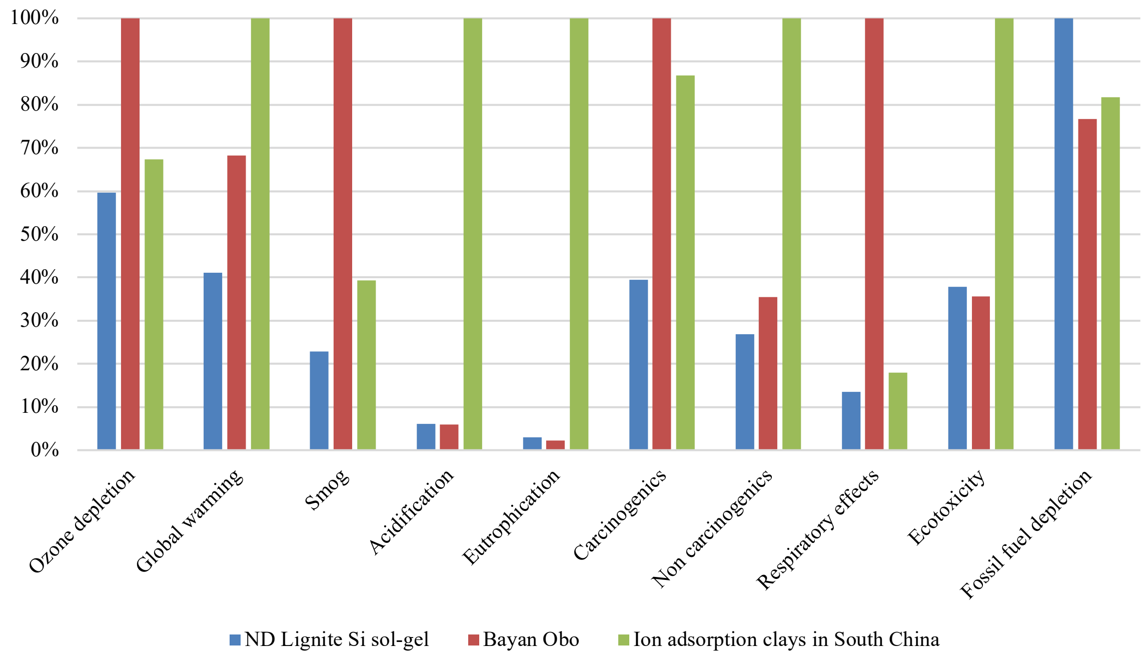

Figure 4. Comparison of environmental impacts for recovering 1 kg neodymium oxide from Chinese production routes and our biosorption approach.

Sensitivity analysis and future direction recommendations

As the REE market is volatile and biosorption is a novel technology that has not been commercialized/tested at the pilot scale, there are significant uncertainties in our TEA and LCA results. To explore the sensitivity of our analysis, the net profit effects from changing the main economic and environmental factors are shown in Figure 5 (TEA) and Figure S9 (LCA). By examining historical prices of mineral acid[55] and scandium oxide[3,46,47,48,49,50,51,52,53] in the last 9 years (2011-2019), it can be found that the maximum and minimum prices of mineral acid were in ±11% of our TEA assumption ($140/tonne) and scandium oxide price ranged between -17% and +9% of our assumption ($4,689/kg). Therefore, the sensitivity analysis was performed in ±15% range.

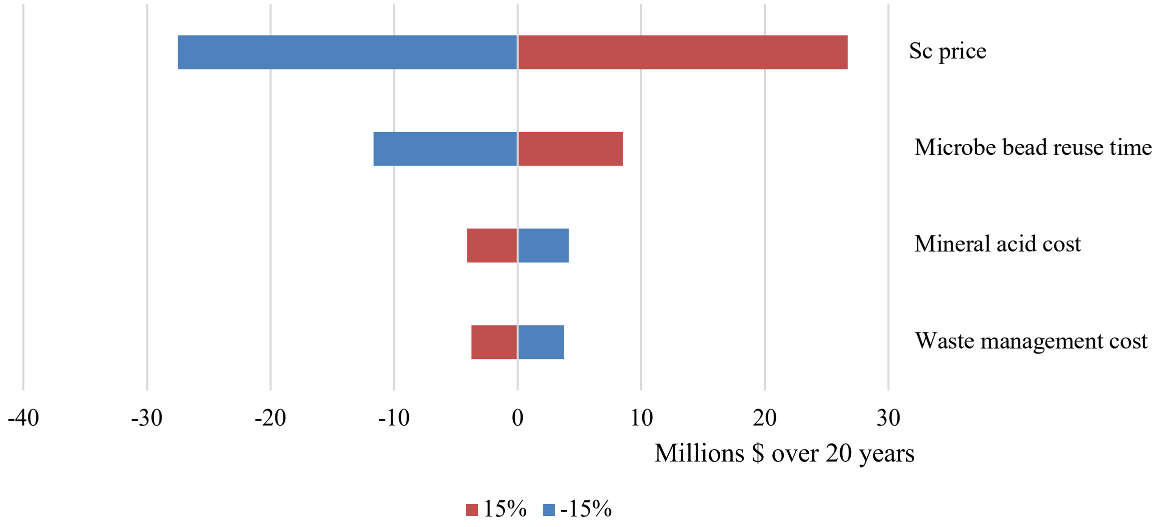

Figure 5. Sensitivity analysis on the net present value by changing each major economic factor by 15% for REE recovery from ND lignite using the Si sol-gel biosorbent.

Sc price: REE composition is different across feedstocks, and Sc plays the most significant role in the economic feasibility for our process (Figure 5) as well as others reported.[9,10] The market size of Sc is limited (~10 tons/yr worldwide), and there are multiple competing technologies emerging worldwide specifically for Sc extraction.[56] Therefore, Sc price could be susceptible to a decrease in the future due to potential market saturation. A reliable Sc source, on the other hand, could support robust growth in several promising applications such as aluminum-scandium alloys and solid oxide fuel cell markets. Based on our TEA, we project that the breakeven price of Sc oxide (99+% pure) would be 87% of the current assumption ($4,075/kg) below which our proposed REE recovery process would not be profitable. The environmental impacts of our process are also highly dependent on the Sc price due to economic allocation. Variation in the REO prices can alter the allocation factors used for the LCA: If Sc oxide price decreases, the environmental impacts of Nd_2O_3 recovery (i.e., our functional unit) would increase, as more weights are allocated to Nd_2O_3.

Acid consumption cost: Acid consumption in the leaching step is one of the major cost drivers and environmental hotspots for both PRB fly ash and ND lignite. Any effort to reduce acid consumption, such as recycling residual acids or increasing pulp density without a significant loss in REE leaching efficiency, will have a major impact on the economic and environmental performances of the recovery process.

Waste management: The biosorption process itself does not introduce any acid or harmful chemicals to the environment. Therefore, waste management focuses primarily on wastewater and leachate treatments. Unlike fly ash which is regulated for disposal and thus is already stored in ash ponds, ND lignite residuals are assumed to go through additional dewatering and disposal processes (with associated costs included), in addition to the paste tailing storage cost of \$0.13/kg.[57] However, there is an on-going study showing that lignite could be upgraded during REE recovery so that burning it for energy not only eliminates the paste tailing storage cost but adds more value due to the resulted cleaner coal.[58] Fly ash is often stored in coal ash ponds due to the hazards associated with airborne particles and potentially toxic trace elements (e.g., arsenic, mercury, and selenium),[59] and thus we did not assume any credits for processing them or any liabilities for placing them back in the ponds. Arsenic form hydrated oxyanions is not adsorbed by the cell surface and remains in the leachate solution.[34] Small amounts of uranium and thorium are adsorbed by the cell surfaces, but they are expected to be separated from desorbed REEs after the oxalic acid precipitation step.[60]

Cell immobilization and biosorbent reusability: REE absorption capacity and reusability of the biosorbent are keys to the economic and environmental performances. Si sol-gel exhibited a higher absorption capacity and greater reuse potential based on the current performance, which could be improved further as the technology matures. The REE adsorption capacity of biosorbents decreases, as the number of reuse cycles increases (Eqs. 1-2). Figures S10 and S11 show that when the biosorbent reuse cycles reach 61 times for PEGDA and 370 times for Si sol-gel, the net present values would decrease afterwards. The sensitivity graph (Figure S11) showed that if Si sol-gel is reused less than 75 times, the proposed biosorption approach would not be profitable.

Challenges of biosorbents for REE recovery: A major advantage of the presented biosorption process is that it is decoupled from cell viability, which expands the allowable range of operating conditions (e.g., solution composition, pH, temperatures) and enables multiple reuse cycles. However, there are two potential challenges associated with the proposed method. First, matrix materials could wear and tear due to repeated exposure to potentially corrosive fluids (e.g., feedstock leachates and citrate desorbent), leading to biomass loss over time, which is partially reflected in Eqs. (1-2). Second, although encapsulation likely protects the cells against decomposition to some extent, biomass could be decomposed by chemical or biological (e.g., predation) processes during prolonged operation, leading to biosorbent fouling.[61,62] Further engineering and testing will be required to fully gauge the impacts of these effects and overcome technical challenges that arise.

Conclusion

TEA and LCA results confirmed the economic and environmental potentials of adopting the proposed biosorption technology for REE recovery from coal byproducts. Based on our comparison of ND lignite with PRB fly ash, we found that the biosorption technology was well-suited for REE recovery from low-grade feedstocks that can be leached with minimal acid consumption and contain soluble Sc content. The analysis helped optimize pulp density and biosorbent reuse cycles for sustainable process development and scale-up. Future work may be directed at scandium recovery from fly ash, improving leaching efficiency at a high pulp density, testing more efficient leaching method such as alkali roasting before leaching, and field validation of biosorbent reusability on a large scale.

Supporting Information.

The Supporting Information is available free of charge on the ACS Publication website. A PDF file including additional information and two Excel files demonstrating TEA and LCA calculations.

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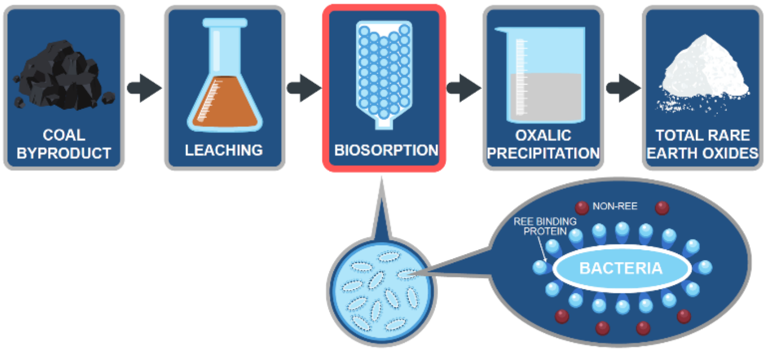

Synopsis: PEGDA and silica nanoparticle-based biosorption could offer an economical and environmentally sustainable pathway to recover rare earths from coal byproducts.