Rare-Earth Elements Recovery from Electronic Waste: Techno-Economic and Life Cycle Analysis

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**ABSTRACT:** Rare-earth elements (REE) possess unique magnetic, luminescent, and catalytic properties, making them key resources for diverse applications. They are critical components in electronic devices, industrial components, military defense equipment, and various clean energy technologies. Electronic waste (e-waste) constitutes a rich source of REE. E-waste REE recovery can lower the dependence on mining, stabilize REE market prices, and reduce the environmental impact of landfilling. Thus, researchers have been focused on developing technologies to recover REE from e-waste. This article presents a technoeconomic (TEA) and environmental (LCA) assessment of didymium oxide recovery from hard drive shreds through acid-free dissolution recycling. The goal of this study is to assess the economic feasibility and the environmental impacts of the process. Results show that a facility with an annual processing capacity of 342.42 tonnes of hard drive shreds per year collects 2.53 tonnes of didymium oxide. The estimated minimum selling price (MSP) for didymium oxide is \$130/kg. Sensitivity analysis suggests that further optimization of the recycling technology could potentially reduce the MSP to approximately \$73/kg. The greenhouse gas (GHG) footprint is estimated at 4.91 kg CO_2/kg REE. Sensitivity analysis shows that REE and $CuSO_4$ recovery efficiency and HDD content are the primary factors impacting the MSP. A comparison of the hydrometallurgical and electrometallurgical processes showed that acid-free dissolution is an attractive e-waste strategy.

KEYWORDS: e-waste recycling, rare-earth elements, techno-economic analysis, life cycle assessment
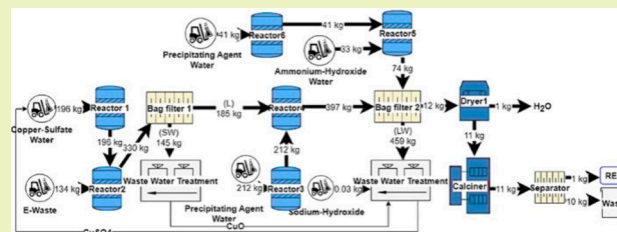
■ INTRODUCTION

Rapid technological development growth has led to an exponential increase in the global sales of electrical and electronic equipment, but limited recycling and reuse options have led to significant waste generation. Global electronic waste (e-waste) generation in 2019 was 53.6 million tonnes.[1] Although pollution concerns are driving government policies to limit landfilling,[2] recycling and reuse of valuable components (e.g., precious metals) of e-waste presents an economic opportunity. The market for managing electronic waste (e-waste) is expected to grow to \$145.11 billion by 2030 with a 13.2% growth rate.[3] E-waste recycling is driven by economic, environmental, and societal factors. With such potential, recycling and reuse of e-waste can reduce the global competition for key materials and commodities, which currently drives the search for alternatives to mining from natural sources. Rare-earth elements (REE) are a valuable resource in electronic equipment. The global REE demand was 119,650 metric tonnes in 2015, and it is expected to grow 5% per year.[4] REEs are expensive and often scarce. Hence, there is a strong need to develop new recycling technologies to ensure a sustainable worldwide supply of REEs and support the market for many dependent applications, including electronic products.[5]

The U.S. Department of Energy (DOE) has classified some of the REEs as critical materials due to their importance in national energy security and the economic growth of advanced technologies used in many sectors of the U.S. economy. Moreover, China accounts for most of the global production of REE. Such geographical concentration of the critical REEs creates concerns for global supply disruption, which has motivated countries to develop other ways to recover REE without requiring mining. One way is to recover REE from e-waste. This can help in the effort to reduce the environmental impacts of REE landfilling, stabilize global REE market prices, and lower the dependence on REE mining.[6]
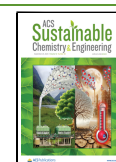
The most critical REEs are dysprosium (Dy), terbium (Tb), neodymium (Nd), and praseodymium (Pr). These REEs are used in rare-earth permanent magnets contained in many electronic devices. The most widely used type of rare-earth magnet is the neodymium—iron—boron (Nd—Fe—B) magnet. This is a permanent magnet containing Nd and Pr, and the
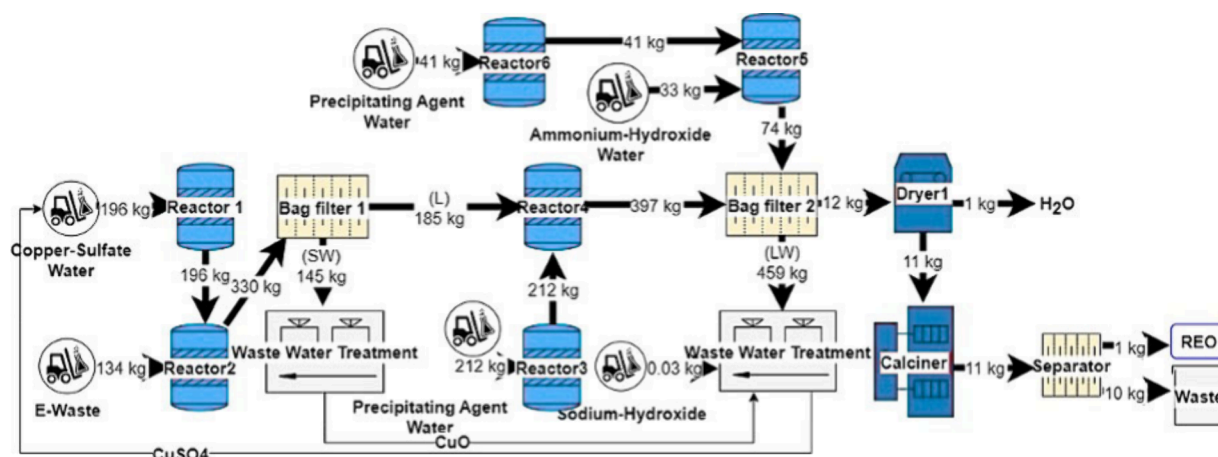
Figure 1. Process flow diagram for recovering 1 kg of REO from 134 kg of HDD shreds. Copper valorization will supply back some copper sulfate, so only 5 to 6 kg of copper sulfate will be consumed per kilogram of REO recovered.

mixture of these two elements is referred to as didymium.[7] Dy and Tb are often added to enhance the high-temperature performance of the magnets. The Nd−Fe−B magnets typically contain approximately 31 wt % of rare-earth metal, and they are used in a wide variety of applications such as wind turbines, automobiles, hard disk drives (HDD), mobile phones, video and audio systems, etc.[8,9] Researchers state that recycling of REE from U.S. HDD could replace up to 5.2% of the world's (excluding China) Nd−Fe−B magnets demand.[10]

REEs are commonly found in oxide form and are processed into alloys. The worldwide demand for didymium oxide in 2021 was around 65,000 tonnes and is expected to grow to 140,000 tonnes per annum by 2030. The increase in the demand for these magnets from 2020 through 2030 is forecasted to be at a compound annual growth rate of 9.7%, and it is expected to reach a peak with an annual increase of 12.5% in 2035. Due to the increasing demand and the shorter life cycles of electronic devices, researchers are trying to develop more cost-effective and sustainable ways to recover REO from e-waste.[11,12]

Several studies have reported various methods for REE recovery from waste Nd−Fe−B magnets.[13] Many studies report the recycling of REO from e-waste through pyrometallurgical and hydrometallurgical approaches due to their high efficiency and rapid recovery. Nonetheless, the main disadvantage of both techniques is that they generate environmental pollutants. Pyrometallurgical treatment requires high-temperature processes that consume a large amount of energy. Firdaus et al. analyzed previous studies on the pyrometallurgical recovery of REO from magnet waste[14] and suggested that e-waste recycling techniques need optimization. On the other hand, the hydrometallurgical method involves the use of strong chemicals, which lead to the formation of large quantities of wastewater. Iannicelli-Zubiani et al. estimated the environmental impact of a hydrometallurgical treatment of waste electrical and electronic equipment through the life cycle analysis methodology and concluded that the environmental performance could be improved by optimizing the absorption step.[14−17]

To fill these research gaps, this study presents a techno-economic analysis (TEA) and life cycle assessment (LCA) of didymium recovery from HDD shreds through acid-free dissolution recycling.[18] To our knowledge, this study presents the most comprehensive sustainability study of acid-free

dissolution recycling of HDD shreds. The acid-free dissolution recycling technology has gained substantial national and global recognition due to its unique features. The technology is now being commercialized and the present study represents the first TEA and LCA investigation at high technology readiness level.[19−21] The TEA and LCA studies are developed to investigate the economic and environmental impacts of this innovative recycling process and determine key parameters for future research and development.[18,22]

MATERIALS AND METHODS

Process Design. This case study evaluates didymium oxide recovery at a single facility in the United States capable of processing about 342 tonnes per year of HDD shreds using various solvents to isolate didymium oxide or Di_2O_3. The U.S. imports about 17,430 tonnes per year of HDDs (0.108 kg/HDD).[23] This facility is designed to produce 2.53 tonnes of REO per year. To evaluate the feasibility of this e-waste recycling project, the process model was designed by using an Aspen Batch Process Developer. Figure S1 shows the 3D process diagram of the equipment and plant distribution design. A flowchart of the e-waste recovery pathway is illustrated in Figure 1. In this process diagram, e-waste refers to the HDD shreds. The recovery process employs various solvent combinations and calcination to produce REO. Summary information on the material balance for HDD shreds is shown in Table 1, which agrees with published data: aluminum, ≤55.0 wt %; iron, ≤22.5 wt %, copper, ≤5.5 wt %; cobalt, ≤2.5 wt % (our data); nickel, ≤1.75 wt %; zinc, ≤0.7 wt %; didymium (Nd, Pr), ≤1.0 wt %.[24] The concentrations of boron and dysprosium in the HDD are negligible and unaccounted for in this study. All the waste coming out of bag filters 1 and 2 was considered as electronic scrap except for water, which was considered as wastewater.[25,26]
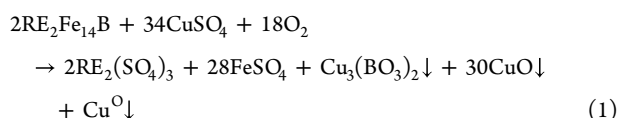
Table 1. Material Balance to Recover 1 kg of REO from HDD Shreds through an Acid-Free Dissolution Process

input	quantity	unit	output	quantity	unit
copper sulfate	5.66	kg	**REO recovered**	1.00	kg
precipitating agent	3.35	kg	copper oxide	1.65	kg
e-waste (HDD shreds)	133.63	kg	precipitating agent	2.91	kg
ammonium hydroxide	1.14	kg	iron	1.43	kg
sodium hydroxide	0.03	kg	ferrous sulfate	3.26	kg
water	243.60	kg	copper borate	0.24	kg
electricity	20.11	kWh	wastewater	52.88	kg
			electronic scrap	138.48	kg

Table 2. Direct Equipment Costs for REE Recovery from HDD Shreds[31]

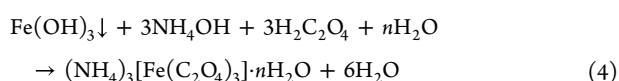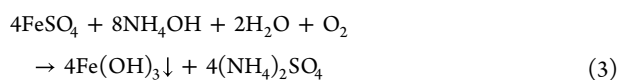| equipment	description	direct cost	capacity
bag filter 1	stainless steel multi bag filter	$630.00	20 t/h
bag filter 2	bag filter 100 L capacity	$630.00	20 t/h
transfer pump	pump a	$3150.00	40 m³/h
in-line circulation pump	pump b	$3150.00	40 m³/h
metering pump	pump c	$3150.00	40 m³/h
reactor 1	plastic tank container	$1995.00	10,000 L
reactor 2	plastic tank container	$1995.00	10,000 L
reactor 3	plastic tank container	$1995.00	10,000 L
reactor 4	plastic tank container	$1995.00	10,000 L
reactor 6	plastic tank container	$1995.00	10,000 L
reactor 5	jacketed stainless steel reactor	$3990.00	30,000 L
dryer	100 L conical dryer	$23,100.00	2550 × 1000 × 2000 mm
waste treatment	EWTSDEMINERAL	$31,980.00	50 m³/h
contingency	contingency	$61,730.00	
oven	1500 L capacity	$126,000.00	2200 × 1000 × 1000 mm

The facility could recover 2.53 tonnes of REO annually from 342 tonnes of HDD shreds. Each recycling batch takes 37 h and yields 12 kg of REO. The first step of the recovery process is the dissolution of copper sulfate in water, which takes about 4 h. After this, the HDD shreds (1607 kg) are selectively leached in the copper salt aqueous solution, as shown in eq 1. Due to the leaching selectivity of the acid-free dissolution process, the HDD shreds are represented as $2RE_2Fe_{14}B$ in the chemical equations.

$$2RE_2Fe_{14}B + 34CuSO_4 + 18O_2$$
$$\rightarrow 2RE_2(SO_4)_3 + 28FeSO_4 + Cu_3(BO_3)_2\downarrow + 30CuO\downarrow$$
$$+ Cu^O\downarrow \quad (1)$$

The copper sulfate recovery step designed for this study was performed by using a wastewater treatment unit that converts copper oxide into copper salt.[27,28] Bag filter 1 was used to filter the solution, and all copper, aluminum, and 90% of the iron were transferred to the solid waste container. This solid stream maintains a value similar to that of the raw HDD shreds because it retains many of the precious metals, like gold, in the original HDD. The ability to recover these precious metals is not impacted by the dissolution process. The liquid exiting bag filter 1 was sent to reactor 4, where it was mixed for about 30 min with an aqueous solution of an oxalate anion containing precipitating agent coming from reactor 3 (eq 2). After this, the contents of reactor 4 were filtered using bag filter 2, and the liquid stream was pumped into the wastewater treatment container. The cake was fed into the dryer.

$$RE_2(SO_4)_3 + 3C_2O_4^{2-} + nH_2O$$
$$\rightarrow RE_2(C_2O_4)_3 \cdot nH_2O\downarrow + 3SO_4^{2-} \quad (2)$$
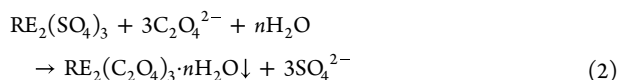
A purification stage was performed in reactor 5 to remove any remaining impurities, mainly iron. In this stage, water was added to the cake from bag filter 2, and after mixing into a suspension, ammonium hydroxide and water were added. The reactor was then heated (≤ 50 °C), and the contents were mixed (eq 3). After precipitation with a source of an oxalate ion, the products were discharged to reactor 5. The contents inside reactor 5 were heated (≤ 60 °C), mixed (eq 4), and filtered in bag filter 2. The liquid stream was sent to the wastewater treatment container, and the cake was transferred to reactor 5. The purification steps were then repeated.

$$4FeSO_4 + 8NH_4OH + 2H_2O + O_2$$
$$\rightarrow 4Fe(OH)_3\downarrow + 4(NH_4)_2SO_4 \quad (3)$$

$$Fe(OH)_3\downarrow + 3NH_4OH + 3H_2C_2O_4 + nH_2O$$
$$\rightarrow (NH_4)_3[Fe(C_2O_4)_3]\cdot nH_2O + 6H_2O \quad (4)$$

The wastewater was treated by adjusting the pH using sodium hydroxide (eq 6).[29]

$$H_2C_2O_4 + nH_2O + 2NaOH \rightarrow Na_2C_2O_4\cdot nH_2O + 2H_2O \quad (5)$$
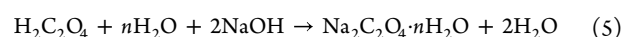
After the purification steps were performed, the liquid stream from bag filter 2 was also sent to the wastewater treatment. The cake was transferred to the dryer, where it was dried for 2 h at a temperature of 150 °C (eq 6).

$$RE_2(C_2O_4)_3\cdot nH_2O \rightarrow t^{\circ}C \rightarrow RE_2(C_2O_4)_3(H_2O)$$
$$+ (n-1)H_2O\uparrow \quad (6)$$
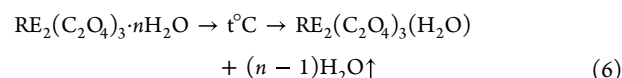
The last step in the rare-earth recovery process is calcination. The stream coming from the dryer was transferred to a calciner, where it is heated to 1000 °C for about 4 h (eq 7). As shown in Figure 1, about 1.0 kg of REO is recovered per batch.

$$2RE_2(C_2O_4)_3(H_2O) + 3O_2 \rightarrow 2RE_2O_3 + H_2O + 12CO_2\uparrow \quad (7)$$
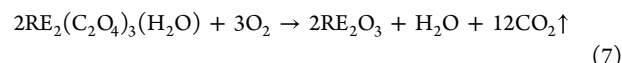
Energy Consumption. For calculating the energy consumption of the recovery process, the approach described by Chowdhury et al. was adopted.[28] For calculating heating energy (Q_{heat}), the energy to raise the temperature of the reaction mixture (Q_{temp}), the energy loss due to heat transfer on the reactor wall (Q_{loss}) and the efficiency of the heating element (η_{heat}) are considered. Equations 8−10 show how to compute the mentioned parameters, where $C_{p,t}$ is the specific heat capacity, which was assumed to be 4.12 (J/kg/K), m_{mix} refers to the mass of the reaction mixture (kg), T_r is the temperature of the target reaction, and T_0 is the ambient temperature (K). A denotes the surface area of the reactor, K_a is the thermal conductivity of the insulation material, s is the insulation thickness, and t is the reaction time. The efficiency of the heating element was assumed to be 95%.[28]

$$Q_{temp} = C_{p,t}m_{mix}(T_r - T_0) \quad (8)$$

$$Q_{loss} = A\frac{K_a}{s}(T_r - T_0)t \quad (9)$$

$$Q_{heat} = \frac{Q_{temp} + Q_{loss}}{\eta_{heat}} \quad (10)$$

To estimate the energy required, four mixing cylindrical reactors were considered for industry-scale operations based on the rotational speed of the lab-scale-sized equipment. Equations 11 and 12 were used to calculate the stirring power demand (E_{stir}) of the chemical mixtures. N denotes the rotational speed of the agitator, d refers to the diameter of the impeller, N_p is a dimensionless impeller power number based on the theory of similarity, ρ_{mix} is the density of the mixture, and η_{stir} is the stirring efficiency.[28]

$$N_{\text{industrial}} = N_{\text{lab}}(d_{\text{lab}}/d_{\text{industrial}})^{2/3} \tag{11}$$

$$E_{\text{stir}} = \frac{N_p \rho_{\text{mix}} N_p{}^3 d^5 t}{\eta_{\text{stir}}} \tag{12}$$

Furthermore, for approximating the energy required for industrial-scale filtration operation, an average value of 5.5 kWh/ton of dry material was used. To estimate the energy consumption to transport liquid from one reactor to another by the pumps (E_{pump}), the formula shown in eq 13 was used. g refers to the acceleration of gravity, Δh is the total hydraulic head, and η_{pump} is the efficiency of the pump. Finally, a roasting energy consumption of 16 kWh/kg of REO was assumed. This process is the thermal treatment that converts RE oxalate into REO and is the most energy-intensive process in the facility. This energy cost can be reduced in the future since the decomposition of REE oxalate to oxide can be accomplished at 800 °C.[30] Table S1 shows the parameters used to compute the energy requirement for this study, and Table S2 summarizes the energy consumption estimates.[28]

$$E_{\text{pump}} = \frac{m_{\text{mix}} g \Delta h}{\eta_{\text{pump}}} \tag{13}$$

Techno-Economic Analysis. The techno-economic analysis (TEA) was performed to analyze the economic performance of the recycling plant. The TEA is based on the Nth-plant assumption that the facility has solved all major technical and engineering challenges. The plant was assumed to be operating 7884 h/year. This TEA is based on a multiyear discounted cash flow rate of return analysis focused on estimating the minimum selling price (MSP) of didymium. The main utilities required for the recovery process included water and electricity.

The equipment used for the study and the purchase cost are listed in Table 2. The cost basis is 2022. The capacities for the used equipment are the following: 20 t/h for all the bag filters, 10 m³/h for the transfer and metering pump, 40 m³/h for the in-line circulation pump, and 10,000 L for the reactors (plastic tank containers) except for reactor 5 (jacketed stainless-steel reactor), the capacity of which was 30,000 L, 1000 L for the dryer, 4500 L for the waste treatment container, and 1500 L for the oven. The equipment cost assumed for this study is the following. A Lang factor of 2.1 was applied to each equipment cost to calculate the capital cost for the project. The equipment cost for bag filters 1 and 2 was determined to be $630. The pumps cost $3150. Reactor 5 was assumed to cost $1995, and the rest of the reactors $1995. The dryer cost is $23,100. The wastewater treatment unit cost is $31,980, and the oven price is estimated to be $126,000. The total capital cost was $267,500, and a contingency of 30% was added to cover unexpected direct costs. More detailed information is provided in Table S3.[21−29]

Operating costs include raw materials, utilities, labor, maintenance, waste management, and other recurring expenses. We assumed the cost of e-waste to be zero for this study. HDD shreds are commonly available in IT assets disposition, HDD disposal, and recycling facilities. Companies are often willing to pay to ensure complete and secure disposal of HDDs due to privacy and security concerns. As HDD shred recycling develops, the cost of e-waste could increase. Labor rates are based on the U.S. Bureau of Labor Statistics 2020 rates for Iowa.[32] The cost of copper sulfate is assumed to be $3.00/kg. The precipitating agent price is $2.72/kg. The price of ammonium hydroxide is $5.50/kg, and the sodium hydroxide price is $1.50/kg. Moreover, the average industrial electricity price for the U.S. was considered to be $0.040/kWh.[33−37]

A discounted cash flow analysis was performed using Microsoft Excel to calculate the net present value and the minimum selling price. The following assumptions were made for computing this analysis: an internal rate of return of 10%, a project lifetime of 20 years, and a loan interest of 7.50% for 10 years. The income tax rate is 39%. The equipment depreciates at a double-discounted rate over a 7 year period.

Sensitivity Analysis. An in-depth analysis of all of the process input variables was performed to identify how many variations in these values would impact the MSP of didymium oxide. The sensitivity analysis parameters include electricity use (4.706 MWh), plant engineer salary ($65,000/year), NH$_4$OH price ($5.5/kg), precipitating agent price ($2.72/kg), general employee salary ($25,000), capital cost ($267,485), CuSO$_4$·5H$_2$O price ($3.0/kg), machine operator salary ($43,000), CuSO$_4$ recovery (70%), operating hours (7884 h), REO recovery (0.75%), and REE content in HDD (0.67%). Each sensitivity analysis parameter was varied by ±20%.

Life Cycle Analysis. LCA was performed to quantify and assess the potential environmental impact and resources used for the recovery process. LCA is a sustainability assessment method defined by the ISO 14040 and ISO 14044 standards.[38] In this study, we define the life cycle system boundary as the boundaries of the e-waste facility. The functional unit is 1 kg of REO.

We have considered acidification, ecotoxicity, eutrophication, global warming, ozone depletion, respiratory effects, and non-carcinogenic impact categories. Acidification evaluates the potential acidification of water and soil caused by the emissions of pollutants such as nitrogen and sulfur oxides. Ecotoxicity indicates the impact of toxic substances released into the environment on freshwater, marine, and terrestrial organisms. Eutrophication refers to the enrichment of water and soil by nutrients due to human discharge of nitrogen and phosphorus. Potential global warming is measured by greenhouse gas emissions into the air. Ozone depletion indicates that air emissions contribute to the depletion of the stratospheric ozone layer. The potential respiratory effects caused by the organic substances were also evaluated. Carcinogenic reflects human toxicity contributors from chemicals released into the environment with cancer potency. Noncarcinogenic is an indicator of human toxicity contributors without cancer potency.[39−41]

This assessment was conducted in OpenLCA by using the EcoInvent database with the TRACI impact assessment method and various impact allocation methods. For the economic allocation method, we assume that the output iron salt byproduct is being sold for $1.20 per kg. This allocation method is based on the economic value of the product flows. For this analysis, it was assumed that all output materials other than REO and iron salt are considered waste for the output of the LCA. Table S4 and Table S5 show the allocation factors for the economic and physical allocation method without the selling, as well as with the selling of electronic scrap, and Table S6 summarizes the life cycle inventory with the cost and revenues used for the LCA. The causal allocation method results and the economic allocation method results are both shown in the Life Cycle Analysis section.[42]

Monte Carlo Simulation. Monte Carlo Simulation is also referred to as a multiple-probability simulation, and it is the most frequent model for approaching uncertainty in LCA. It is used to comprehend the effects of risk and uncertainty by modeling the probability of various outcomes in processes where the involvement of random variables makes prediction difficult. This kind of simulation uses random samples from predetermined statistical distributions of each input parameter for calculating the target output. Moreover, this process is repeated many times to obtain a large sample of model outputs. Finally, an approximate representation of the output distribution can be made using the sample's frequency distribution.[43,44] Monte Carlo simulation was performed using OpenLCA. One thousand trials were run, and the TRACI impact assessment and causal allocation method were used in the simulation.

■ RESULTS AND DISCUSSION

Techno-Economic Analysis Results. The estimated MSP for didymium oxide is $130/kg. Figure 2 shows the operating costs per kilogram of REO recovered. Utility costs are the largest expense, representing 31% of the costs, and they include both heating and power consumption costs. CuSO$_4$ is the second largest cost (11%) followed by the precipitating agent
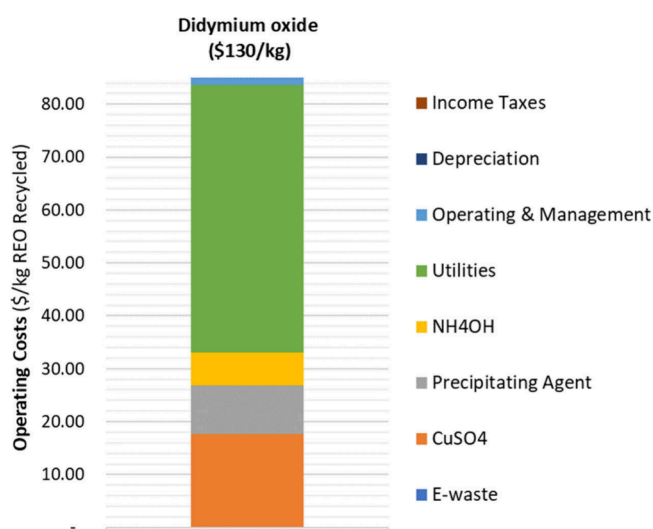
Figure 2. Operating costs per kilogram of didymium oxide recovered from HDD shreds (e-waste).

cost. Operation and management costs are negligible because most units require minimal supervision and maintenance.

The copper sulfate recovery step modeled for this study has reduced the operating cost per kilogram of didymium oxide recovered by $38.77/kg. Assuming the price of 1 kg of CuSO$_4$ is $3 and 70% of it is recycled, the amount paid for copper sulfate would be reduced from $56.45/kg to $17.68/kg. The obtained minimum selling price for didymium oxide was 130 $/kg. As the market price in 2021 was around 150 $/kg of didymium oxide, this recovery process is a favorable and economically profitable option.[45]

Figure 3 shows the sensitivity analysis results. The variables that have a higher impact on the MSP are the content of
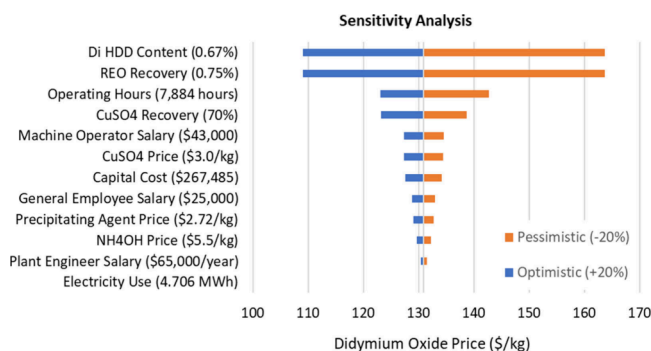

Figure 3. Sensitivity analysis for the REO recovery process from HDD shreds.

didymium in the HDD and the REO recovery percentage. A 20% increase in didymium HDD content (and REO recovery) results in a decrease in the MSP to $109/kg. Moreover, by optimizing the CuSO$_4$ and REO recovery (with a 20% increase) the MSP could be reduced to around $102/kg. Moreover, by optimizing all of the parameters analyzed in the sensitivity analysis, the MSP could be reduced to around $73/kg. Furthermore, the capital costs are not a major driver of the MSP. This is because most of the equipment consists of simple, inexpensive tanks and only a couple of pieces of specialized equipment. However, capital costs are difficult to accurately estimate, and most TEAs have an uncertainty of

±30% or greater. Solvent recycling is a key parameter of the process. Solvent prices have a high uncertainty and could vary by more than 20% over short time periods. Furthermore, wastewater treatment costs could increase if solvents are not recovered efficiently. Thus, more research should be invested in improving solvent recovery or finding more sustainable alternatives.

Life Cycle Analysis. Figure 4 shows the percentage contribution to the environmental impact of the materials and processes involved in recovering 1 kg of REO. Different materials and processes drive the environmental impact categories considered in this study. Electricity is the primary source of greenhouse gas emissions, as shown in the global warming potential category, followed by heat and aluminum. Copper, oil and gas, and other material and energy resources contribute the most in other environmental impacts. This suggests that improving the sustainability of e-waste recovery involves a comprehensive strategy encompassing all material and energy inputs.

In this study, we investigated the recycling of copper sulfate to reduce production costs, and this results in environmental benefits. The designed copper recovery step decreased the consumption of copper sulfate per kilogram of REO recovered by ∼70%, reducing the environmental impact of copper sulfate.

This e-waste recovery process avoids ammonium hydroxide environmental impacts. Ammonium hydroxide is consumed in this process, avoiding the release of nitrogen to water and soil. The business-as-usual case for ammonium hydroxide is to be used as a fertilizer. Up to 85% of the ammonia production is used to produce chemical fertilizers. The use of fertilizers increases the amount of nitrogen and phosphorus in water and soil, causing an increase in the levels of acidification and eutrophication. Consuming ammonium hydroxide in our recovery process avoids acidification and eutrophication impacts compared to using this ammonium hydroxide for fertilizers.

Blasting for manufacturing is the major contributor to ecotoxicity; oil and gas production is the largest contributor to ozone depletion; and copper production contributes to the carcinogenics category. Furthermore, the electricity production required to power the facility is the largest contributor to global warming and respiratory effects. This occurs because most of the elements used for the recovery process are liquids and solids and they are used in ways that do not result in air emissions. The detailed values for the emissions involved in recovering 1 kg of REO using causal allocation are shown in Table S7.[46−48]

There are different allocation methods used to help evaluate environmental performance improvement measures and normally provide very different results. LCA results of the studied process using causal and economic allocations are shown in Table 3. Economic allocation is sensitive to price changes and considers the market values of the output product flow, and for this study, two output flows were assigned as revenue sources which are REO and iron. Physical allocation calculates the environmental impact considering only the total mass and causal only considers what is going to happen to the output material.[49]

Figure 5 compares the environmental impact results of the acid-free dissolution process studied in this paper and those obtained in a previous study where other methods were used to recover REO from electronic waste. The data utilized in this comparison was obtained from the paper by Li et al.[50] In their
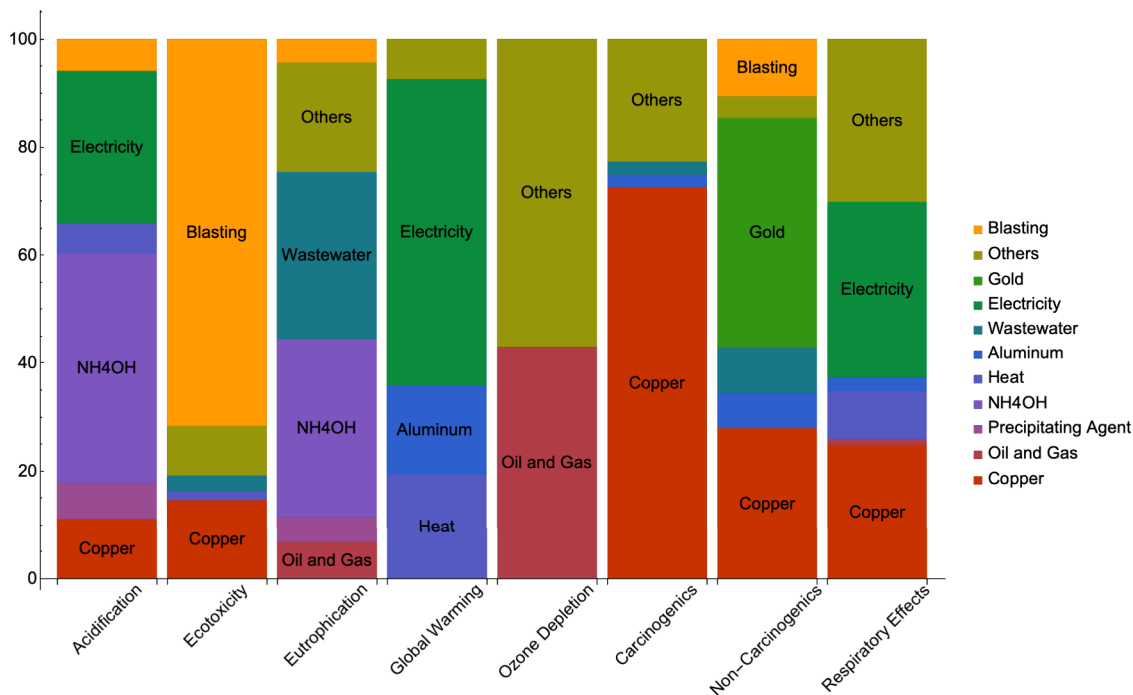
Figure 4. Environmental impact contribution of the input material and energy flows for processes required to recover 1 kg of REO from HDD shreds using causal allocation (impact assessment method: TRACI).

Table 3. Environmental Profiles for the Processes and Materials Needed to Recover 1 kg of REO from HDD (Impact Assessment Method: TRACI)

| impact category	unit	causal allocation	physical allocation	economic allocation
acidification	kg SO$_2$ eq	1.76×10^{00}	1.29×10^{00}	2.53×10^{02}
ecotoxicity	CTUh	1.26×10^{01}	9.23×10^{00}	1.81×10^{03}
eutrophication	kg N	1.67×10^{-03}	1.23×10^{-03}	2.40×10^{-01}
global warming	kg CO$_2$ eq	6.69×10^{00}	4.91×10^{00}	9.62×10^{02}
ozone depletion	kg CFC-11 eq	3.51×10^{-07}	2.58×10^{-07}	5.05×10^{-05}
carcinogenics	CTUh	1.24×10^{-01}	9.08×10^{-02}	1.78×10^{01}
non-carcinogenics	CTUh	2.57×10^{02}	1.89×10^{02}	3.69×10^{04}
respiratory effects	kg PM2.5 eq	1.57×10^{-02}	1.15×10^{-02}	2.26×10^{00}
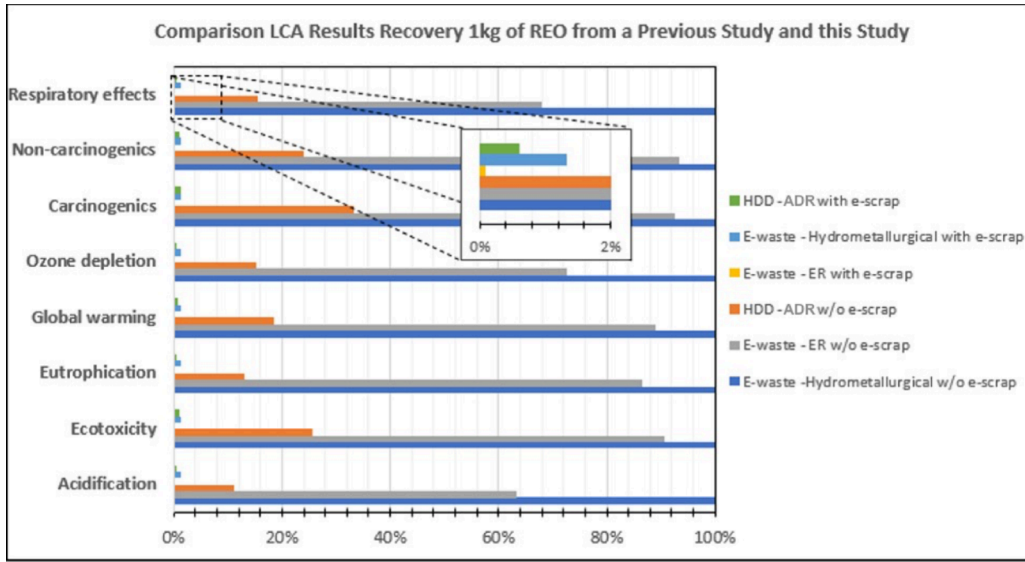
Figure 5. Environmental impact of recovering 1 kg of REO from electronic waste using acid-free dissolution, hydrometallurgical, and electrochemical recovery processes (impact assessment method, TRACI; allocation method, economic using the most recent REO prices).[37]

Table 4. Global Warming Potential Monte Carlo Simulation Results for Producing 1 kg of REO through Different Processes (Impact Assessment Method, TRACI; Allocation Method, Economic)

REO production route	mean	median	SD	CV	5% percentile	95% percentile	mean std. error
ADR	4.1×10^{01}	4.0×10^{01}	4.0×10^{00}	1.0×10^{-02}	3.6×10^{01}	4.8×10^{01}	1.0×10^{-02}
ADR (w/ scrap)	5.3×10^{00}	5.2×10^{00}	6.6×10^{-01}	1.0×10^{-02}	4.6×10^{00}	6.2×10^{00}	2.0×10^{-03}
hydro	6.0×10^{00}	5.9×10^{00}	5.0×10^{-01}	1.0×10^{-02}	5.3×10^{00}	6.9×10^{00}	1.6×10^{-03}
hydro (w/ scrap)	7.5×10^{01}	7.4×10^{01}	6.2×10^{00}	1.0×10^{-02}	6.6×10^{01}	8.6×10^{01}	2.0×10^{-02}
ER	3.4×10^{00}	3.4×10^{00}	1.5×10^{-01}	4.0×10^{-03}	3.2×10^{00}	3.6×10^{00}	5.0×10^{-04}
ER (w/ scrap)	5.3×10^{00}	5.3×10^{00}	5.2×10^{-01}	1.0×10^{-02}	4.7×10^{00}	6.2×10^{00}	1.7×10^{-03}

research, the authors conducted a comprehensive analysis of the life cycle and value recovery of precious metals and rare-earth elements from e-waste. The computed graph compares LCA results of the ADR recovery process, hydrometallurgical, and electrochemical recovery (ER) approaches for two different scenarios, one considering the electronic scrap from the output sold and the other without selling it. The price used in the scenario where electronic scrap is being sold was assumed to be $42.63 per kg for the three different recovery processes; this price was computed by multiplying the composition of each component of milled e-waste that appears in the paper by Li et al. by the price of each element of the composition. The prices used for each element are the following: $6.26 per kg of Cu scrap, $24.95 per kg of tin, $2.13 per kg of lead, $22.47 per kg of nickel, $0.57 per kg of steel, $2.78 per kg of zinc, $804.64 per kg of silver, and $63,687 per kg of gold.[37−42]

The values for the environmental impact results for the four scenarios are summarized in Table S8. The results showed the advantage of the recovery process in this paper for the scenario without selling electronic scrap (i.e., w/o e-scrap) and a competitive result of the analyzed recovery method for the scenario of selling electronic scrap (with e-scrap). To develop the comparison, the system boundary employed in the paper by Li et al. was modified. The preprocessing and the REE recovery steps were the only ones considered for the LCA results shown in Table S9 and Table S10. Some assumptions were made to develop the LCA analysis of the hydrometallurgical (hydro) and ER methods. The material balance provided in the study by Li et al. is for recovering 1 kg of gold. To convert this data into recovering 1 kg of REO, we have multiplied these values by 0.29 because 0.29 kg of gold was obtained per kg of REO recovered.

Monte Carlo Simulation Results. Table 4 summarizes the Monte Carlo simulation results of global warming potential obtained for the six routes for producing REO. From this table, we can conclude that the mean for the global warming emission potential for the proposed REO recovery from HDD would fall within 4.60−6.20 kg of CO_2 eq with 95% confidence. It can also be observed from the obtained results that e-waste recycling through electrometallurgical (electro) recovery has the lowest standard deviation value, meaning that the values obtained from the simulations are more closely grouped and therefore more precise than the result obtained for the other routes. Monte Carlo simulation results for the REO recovery process from HDD for other environmental categories are reported in Table S11, and Figures S2 and S10 show representations of these results on histograms. From the overall results, we can conclude that noncarcinogenics have the largest degree of uncertainty due to the uncertainty associated with their major contributors, which are gold and copper production. The results also indicate that the values obtained

for ozone depletion are the most precise of all the impact categories analyzed for this study due to the low standard deviation. The mean REO GWP increases with the sale of scrap because the economic ratio of REO to scrap is greater than the mass ratio. Thus, selling the scrap increases the REO environmental burden when an economic allocation is considered. Allocation methods should be considered carefully when there is insufficient market data to evaluate the market-based displacement impacts, which is the most common LCA approach.

◼ CONCLUSIONS

The growing demand for electrical and electronic equipment has led to increased generation of e-waste, which contains valuable REE. Scientists are seeking economic and sustainable methods to recover REE from e-waste. REE recovery can lower the dependence on mining, reduce environmental risks caused by landfill disposal of electronic waste, and stabilize global REE market prices.

This study investigated the costs and environmental impacts of REE recovery from HDD shreds by using an acid-free dissolution method. We estimated the capital investment, operating costs, and minimum selling price of the REE recovery. We also estimated the life cycle environmental impacts of REE recovery, including global warming potential, ecotoxicity, acidification, and other midpoint impact categories. We conducted sensitivity analysis of the economic results and Monte Carlo simulations of the environmental impacts to improve our understanding of the key system parameters. We compared the results to those of similar studies.

The copper sulfate recycled stream and the obtained minimum selling price shown in the TEA results demonstrated that the recovery process is profitable, that waste was avoided, and that the input material needed was reduced by recycling copper sulfate. The MSP of $130/kg is competitive with the current REO prices. As discussed in the sensitivity analysis section, further optimization of the recycling technology could potentially reduce the MSP to approximately $73/kg. The obtained ADR LCA results are comparable to the REO recycling routes using electrometallurgical and other hydrometallurgical methods. In general, the three recovery methods show an advantage if the output electronic scrap is being sold. Overall, the results obtained in this work for the conducted TEA and LCA demonstrate that the recovery of didymium oxide from HDD shreds is economically viable and can be conducted as a successful commercial operation.

Future work may study the valorization of other electronic waste byproducts from shredded HDD after the acid-free dissolution process, performing a TEA and sensitivity analysis for recovering copper(II) oxide to evaluate if recycling is economically beneficial rather than treating it as a waste and finding new pathways to recover REE from e-waste. Future

LCA could evaluate the sustainability of the ADR technology for REE recovery from other resources such as electric vehicles, turbines, solar panels, and other electronic devices.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.4c04100.

> Detailed information on material and energy flows, equipment costs, life cycle inventory data, and uncertainty analysis (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

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