



Full length article

Comparative life cycle analysis for value recovery of precious metals and rare earth elements from electronic waste

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ARTICLE INFO

Keywords:

Electronic waste

Electrochemical recovery

Life cycle assessment

Precious metal recovery

ABSTRACT

There is an ever-increasing concern regarding the electronic waste (e-waste), which is the fastest growing waste stream in the world. Incentivized by various legislations and the intrinsic value of critical metals inside, recycling of e-waste is becoming an attractive business opportunity that also benefits the environment. A novel electrochemical recovery (ER) process has been developed as a promising alternative to the existing pyrometallurgical and hydrometallurgical processes based technologies to recover base metals, precious metals, and rare earth elements (REEs) from e-waste. Experimental results indicate that the ER process has lower chemical consumption, enhanced control, and reduced energy demand compared to the pyrometallurgical and the hydrometallurgical processes. To quantify and compare the environmental performances of the three technologies, life cycle analysis has been conducted. Results show that the ER process outperforms the other two processes in almost all impact categories adopted in TRACI and ILCD while there is no clear winner between the hydrometallurgical and the pyrometallurgical processes. The highest impactful input for the ER method is hydrochloric acid, and for the pyrometallurgical method is copper scrap, while for the hydrometallurgical method, it is hydrogen peroxide, an oxidizer that accelerates base metal extraction process, that dominates the overall environmental footprint. The environmental viability of the ER process warrants the further development of ER process at industrial scale.

1. Introduction

The rapid technological developments in the information and communication technology (ICT) sector has allowed the consolidation of a highly competitive industry that fuels consumers to keep up with the latest available technology, but at the expense of a significant decrease in the lifespan of the electronic devices (İşildar et al., 2018). As a consequence, electronic waste (e-waste) has rapidly positioned as the fastest growing waste stream in the world (Awasthi et al., 2018). Around 2.44 million short tons of e-waste was produced per year. This scenario has generated widespread concern among researchers and legislators regarding the management and disposition of e-waste, due to the high toxicity of its components, such as halogenated flame retardants and heavy metals as well as the valuable metals inside (Jelea

et al., 2010; Perkins et al., 2014; Zhang et al., 2012a; 2012b). Common end-of-life options of e-waste are landfilling, incineration, refurbishment and recycling (Kiddee et al., 2013). Even though recycling is encouraged for the economic, environment, and human health benefits, as there are no U.S. regulations mandating to recycle electronic waste, and the portion of recycling e-waste in end processing is not high as desired (Namias, 2013), Kumar et al (2017) presented an overview about the statue of e-waste recycling in the U.S. and effort has been done to incent the recycle.

The recycling industry has seen a business opportunity in the surge of e-waste, mainly associated to the intrinsic value of the metals present in e-waste. Metals such as copper, tin, nickel, silver, gold, and palladium can be found in e-waste at concentrations that exceed those in minerals ores (Kumar et al., 2017). Moreover, process development has

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E-mail address: fzhao@purdue.edu (F. Zhao).<https://doi.org/10.1016/j.resconrec.2019.05.025>

Received 15 November 2018; Received in revised form 18 March 2019; Accepted 22 May 2019

Available online 28 May 2019

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made the mining of metals from e-waste more cost-effective than the extraction of minerals from natural sources (Zeng et al., 2018), with additional environmental benefits toward the stabilization of mineral supplies and the reduction of extractive mining. In brief, the processing technologies to reclaim metals from e-waste can be divided in two basic steps: pre-processing, which is based on physical transformations, includes dismantling, sorting, and shredding; and end-processing i.e. chemical transformation that allow the separation and recovery of the metals in different streams. Among these steps, the highest value generation can be attributed to the end-processing. Pyrometallurgical, hydrometallurgical, and combination of both processes dominate the literature and industrial implementation of e-waste end-processing (Kaya, 2016; Kumar et al., 2017).

Pyrometallurgical process is the most common process for recovering metal from e-waste and includes a high temperature furnace for melting. It has high reaction rate and the wastes are easily to be separated. Due to the emission regulations, pyrometallurgical process always requires larger scale facility. It also has issue of high energy requirement. The minimum viable smelting operation requires an annual throughput of 30 kt/year (Diaz et al., 2016) and the capital investment is around \$ 15 million (Diaz and Lister, 2018). Hydrometallurgical process involves leaching. It can be applied at small scale and has been considered cleaner and more economic than the pyrometallurgical process (Ghosh et al., 2015; Namias, 2013). However, despite that hydrometallurgical process is much less energy-intensive, its issues with long processing time and toxicity cannot be neglected. Electrochemical methods have been developed to reduce the use of reagents (Ghosh et al., 2015). Recently, electrochemical based processes are being developed as an alternative to existing pyrometallurgical and hydrometallurgical techniques, which can accomplish low chemical consumption, enhanced control, and reduced energy demand. In this study, the novel ER process using a weak oxidizer (Fe^{3+}) is chosen for the analysis which can be renewed electrochemically. The detailed process and presented in some previous works (Diaz et al., 2017; Lister et al., 2014). This process has lower capital investment than the pyrometallurgical route while use less chemicals than the hydrometallurgical route. Diaz et al. (2017) conducted a techno economic analysis of the ER process and black copper smelting routine for the recovery of metals from e-waste. A base scenario of 20 kt/year of cell phone material is considered for the simulation and the capital investment is around \$ 6.8 million (Diaz and Lister, 2018).

The environmental benefits of metal extraction from e-waste over primary sources, using either the pyrometallurgical or the hydrometallurgical processes, have been documented in multiple studies in recent years using life cycle assessment (LCA). Rodriguez-Garcia and Weil (2016) provided a statistical overview of LCA literatures related to the waste electrical and electronic equipment (WEEE) recycling in recent. Bigum et al. (2012) studied the environmental impacts of recovering metals from high-grade WEEE with pyrometallurgical processes via using literature review. In their study, preprocessing such as shredding and sorting of the WEEE was included in the system boundary, and the results showed e-waste recycling has significant environmental savings compared with mining and refining virgin metals. LCA of hydrometallurgical processes studied metal recovery from a variety of e-waste streams such as printed circuit board (PCB) (Rubin et al., 2014; Xue et al., 2015), physically treated residue of WEEE (Rocchetti et al., 2013), and mobile phones (Iannicelli-Zubiani et al., 2017). Rubin et al. (2014) compared the environmental impacts of recovering copper from PCB scrap using either sulfuric acid or aqua regia (nitric and chloridric acid). Xue et al. (2015) applied LCA methodology to assess a hydrometallurgical PCB recycling process that includes product collection, and the results highlighted the environmental benefits of metal recycling from e-wastes than primary metal production from virgin materials. Rocchetti et al. (2013) studied the environmental impacts of recovering metals including yttrium, cobalt, gold, and silver from physically treated residue of WEEE using

hydrometallurgical processes. The global warming potential of value recovery from different e-waste demonstrated a significant reduction of carbon footprint compared with primary metal production. Iannicelli-Zubiani et al. (2017) assessed the environmental impacts of recovering copper, silver, and gold from mobile phone in a hydrometallurgical pilot plant. Their LCA was conducted from cradle-to-gate and the results showed nitric acid leaching in gold recovery contributed most of the environmental impacts, which pointed out the critical steps for eco-design improvement. Hong et al. (2015) conducted the LCA of e-waste treatment in China and compared the environmental impacts of recycling with and without the end-life disposal. Their results showed e-waste recycling with end-life disposal can lower the environmental burden and is environmental beneficial.

Despite their contribution, the aforementioned studies did not directly compare the life cycle impacts of different e-waste processing technologies. Existing literature focused on either the pyrometallurgical or the hydrometallurgical methods (Iannicelli-Zubiani et al., 2017; Rocchetti et al., 2013; Rubin et al., 2014), or briefly stated the environmental impacts of disposal with primitive e-waste recycling operations: sorting with human labor and incineration (Kumar et al., 2017; Zhang et al., 2012a). Bailey (2016) conducted a comparative LCA for recycling neodymium magnets from e-waste with hydrometallurgical and pyrometallurgical techniques. His results showed there is no winner between the two techniques. However, this study was limited to recycling one type of particular element from e-waste. Therefore, there is a lack of LCA studies comparing the environmental impacts of different e-waste recycle technologies using consistent methodologies and boundary conditions, as well as reliable data for each process step in different technologies.

To fill these research gaps, this research investigated and compared the environmental impacts of recovering precious metals and rare earth elements from e-waste using three different recycling approaches. Life cycle impact assessment was carried out on the hydrometallurgical, pyrometallurgical and the novel comprehensive electrochemical recovery (ER) technologies following the ISO 14,040 standard. Precious metal recovery from e-waste was assessed, and the key contributors to the environmental impacts were identified.

2. LCA methodology

ISO standard LCA methodology is used for this study, which includes four steps: goal and scope definition, life cycle inventory analysis, life cycle impact assessment, and interpretation (ISO, 2006).

2.1. Goal and scope

This research was performed to evaluate the environmental impacts of different methods to recover precious metals from e-waste. The goals were to 1) quantify the environmental impacts of recovering precious metals from the ER, the hydrometallurgical, and the pyrometallurgical technologies and compare within the different methods, and 2) identify the key process or material in each method for e-waste recycling that has the most significant impact on the environment. The geographic region of e-waste recycling is assumed to be in the United States as the ER process is developed for application in the U.S. We chose a single country because the goal is to highlight the differences in processing technologies rather than confounding the impacts with reverse logistics. The final outputs include a variety of materials such as gold (Au), silver (Ag), copper (Cu) and rare earth elements (REEs) that are extracted at different stages of the recycling processes. The functional unit is defined as 1 kg of gold recovered from e-waste as gold contributes the most for the recovery revenue. Moreover, the recovery efficiency of gold are same for all three processes. Therefore, the life cycle impacts of recycling 1 kg of gold from e-waste are compared for different value recovery technologies. As there are exists other metals besides gold and they also have unneglectable value, additional scenarios have been

conducted for assessing the environmental impacts of metals recovery.

The system boundary of this LCA is grave-to-gate for three recycling routes: the ER process, the hydrometallurgical process, and the pyrometallurgical process. Detailed processes flows are provided in the following section. All processes start with the feedstock material of e-waste, which undergoes preprocessing and size reduction. The starting material volumes and compositions were assumed to be the same so that the three methods can be compared without bias. The LCA ends at the precious metals and REE materials extraction stage. In the alternative study, two of the scenarios separated the precious metal and REEs recovery.

2.2. Life cycle inventory

In this work, a comparative life cycle analysis among the traditional pyrometallurgical/ hydrometallurgical processes and an electrochemical based alternative has been performed. The material inputs, energy flow and emission data were gathered at Idaho National Laboratory based on the lab scale experiments on the electrochemical recovery. The corresponding data for the hydrometallurgical and the pyrometallurgical processes were based on available literature (Behnamfard et al., 2013; Ghodrat et al., 2016). It was assumed that the facility can process 10 ton/day of small IT e-waste using hydrometallurgical methods and 54.8 ton/ day using the other two methods (Diaz and Lister, 2018; Lister et al., 2016). A common pre-processing has been considered for the three alternatives so that the system boundary for the LCA was limited to the end-processing of the non-magnetic fraction of e-waste. A description of the processes alternatives is presented below.

2.2.1. Hydrometallurgical processes

Hydrometallurgical process is based on the oxidation of the metals using mineral acids, caustic leaching agents or suitable oxidants. The oxidation (extraction) of metals is performed in series of steps that allow the selective recovery of different value streams. Acid leaching is commonly employed for the removal of base metals (Cu, Ni, Sn, Zn, etc.), while stronger oxidants such as halides or complexing agents, such as cyanide, thiosulfates, and thiourea are used for the extraction of precious metals. After extraction, the respective leaching solutions could go through a purification/concentration step using either solvent extraction, adsorption or ion-exchange, or directly to metal recovery through chemical reduction or electro-refining (Cui and Zhang, 2008). The process diagram for a hydrometallurgical process option based on available literature (Behnamfard et al., 2013; Cheng et al., 2013) and described elsewhere (Lister et al., 2016) is presented in Fig. 1.

Based on the techno economic analysis presented elsewhere (Lister et al., 2016), the amount of major materials, energy inputs as well as emissions for all three extraction processes to obtain 1 kg of gold are

organized in tables for inventory analysis. Table 1 is the inventory for the hydrometallurgical process. In addition, Table S1 shows the breakdown of metals for this process, which can use to valid the mass balance for this process.

2.2.2. Pyrometallurgical processes

Pyrometallurgical processing for the reclamation of metals from waste streams has been a well-established technology for over two decades. Hagelüken et al. (2005) integrated lead and copper smelters that currently operate for the processing of e-waste in the Umicore's integrated metals smelter and refinery plant in Belgium. Cu smelters rather than lead smelters are more suitable for the processing of e-waste since in the lead smelters produce a copper matte product that still needs to be refined through black copper smelting (Khaliq et al., 2014). Additional advantages of Cu smelters over lead smelters include production of less harmful fumes and the alternative to recover precious metals using conventional electro-refining. For this LCA, a black Cu smelting (BCS) route has been selected as shown in Fig. 2. The process can be described in four consecutive steps: a reduction furnace where coal and/or the polymers present on the e-waste are used as reducing agent to obtain the crude black Cu, an oxidation furnace that allow the separation of metal impurities as oxide slag, the fire refining where natural gas is used to remove the oxygen in the molten Cu to produce Cu anode, and the electro refining processes, which include Cu and precious metals electro-refining. A more detailed description of the BCS process as well as techno economic analysis of the process applied to electronic waste are available in the referenced literature (Diaz and Lister, 2018; Ghodrat et al., 2016). Similar to the hydrometallurgical process, the inventory for the pyrometallurgical process is listed in Table 2.

2.2.3. Electrochemical recovery process

The ER process (Fig. 3) is based on the integration of the metals extraction and metal electrowinning steps, which are separated in a traditional hydrometallurgical process. This process integration allows a completely different extraction chemistry to be used for the extraction of base metals. In the ER process, Fe^{3+} is generated at the anode of a flow-through electrochemical cell (Eq. 1) from a leaching solution containing FeCl_2 and HCl . The leaching solution with the oxidant is fed to series of e-waste packed columns where the base metals (Cu, Sn, Zn, Ni, etc.) are oxidized (Eq. 2).

After leaving the extraction columns the leachate returns to the cathode side of the electrochemical cell, where the base metals are electrowon and recovered (Eq. 3). Ag from the e-waste is also oxidized but the low solubility of the AgCl maintains most of the oxidized silver within the columns. A series of at least three columns help to maintain the silver within the extraction system (due to galvanic reactions), and assure the complete reaction of the oxidant before it is returned to the

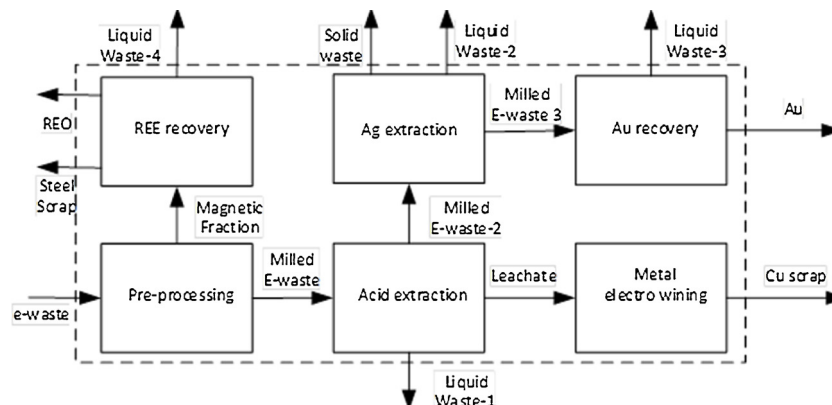


Fig. 1. System boundary of hydrometallurgical processing.

Table 1
Life cycle inventory to obtain 1 kg of gold for the hydrometallurgical process.

Input / Output	Consumption	Unit	Unit Process
<i>Inputs from technosphere</i>			
Electricity	2867.44	kWh	Electricity, medium voltage, at grid, 2015/US US-EI U
Hydrochloric acid	55.33	kg	Hydrochloric acid, 30% in H ₂ O, at plant/US- US-EI U
Hydrogen peroxide	10197.27	kg	Hydrogen peroxide, without water, in 50% solution state {GLO} market for APOS, U
Sodium thiosulfate	7.57	kg	Sodium persulfate {GLO} market for APOS, U
Sodium sulfate	11.86	kg	Sodium sulfate, anhydrite {RoW} market for APOS, U
Sulfuric acid	7439.01	kg	Sulphuric acid, liquid, at plant/US- US-EI U
Sodium hydroxide	2.89	kg	Sodium hydroxide, without water, in 50% solution state {GLO} market for APOS, U
Zinc	2.77	kg	Zinc {RoW} primary production from concentrate APOS, U
Sodium metabisulfite	1.25	kg	Sodium persulfate {GLO} market for APOS, U
Oxidant (FeCl ₃)	1.98	kg	Iron (III) chloride, 40% in H ₂ O, at plant/US* US-EI U
Calcium carbonate	6686.74	kg	Limestone, crushed, washed {RoW} market for limestone, crushed, washed APOS, U
<i>Direct emissions</i>			
Residue	229.25	kg	Solid waste
Water	52428.32	L	Waste water, to water

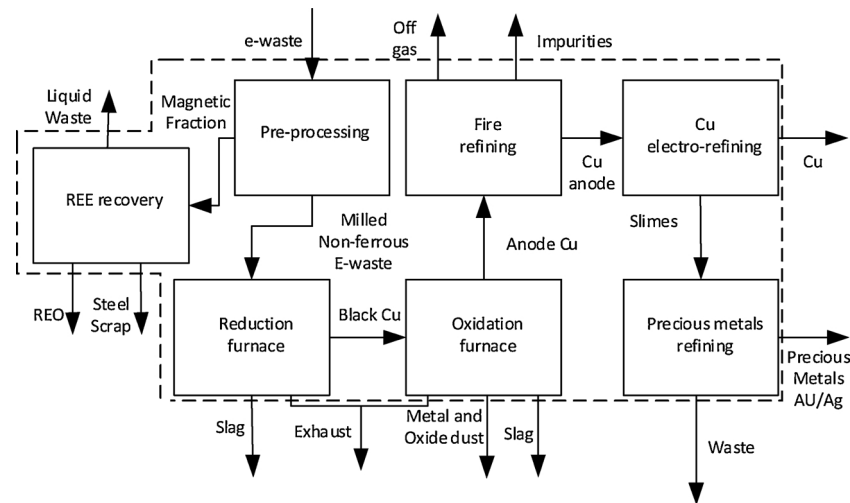


Fig. 2. System boundary of pyrometallurgical processing.

Table 2
Life cycle inventory to obtain 1 kg of gold for the pyrometallurgical process.

Input / Output	Consumption	Unit	Unit Process
<i>Inputs from nature</i>			
Water	1127.20	kg	Water, cooling, surface
Air	2762.85	kg	Air
<i>Inputs from technosphere</i>			
Electricity	5847.17	kWh	Electricity, medium voltage, at grid, 2015/US US-EI U
Copper scrap	1316.86	kg	Copper scrap, sorted, pressed {GLO} market for APOS, U
Coal	549.41	kg	Charcoal {GLO} market for APOS, U
Enriched air	3716.21	kg	Air and energy data from (Belaissaoui et al., 2014)
FCS slag	192.65	kg	Iron ore, 65% Fe, at beneficiation/GLO US-EI U
	71.97	kg	Activated silica {GLO} market for APOS, U
	84.41	kg	Calcium silicate, blocks and elements, production mix, at plant, density 1400 to 2000 kg/m ³ RER S
Natural gas	286.56	m ³	Natural gas, high pressure {RoW} market for APOS, U
Hydrochloric acid	37.35	kg	Hydrochloric acid, 30% in H ₂ O, at plant/US- US-EI U
Sodium hydroxide	2.89	kg	Sodium hydroxide, without water, in 50% solution state {GLO} market for APOS, U
Sodium sulfate	11.86	kg	Sodium sulfate, anhydrite {RoW} market for APOS, U
Electrolyte	2.54	kg	Sulphuric acid, liquid, at plant/US- US-EI U
Calcium carbonate	51.17	kg	Limestone, crushed, washed {RoW} market for limestone, crushed, washed APOS, U
<i>Direct emissions</i>			
Slag (Precious metal refining)	14.62	kg	Solid waste
Slag (Reduction furnace)	1438.0	kg	Solid waste
Slag (Oxidation furnace)	590.11	kg	Solid waste
CO ₂ release (Natural gas) ^a	8021.37	kg	Carbon dioxide
Exhaust gas	7276.70	kg	Exhaust to air

^a CO₂ release value calculation is based on reference (Vahidi et al., 2016).

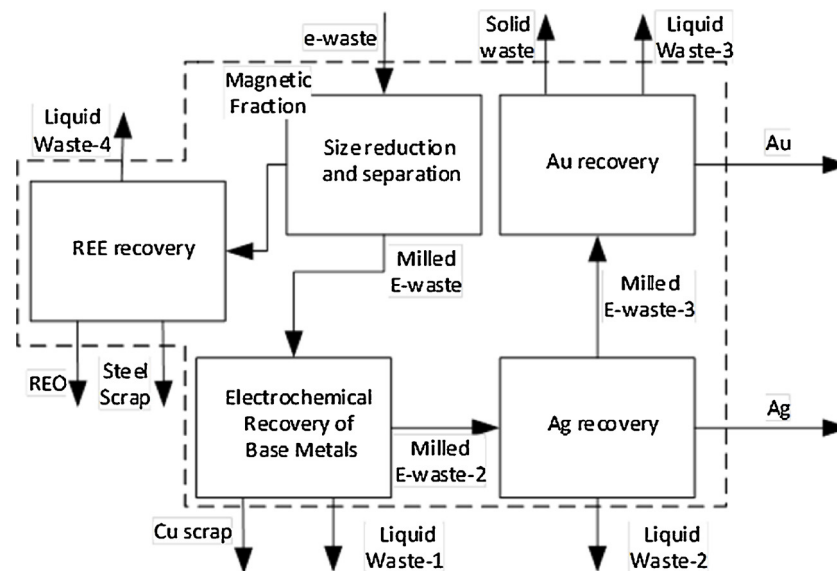
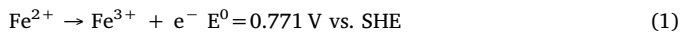


Fig. 3. System boundary of ER processing.

electrochemical cell. Spent e-waste material is removed from the extraction series and replaced with a new column to continue the cycle. Ag is then selectively recovered through complexation with Na_2SO_3 .



A significant reduction in chemical consumption can be achieved due to the regeneration of the oxidant used to extract the base metals, which represent over 90% of the total metal content in the e-waste. Precious metals are then selectively extracted and recovered as in the traditional hydrometallurgical processes. The novelty of this process is using the oxidizer which can be renewed electrochemically. Therefore, the chemical consumption can be reduced in the base metals extraction. Moreover, the extraction and metal recovery are performed in a single unit operation. Table 3 is the inventory for the ER process. In addition, Table S2 shows the breakdown of metals for the ER process for the mass balance checking.

Table 4 shows the recycling efficiencies per element and global based on the Eq. (4), it considered all inlets and outlets. In this case, the weight of inlets are same and it is the weight of the total e-waste. The column for weight is the original weight in 1 ton of e-waste. The recovery efficiency for gold is 97% for all three processes if compared the

Table 4

Recycling efficiencies per ton e-waste of different elements in three processes.

	Weight (kg)	ER	hydrometallurgical	pyrometallurgical
Cu	160.1	15.9%	15.9%	37.7%
Sn	16.6	1.7%	1.7%	0.1%
Pb	1.9	0.2%	0.2%	0.0%
Ni	20.4	1.5%	1.5%	0.8%
Fe	166.6	14.8%	14.8%	8.2%
Zn	12.4	0.1%	0.1%	0.0%
Ag	2.4	0.2%	0.2%	0.1%
Au	0.5	0.0%	0.0%	0.0%
Pr	0.2	0.0%	0.0%	0.0%
Nd	1.8	0.1%	0.1%	0.1%
Dy	0.2	0.0%	0.0%	0.0%
Total	383.0	0.345	0.345	0.470

extracted gold with the gold contained in the e-waste.

$$\eta = \frac{\sum m_{i,out}}{\sum m_{i,in}} \times 100\% \quad (4)$$

It should be mention that the efficiency is higher for Cu in the pyrometallurgical process because there is a significant input of copper scrap as feedstock to the process. Convert to the efficiency of metal recovery rate, the %Cu recovery for the pyrometallurgical process is around 240%. For the ER and hydrometallurgical processes, the Pb and

Table 3

Life cycle inventory to obtain 1 kg of gold for the ER process.

Input / Output	Consumption	Unit	Unit Process
Inputs from nature			
water	6964.40	kg	Water, cooling, surface
Inputs from technosphere			
Electricity	1131.97	kWh	Electricity, medium voltage, at grid, 2015/US US-EI U
Hydrochloric acid	343.08	kg	Hydrochloric acid, 30% in H_2O , at plant/US- US-EI U
Sodium hydroxide	2.89	kg	Sodium hydroxide, without water, in 50% solution state {GLO} market for APOS, U
Sodium thiosulfate	43.48	kg	Sodium persulfate {GLO} market for APOS, U
Sodium sulfate	11.86	kg	Sodium sulfate, anhydrite {RoW} market for APOS, U
Zinc	2.77	kg	Zinc {RoW} primary production from concentrate APOS, U
Sodium metabisulfite	1.19	kg	Sodium persulfate {GLO} market for APOS, U
Oxidant (FeCl_3)	0.20	kg	Iron (III) chloride, 40% in H_2O , at plant/US* US-EI U
Calcium carbonate	468.98	kg	Limestone, crushed, washed {RoW} market for limestone, crushed, washed APOS, U
Direct emissions			
Residue	229.25	kg	Solid waste
Water	7307.70	L	Waste water, to water

Sn are 100% recycled.

The LCA was carried out by using SimaPro 8.3. The Ecoinvent 3 database was used for most of the unit processes except for electricity, water, hydrochloric acid and sulfuric acid. Since the experiment was conducted in the United States, the datasets from the “US- US-EI U” are used to match the location. “Electricity, medium voltage, at grid, 2015/ US US-EI U” was used to consider the mix-produced electricity and processed water (RER S) was used as the water usage in the electrochemical recovery process. In the pyrometallurgical method, the enriched air contains 70% oxygen. Followed by one study of enriched air (Belaissaoui et al., 2014), here we assumed 1 kg of enriched air consumes 2.5 kg air and 0.15 kWh electricity. Direct emissions to environment in the three processes were roughly categorized as solid waste, air emission and waste water.

2.3. Impact assessment

To fully facilitate the use of results, TRACI (Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts) and ILCD (International Reference Life Cycle Data) methods were used to assess the environmental burdens. Both methods are widely used in LCA studies, they provide characterization factors for Life Cycle Impact Assessment (LCIA). TRACI is developed by United States Environmental Protection Agency (EPA) and it is widely used in the U.S. ILCD is developed by the Institute for Environment and Sustainability in the European Commission Joint Research Centre (JRC), in co-operation with the Environment DG which is widely used in Europe. To show more details in the impact assessment categories, here ILCD method for midpoint impact assessment was used.

2.4. Allocation

As the revenue generation is the driving force in the market, the first alternative study (scenario 1) is performed based on economic allocation (Santero and Hendry, 2016). The goal is to compare the environmental impacts on different outputs. We used allocation within all final products based on their revenue to analyze the environmental impacts of each metal. Table 5 shows the amount of gold, silver and some other products during the extraction. Values in percentage are ratios corresponding to share of values in the revenue stream. The amount of silver extracted in the pyrometallurgical process is larger than the other processes, while the ER method has the least silver extracted. In the LCA study, economic allocation is a good approach to evaluate the environmental impacts when the material values are different. Based on the value of different final products, the allocation factors are calculated based on the market price and the amount of elements.

Table 5
Weight and revenue of metal recovery.

Extract	hydrometallurgical		pyrometallurgical		ER		Unit price
	Weight (kg)	% Price	Weight (kg)	% Price	Weight (kg)	% Price	\$/kg
Gold (kg)	1	91.02%	1	75.53%	1	91.92%	41,043. ^a
Silver (kg)	5	5.85%	4.57	4.43%	3.94	4.65%	527.27 ^b
REE (kg)	3.54	0.52%	3.676	0.45%	3.67	0.54%	66 ^c
Steel (kg)	395.8	0.13%	407.11	0.11%	407.1	0.14%	0.15 ^d
Copper scrap (kg)	181.25	2.48%	–	–	198.7	2.75%	6.17 ^e
Copper (kg)	–	–	1462.1	19.47%	–	–	7.24 ^f
Total revenue							
Revenue (\$)	45,091		54,338		44,650		–

^a Gold prices was from citation (Gold Prices Today (2019)).

^b Silver prices was from citation (Silver Prices Today (2019)).

^c REE prices was from citation (Diaz and Lister, 2018).

^d Copper scrap prices was from citation (Today's Current Scrap Metal Prices (2019)).

^e Steel prices was from citation (“Steel - Rockaway Recycling,” n.d.).

^f Copper prices was from citation (1 Week Copper Prices and Copper Price Charts, 2019).

Other scenarios in this case study are focusing on the precious metals and REE recovery. In the scenario 2, we separated the precious metals recovery and REEs recovery processes and evaluated their impacts as the REEs recovery for each method are same. In this scenario, we assume the REEs are by-products. Accordingly, this case study examines the environmental impacts of precious metal recovery excluding the impacts of REE recovery. A special precaution should be taken during the pre-processing steps that are common for both precious metals and REE recovery. The energy consumed in the pre-processing step will be only included into the precious metals' recovery. In the scenario 3, the energy consumed in pre-processing would be allocated based on the value of metals and REE. The other processes are same as the scenario 2. These three scenarios are used to evaluate the environmental impacts of recovering gold and REE under difference considerations.

3. Results

3.1. Life cycle assessment: baseline

Table 6 and Table S3 show the comparative life cycle impacts of extracting 1 kg of gold with the ER method, the hydrometallurgical method and the pyrometallurgical method by using TRACI and ILCD in SimaPro 8.3 separately. Overall, the ER process (column ER of both Table 6 and Table S3) has lower environmental impacts than the hydrometallurgical process (column H of both Table 6 and Table S3) for all categories. Same conclusion obtained when compared to the pyrometallurgical process (column P of both Table 6 and Table S3). The ER process also has lower operational and capital cost compared to the other two processes.

Meanwhile, there is no clear winner between the hydrometallurgical method and the pyrometallurgical method in terms of environmental impacts. Fig. 4 set the highest value of three processes in each environmental category as the base for comparison and it clearly shows the overall environmental impacts for different processes. The hydrometallurgical process has a higher impact than the pyrometallurgical process in six out of ten categories in the TRACI method and 11 out of 16 in the ILCD method. Other than the environmental impacts, the hydrometallurgical method has lower capital cost and operational cost compared to the pyrometallurgical method.

From the comparison above, it is found the hydrometallurgical process has much impacts in the categories related to the acidification and human health, and the pyrometallurgical process is significant in the global warming. To identify the major source of the environmental impacts for different processes, the contribution of each component in all processes was analyzed. Fig. 5 shows the detailed LCA results for

Table 6
Overall results for the different processes per 1 kg of gold (TRACI).

Impact category	Unit	Hydrometallurgical(H)	Pyrometallurgical(P)	H/P	ER	ER/P
<i>Environment</i>						
Ozone depletion	kg CFC-11 eq	1.76E-03	4.38E-04	402%	3.48E-04	79%
Global warming	kg CO ₂ eq	1.79E+04	5.79E+04	31%	1.23E+03	2%
Smog	kg O ₃ eq	8.77E+02	4.42E+02	199%	5.90E+01	13%
Acidification	kg SO ₂ eq	1.68E+02	4.61E+01	364%	5.93E+00	13%
Eutrophication	kg N eq	5.18E+01	1.11E+02	47%	3.45E+00	3%
Carcinogenics	CTUh	3.85E-03	1.08E-03	356%	6.81E-05	6%
Non carcinogenics	CTUh	5.97E-03	2.91E-02	21%	4.92E-04	2%
Respiratory effects	kg PM2.5 eq	1.92E+01	8.11E+00	236%	4.72E-01	6%
Ecotoxicity	CTUe	2.38E+05	5.40E+05	44%	7.16E+03	1%
Fossil fuel depletion	MJ surplus	2.72E+04	8.00E+03	340%	1.35E+03	17%
<i>Cost^a</i>						
Operational	\$	1,030,957	1,474,021	70%	580,740	39%
Capital	\$	3,952	4,452	89%	1111	25%

^a Calculations are based on papers (Diaz and Lister, 2018; Lister et al., 2016).

individual inputs of the three processes with the TRACI method. 100% stacked columns histogram was used to compare the percentage of the component impacts in each process. The scale of the column cannot reflect the impacts between different methods. The inputs for three processes are not identity, thus some components only in specific process (i.e., H₂O₂ only shows in the hydrometallurgical process and copper scrap only uses in the pyrometallurgical process).

For most of the environmental categories, one or two components dominated the impacts. Moreover, some components dominated several categories, which provides the chance to significantly decrease the environmental impacts with only improving one sub-processing.

In the hydrometallurgical process, blue and dark yellow are the predominate color, corresponding respectively to two input materials 1) Hydrogen peroxide (H₂O₂) in the base metal extraction process with significant impacts on almost all environmental categories; 2) Sulfuric acid (H₂SO₄) which contributes around 60% of the total impact for the acidification and 30% for the respiratory effects. Refer the inventory table for this process, the large amount of the hydrogen peroxide can explain this result.

The noticeably major contributor for the pyrometallurgical process

are: (1) natural gas in the global warming category due to the carbon dioxide emission after combustion; (2) electricity in the ozone depletion and fossil fuel depletion categories; and (3) copper scrap added in the reduction furnace in rest of the categories, and 99% copper in the pyrometallurgical process indicates where the copper scrap goes.

For the ER process, the two input materials contributed significantly to the environmental impacts are hydrochloric acid (HCl) and electricity. Hydrochloric acid contributed much of the total impact in all categories. Especially for ozone depletion, it amounts to 90% of the total impact. Electricity in the ER process is the second largest contributor in all environmental categories with impact of up to 60%. Most of the hydrochloric acid in the ER process was used to extract REEs. Therefore, the impact of the hydrochloric acid will be significantly reduced if precious metal is the only recovery target.

3.2. Alternative case study

Based on the baseline study, LCA was carried out for three alternative scenarios to investigate the comparison between the three precious metal recovery methods under different treatments of REEs. Then

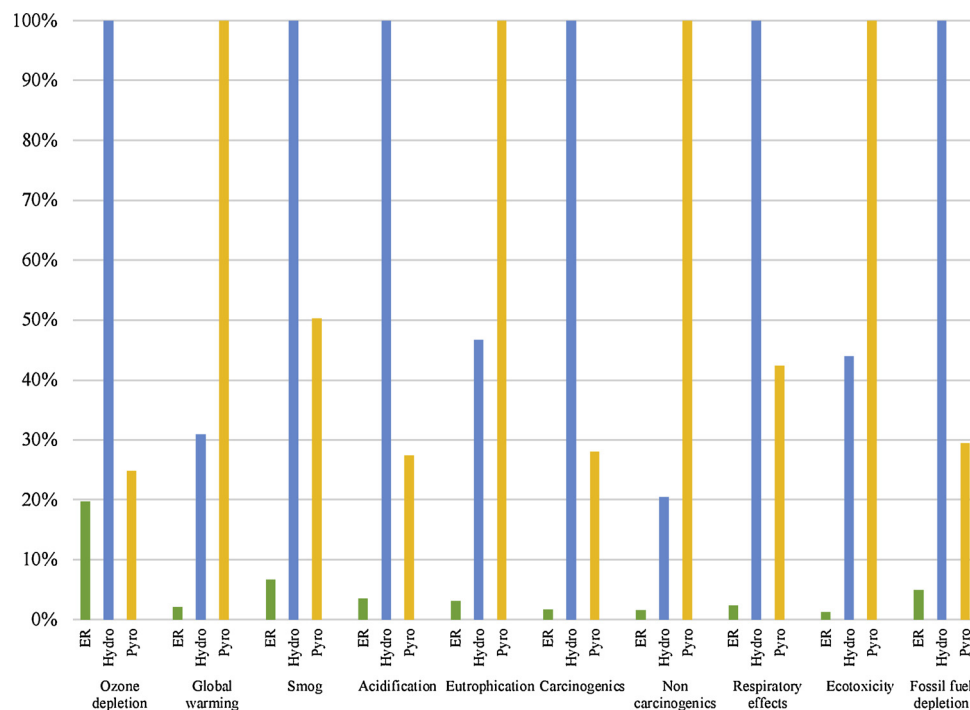


Fig. 4. Comparative life cycle impact of producing 1 kg gold.

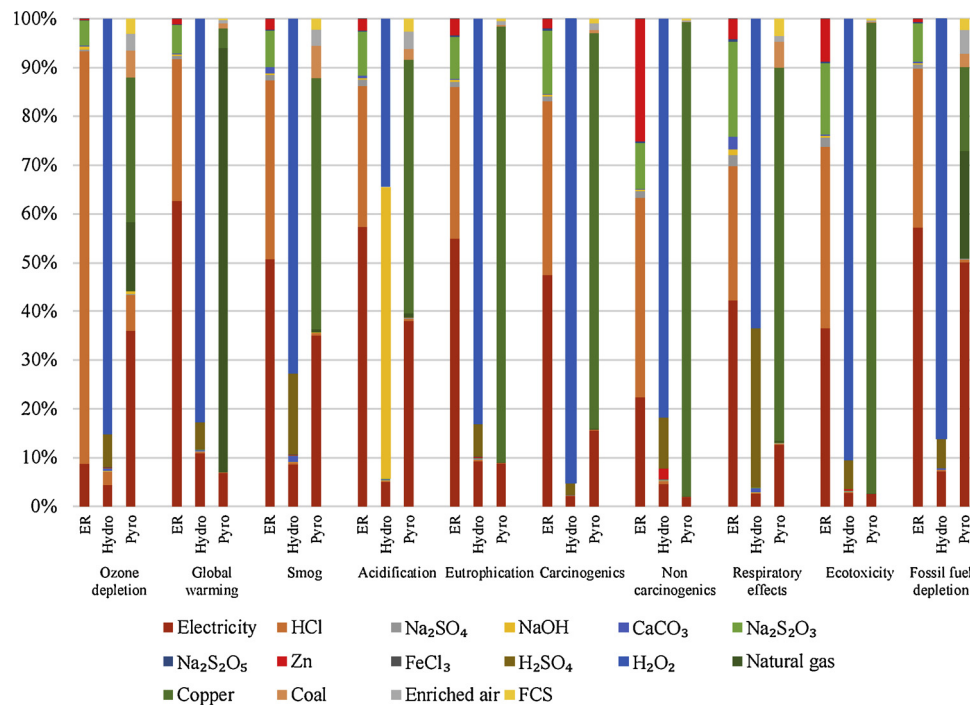


Fig. 5. Environmental impacts of each component in different processes for 1 kg of gold recovery.

the environmental impacts of best process in these alternative scenarios—recovery through the ER process—was compared with REE production through the virgin method.

Aforementioned, several scenarios with the same functional unit, 1 kg of gold recovered from e-waste, is studied. The initial feed-stock (i.e., e-waste) was divided into two groups of materials after magnetic separation: one that contains precious metals and the other that contains REEs. The following three alternative scenarios carried out the LCA using different treatment of the REEs by-product in gold recovery.

Alternative scenario 1 allocated all environmental impacts of processing precious metal and REEs using price of the materials. Scenario 2 separated the precious metal recovery and REE recovery processes, with all energy required in the pre-processing such as size reduction and magnetic fraction was attributed for the precious metal recovery. Scenario 3 also separated the recovery of precious metal and REE, while also allocating the electricity consumption during pre-processing using the price factors in Table 5.

Table 7 and Table S4 list the results for the LCA to produce 1 kg of gold with price allocation of all products (Scenario 1). Overall, the ER method was better than the other two approaches in term of environmental impacts. As the by-product of precious metal recovery, the life cycle impact of recovering REE was also investigated under scenario 1 as shown in Table 8 and Table S5 show the life cycle impact to product

1 kg REEs under the scenario 1. The values in the tables are converted to be based on 1 kg REE recovery. Since the amount of REEs recovered was similar for all three methods when obtaining 1 kg of gold, the ratio of environmental impacts between methods did not change significantly compared to the results based on 1 kg of gold recovery (Table 7 and Table S4). For the pyrometallurgical and ER methods, the amount of REEs were almost the same when obtaining 1 kg of gold. In contrast, the hydrometallurgical process resulted in less REEs, which explains why the H/P column in Table 8 is a slight lower than same column in Table 7.

Table 9 and Table S6 are the results for the LCA in Scenario 2 where 1 kg of gold was produced without considering the REE recovery process. Although all the energy consumption during the pre-process was counted in the precious metal recovery process, the total impacts were still lower than the results in scenario 1 without the REE recovery. The H/P was lower in scenario 2 than in scenario 1 as the fraction of revenue contributed from gold (75.53%) of the pyrometallurgical process was lower than in the hydrometallurgical process (91.02%) and ER (91.92%). In this scenario, the ER process had lower environmental impacts with the ozone depletion impact similar to the pyrometallurgical process and the other impacts much lower than the other two methods. There was still no obvious winner between the hydrometallurgical and pyrometallurgical methods in this scenario.

Table 10 and Table S7 show the life cycle impact to producing REE

Table 7
Comparative life cycle impact of producing 1 kg gold (Scenario 1, TRACI).

Impact category	Unit	Hydrometallurgical(H)	Pyrometallurgical(P)	H/P	ER	ER/P
Ozone depletion	kg CFC ₁₁ eq	1.60E-03	3.31E-04	484%	3.20E-04	97%
Global warming	kg CO ₂ eq	1.63E+04	4.37E+04	37%	1.13E+03	3%
Smog	kg O ₃ eq	7.98E+02	3.34E+02	239%	5.42E+01	16%
Acidification	kg SO ₂ eq	1.53E+02	3.48E+01	439%	5.45E+00	16%
Eutrophication	kg N eq	4.71E+01	8.38E+01	56%	3.17E+00	4%
Carcinogenics	CTUh	3.50E-03	8.16E-04	430%	6.26E-05	8%
Non carcinogenics	CTUh	5.43E-03	2.20E-02	25%	4.52E-04	2%
Respiratory effects	kg PM2.5 eq	1.75E+01	6.13E+00	285%	4.34E-01	7%
Ecotoxicity	CTUe	2.17E+05	4.08E+05	53%	6.58E+03	2%
Fossil fuel depletion	MJ surplus	2.48E+04	6.04E+03	410%	1.24E+03	21%

Table 8
Comparative life cycle impacts of producing 1 kg REE (Scenario 1, TRACI).

Impact category	Unit	Hydrometallurgical(H)	Pyrometallurgical(P)	H/P	ER	ER/P
Ozone depletion	kg CFC ₋₁₁ eq	2.59E-06	5.36E-07	482%	5.12E-07	95%
Global warming	kg CO ₂ eq	2.63E+01	7.09E+01	37%	1.81E+00	3%
Smog	kg O ₃ eq	1.29E+00	5.41E-01	238%	8.68E-02	16%
Acidification	kg SO ₂ eq	2.47E-01	5.64E-02	437%	8.73E-03	15%
Eutrophication	kg N eq	7.61E-02	1.36E-01	56%	5.08E-03	4%
Carcinogenics	CTUh	5.66E-06	1.32E-06	428%	1.00E-07	8%
Non carcinogenics	CTUh	8.77E-06	3.56E-05	25%	7.24E-07	2%
Respiratory effects	kg PM2.5 eq	2.82E-02	9.93E-03	284%	6.94E-04	7%
Ecotoxicity	CTUe	3.50E+02	6.61E+02	53%	1.05E+01	2%
Fossil fuel depletion	MJ surplus	4.00E+01	9.79E+00	408%	1.99E+00	20%

Table 9
Comparative life cycle impact of producing 1 kg gold (scenario 2, TRACI).

Impact category	Unit	Hydrometallurgical(H)	Pyrometallurgical(P)	H/P	ER	ER/P
Ozone depletion	kg CFC ₋₁₁ eq	1.72E-03	4.01E-04	429%	3.12E-04	78%
Global warming	kg CO ₂ eq	1.78E+04	5.77E+04	31%	1.15E+03	2%
Smog	kg O ₃ eq	8.68E+02	4.37E+02	199%	5.46E+01	12%
Acidification	kg SO ₂ eq	1.65E+02	4.57E+01	360%	5.51E+00	12%
Eutrophication	kg N eq	5.15E+01	1.11E+02	47%	3.21E+00	3%
Carcinogenics	CTUh	3.84E-03	1.08E-03	357%	6.33E-05	6%
Non carcinogenics	CTUh	5.92E-03	2.90E-02	20%	4.57E-04	2%
Respiratory effects	kg PM2.5 eq	1.89E+01	8.07E+00	235%	4.32E-01	5%
Ecotoxicity	CTUe	2.37E+05	5.40E+05	44%	6.60E+03	1%
Fossil fuel depletion	MJ surplus	2.70E+04	7.90E+03	342%	1.26E+03	16%

Table 10
Comparative life cycle impacts of producing 1 kg REE (Scenario 2, TRACI).

Impact category	Unit	Hydrometallurgical(H)	Pyrometallurgical(P)	H/P	ER	ER/P
Ozone depletion	kg CFC ₋₁₁ eq	1.13E-05	9.96E-06	113%	9.96E-06	100%
Global warming	kg CO ₂ eq	3.07E+01	2.17E+01	141%	2.17E+01	100%
Smog	kg O ₃ eq	2.43E+00	1.20E+00	203%	1.20E+00	100%
Acidification	kg SO ₂ eq	9.35E-01	1.13E-01	827%	1.13E-01	100%
Eutrophication	kg N eq	9.65E-02	6.57E-02	147%	6.57E-02	100%
Carcinogenics	CTUh	2.15E-06	1.31E-06	164%	1.31E-06	100%
Non carcinogenics	CTUh	1.49E-05	9.44E-06	158%	9.44E-06	100%
Respiratory effects	kg PM2.5 eq	6.20E-02	1.07E-02	579%	1.07E-02	100%
Ecotoxicity	CTUe	2.73E+02	1.51E+02	180%	1.51E+02	100%
Fossil fuel depletion	MJ surplus	3.93E+01	2.49E+01	158%	2.49E+01	100%

Table 11
Comparative life cycle impacts of producing 1 kg REE.

Impact category	Unit	ER 1 (Scenario 1)	ER 2 (Scenario 2)	REO from ion adsorption clay (Virgin)	ER 1/ Virgin	ER2/ Virgin
Ozone depletion	kg CFC ₋₁₁ eq	5.12E-07	9.96E-06	1.21E-05	4%	82%
Global warming	kg CO ₂ eq	1.81E+00	2.17E+01	2.26E+02	1%	10%
Smog	kg O ₃ eq	8.68E-02	1.20E+00	7.09E+00	1%	17%
Acidification	kg SO ₂ eq	8.73E-03	1.13E-01	1.51E+01	0%	1%
Eutrophication	kg N eq	5.08E-03	6.57E-02	6.23E+00	0%	1%
Carcinogenics	CTUh	1.00E-07	1.31E-06	4.91E-06	2%	27%
Non carcinogenics	CTUh	7.24E-07	9.44E-06	3.84E-05	2%	25%
Respiratory effects	kg PM2.5 eq	6.94E-04	1.07E-02	1.24E-01	1%	9%
Ecotoxicity	CTUe	1.05E+01	1.51E+02	5.82E+02	2%	26%
Fossil fuel depletion	MJ surplus	1.99E+00	2.49E+01	1.41E+02	1%	18%

as a separate process following the precious metal recovery in scenario 2. The energy consumption in pre-processing was not considered in evaluating the environmental impacts of REE production in this scenario. The values in the tables are converted to be based on 1 kg REEs. As the pyrometallurgical process and the ER process has the same input materials and similar amount REEs output, the environmental impacts of recovery 1 kg REEs in these two methods are same. The hydrometallurgical process had higher impact due to the use of sulfuric acid in REEs extraction, which led to high acidification and respiration effect

as shown in the H/P column in Table 10 and Table S7.

The scenario 3 allocated the electricity consumption during pre-processing between precious metal and REEs based on the price. The results for scenario 3 are not listed in this paper as they were very similar to scenario 2 as the total impact of pre-processing electricity consumption turned out to be relatively small. In scenario 3, the impacts of producing 1 kg of gold from all three processes were slightly lower than scenario 2 as some of the energy consumed is distributed to the REEs extraction.

The final analysis evaluated the life cycle impacts to produce 1 kg REEs with the ER method and the virgin method (Arshi et al., 2018). Since the ER method was identified to have the least environmental impacts when extracting gold from e-wastes, its results in scenario 1 and 2 were compared with the virgin method. The results are shown in Table 11.

Considering the REEs by-product of recycling gold, the impact of producing 1 kg REEs is ignorable compare to extract REEs from ion adsorption clay (Arshi et al., 2018; Diaz et al., 2016). If account the potential of recycling acid in the ER process, it is much more environmentally friendly than extracting REEs from ion adsorption clay.

However, if only recovery REEs by the ER process, even the impacts are lower than the virgin method for all categories, it is still higher than the impacts results in scenario 1. This shows only recover REE from e-waste is costly for the environment. It is necessary to include precious metals recycling into the REE recovery pathway for e-waste recycling to ensure the environmental sustainability of REE recycling.

Vahidi et al (2016) discussed and summarized the environmental profiles of several REEs studies which the REEs produced via the bastnasite/monazite route. For instance, the study of Sprecher et al. (2014) indicated the global warming impacts for 1 kg of REEs is in the range of 12–16 kg CO₂ eq and Ozone depletion is around 2 E-06–3.5E-06, and study of Zaimes et al. (2015) showed the respiratory effect of 1 kg REE production is around 0.16–0.18 kg PM_{2.5} eq. Compared the results for bastnasite/monazite route with the above alternative studies, recover REE from the ER process has lower environmental impacts.

4. Discussion

In this LCA study, the factories for processing and collecting e-waste were assumed at the U.S., thus the transportation of the materials are not included in the boundary. If consider the distance and methods for transportation, the LCA results may vary. In addition, the unit process ‘Electricity, medium voltage, at grid, 2015/US US-EI U’ is used. Even each process used electricity for the metal recovery, it has the largest impact on the ER process. In Fig. 5, the electricity (red) contributed much for the ER process, especially for the global warming and fossil fuel depletion categories. Alternative green energy source would make the ER process more attractive in terms of environmental impacts.

Since some components and compounds were not in the Simapro database, assumptions and user defined component were used in this study. Sodium persulfate (Na₂S₂O₈) was used in the place of sodium thiosulfate (Na₂S₂O₃) and sodium pyrosulfate (Na₂S₂O₅) in Simapro, as the latter two were not in the database. FSC slag used in the reduction furnace of pyrometallurgical process is a mixture of FeOx -CaO-SiO₂, and it was created in Simapro with given the ratio of the three existing components: Fe, activated silica and calcium silicate (Table 2).

To cut off original materials (like e-waste from cell phone) from the system boundary, some self-defined inputs and outputs were used. The emission of hydrometallurgical extraction process was mostly waste water with little solid waste produced. ‘Waste water’ in the Simapro databased was used for this water borne emission. This waste water can be further processed to reduce the environmental impact; however, the water treatment was not included in our system boundary.

In terms of chemicals, the oxidant for the ER process is recycled, which make it more efficient. The chemical consumption (hydrochloric acid, hydrogen peroxide and sulfuric acid) in both the ER and the hydrometallurgical process are the key factors with the greatest contribution to the environmental impacts. Refer the inventory tables, the lower overall environmental impact in the ER process compared with hydrometallurgical process can be attributed to its significant lower chemical consumption. Furthermore, acid used in the hydrometallurgical process makes it has 3 times more impacts than the pyrometallurgical and the ER process in the category of acidification. Compare the pyrometallurgical process with the hydrometallurgical process, the trend of results are similar as the study of Bailey (2016).

The pyrometallurgical process has the less impacts in more categories as it used less chemicals which are toxic and harmful for the human health. As for the categories which the pyrometallurgical process ranks the top, most of them were dominated by the input of copper scrap. Only the global warming impacts was due to the natural gas consumption. As mentioned in the recovery efficiency table, the Cu can be recovered more than 200% in the process. The added copper scrap was processed to higher level copper, and the revenue for the pyrometallurgical process is 20% more than the others. In the scenario 1, which used allocation to compare the impacts of recovering 1 kg of gold, the impacts of the pyrometallurgical process decreased more than the other two processes compared with the results in the baseline case. If the copper product can be reused as copper scrap, the overall environmental impacts will decrease dramatically. As for the ER process, even this study concluded it is environmentally than the other two methods, the current state is under the laboratory study and the applications of ER process still need to be researched.

5. Conclusion

This study provides a comparative LCA on recovering precious metals from e-waste with the hydrometallurgical, the pyrometallurgical and the ER processes. Simapro 8.3 was used for the inventory analysis with the database like Ecoinvent 3.0 and US- US-EI U. EPA TRACI (USA 2008) and ILCD were used to assess environmental impacts. Final products for recovery include gold, silver, REEs, steel and copper. Gold is the main product due to the high market price. Several scenarios were discussed with different system boundaries. Based on the baseline analysis and the alternative case study, which used 1 kg of gold as functional unit, it is found the ER process has the lowest environmental impacts compared to the other two processes, only except the higher ozone depletion impacts than the pyrometallurgical process. As for the pyrometallurgical and hydrometallurgical processes, they are competitive with each other in term of the environmental impacts. In addition, the LCA on recovering 1 kg REEs was conducted and the results show recovering REEs by the ER process has less environmental impacts than the in-situ extraction.

Acknowledgements

This work is supported by the Critical Materials Institute, an Energy Innovation Hub funded by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Advanced Manufacturing Office.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.resconrec.2019.05.025>.

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