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Applicability and environmental benefits of alternative resource recovery processes from end-of-life PV panels

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ABSTRACT: The feasibility of TU-Fe system to replace the conventional leaching using HNO3 followed by neutralization process was investigated with an attempt to evaluate the unviornmental burden/disburden using the stoichiometric information obtained from the experiments. Under the optimum conditions, the leaching capacity of TU-Fe system was nearly comparable (ca. 87%) to that of HNO_3, while the environmental impacts including GWP could be effectively reduced by employing various alternative leaching agents (i.e., TU-Fe and I_2-KI). The human health impact related to HNO_3 leaching was found to be highest because of using $Ca(OH)_2$ for neutralization and NO_x emission during the electrolysis. On the level of the ReCiPe midpoint method, the environmental impacts associated with HNO_3 leaching were found to be the highest as compared with other leaching agents with respect to the impact category of GWP, TAP, PMFP, MEP, and POFP, while those related to I_2-KI leaching were the highest in the case of ODP, TETP, and FDP. And the environmental impacts produced from thiourea leaching were the highest in MDP only. The uncertainty analysis of environmental impacts associated with the chemical treatment and material recovery process was performed using Monte-Carlo simulation. The impact variances from different leaching agents showed the meaningful statistical differences determined by ANOVA test (p-Value < 0.05).

Keywords: Alternative leaching, End-of-Life PV cell, LCA, Resource recovery, GWP

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Nomenclature or Abbreviation list

Ag   Silver

Al   Aluminium

c-Si   Crystalline silicon

EoL   End-of-Life

EU   European Union

EVA   ethylene vinyl acetate

FEP   Freshwater eutrophication

FETP   Freshwater ecotoxicity

FRELP   Full Recovery End of Life Photovoltaics

GHG   Greenhouse gas

GWP   Global warming potential

HTP   Human toxicity

IEA   International Energy Agency

IRENA   International Renewable Energy Agency

LCA   Life Cycle Assessment

LCIA   Life cycle impact analysis

METP   Marine ecotoxicity

MOTIE   Ministry of Trade, industry, and energy

ODP   Ozone depletion

OECD   Organization for Economic Co-operation and Development

POFP   Photochemical oxidant formation

PV   Photovoltaic

SRMs   Secondary raw materials

TAP   Terrestrial acidification

TETP   Terrestrial ecotoxicity

TU   Thiourea

WEEE   Waste electrical and electronic equipment

1. INTRODUCTION

Climate change occurring worldwide is mainly caused by an increase in carbon dioxide (CO_2) known as the major greenhouse gas (GHG) that has the greatest impact on global warming. Among several CO_2 emission sources, its share from the energy sector accounts for 58.8% of all GHG emissions worldwide (IEA, 2011). Therefore, in order to minimize carbon dioxide generation worldwide, efforts habe been made to use various renewable energy sources, e.g., solar energy, wind energy, biomass energy, geothermal energy etc. is encouraged instead of fossil fuels (Chowdhury et al., 2020). It is expected that they can provide 20-40% of the primary energy in 2050 (Fridleifsson, 2003). Electricity generated from fossil fuels accounts for CO_2 emission of 400 g to 1000 g CO_2 eq/kWh, while CO_2 emission of silicon-based solar panels can be ignored (Shin et al., 2017).

Among these renewable energy sources, photovoltaic (PV) cell directly converts solar energy to electricity without any emitting pollutants, noise, or vibration (Panwar et al., 2011). The solar cell market has grown repidly over the past 10 years, and by 2050, the global volume of waste crystalline silicon (c-Si) PV panel is estimated to be 9.57 million tons (Xu et al., 2018), considering the 15-25 years of lifespan with a possible early loss due to various reasons (e.g. natural disasters) (Chung et al., 2021). However, most end-of-life (EoL) PV modules around the world are sent to landfills. Heavy metals exist in c-Si PV panels such as lead and tin, so if the waste solar panel is not properly treated, it can leach or contaminate soil or groundwater, causing serious environmental pollution problem (Farrell et al., 2020; Xu et al., 2018).

Therefore, international standardization and protocol of disposal methods of c-Si waste PV panels are required though, most countries have not yet established nor adopted specific strategic frameworks and policies. European Union (EU) has established the regulatory frameworks based on the Waste Electrical and Electronic Equipment (WEEE) directive, including EoL PV waste management and recycling technologies for materials recovery. This directive has set rules to ensure that PV manufactures have imposed liability for the costs of collection, handling, and treatment while satisfying the requirements and responsibility stated in the WEEE directive (Sharma et al., 2019).

 "High-rate" recovery is to recovey high valuable-added materials such as precious metals, using leaching or chemical treatment like hydrometallury, electrolysis and chemical precipitation etc., and "low-rate" recovery is de-lamination and thermal treatment such as machanical separation or incineration. In order to recover valuable resources such as silver, silicon, and aluminum from c- Si PV cell, wet smelting (i.e., hydrometallurgy) and dry smelting (i.e., pyrometallurgy) can be applied, both of which produce significant amount of environmental burden; the former applies strong lixiviants such as nitric acid and sulfuric acid, generating toxic wastewater and greenhouse gas (GHG) such as nitrogen oxides which is known to affect global warming 300 times greater than CO_2 (Farrell et al., 2020; Olin et al., 2017) where the later requires a large amount of thermal energy that involves enormous amount of CO_2 generation from heating. On the other hand, biological extraction (i.e., bioleaching), draws attention as well owing to its eco-friendly perspective (Baniasadi et al., 2019; Johnson and Du Plessis, 2015; Petersen, 2010; Srichandan et al., 2019). However, bioleaching is very time-consuming and has a low yield, and even toxic chemicals can be produced during the process (Chen and Pan, 2010; Kumar and Yaashikaa, 2020; Valix, 2017). Recently, more attention is paid to the study of several environmental friendly (so clled "green") leaching processes, such as thiourea leaching, thiosulfuric acid leaching, and halide leaching (Zhang et al., 2012).

In order to minimize the environmental burden and maintain high efficiency in the process of recovering resources for proper management and processing of EoL PV panel, optimal conditions for leaching silver and aluminum were derived through experiments using thiourea instead of toxic lixiviant solutions. Per unit information obtained from the optimum leaching condition is then applied to the input data of LCA in the following section. Note that the stoichiometric information using I_2-KI solution were adopted from the previously published paper (Chung et al., 2021).

2. MATERIAL AND METHODS

2.1 Preparation of waste c-Si solar cell

Waste PV panel is obtained from a landslide site in Korea. For effective recovery of precious metals, it is necessary to separate solar cell from the rest of EoL PV modules. First, aluminum frame is disassembled manually, and then the cable and junction box are torn off from the PV sandwich. Electric furnace was heated for 2 hours at 600℃ to burn the plastic compounds including backsheet and EVA (Ethyl-Vinyl-Acetate) off. Then, the scraps of cells and broken glass particles were sieved (between #20 and #18) to obtain a uniform particle size between 0.84 and 1.00 mm. Finally, only PV cells are collected manually using tweezers to minimize the heterogeneity. The total mass composition is determined by ICP-OES (Varian, USA) after 70% HNO_3 digestion.

2.2 Leaching of c-Si PV cell using thiourea

Text Thiourea-ferric solution is prepared with analytical grade thiourea (CH_4N_2S) purchased from JUNSEI (Japan) and iron(Ⅲ) sulfate hydrate ($Fe_2O_{12}S_3 \cdot xH_2O$) purchased from Sigma-Aldrich (USA) dissolved with freshly prepared deionized (DI) water (> 18; MΩ cm; PALL Cascada, USA) before the experiments. All leaching experiments are carried out under room temperature (25℃) and 0.50 g of PV cell scrap is placed into 50 ml HDPE conical tube containing desired concentration of lixiviant solutions. The tubes were then shaken at 40 rpm for intended times (for kinetics, 1, 5, 10, 30, 60, 120 min, and 4 h for thermodynamic equilibrium) using rotary shaker (WiseMix RT-10, DAIHAN Scientific, Korea). Factors affecting the reaction including concentration of thiourea (TU) and ferric ion, solid to liquid (S:L) ratio, and shaking time were thoroughly examined. After the designated leaching time, the supernatants are filtered through syringe (HENKE-JET, Henke-Sass Wolf, UK) and 0.45 μm syringe filter (Whatman, USA). The dissolved metal concentrations are then analyzed by using ICP-OES (Varian, USA). The 1000 mg/L multi-element standard solution (MES-04-1, AccuStandard, USA) is diluted to prepare a calibration curve (1, 5, 10 and 50 mg/L) and different wavelengths not overlapped with each other are selected for Ag (328.068 nm) and Al (237.312 nm and 396.152 nm) based on ISO 22036.

2.3 Life cycle assessment on the alternative leaching process

The goal of this LCA study was to compare the potential environmental impacts related to three different solvents (i.e., HNO_3, iodine-iodide system (I_2-KI), and thiourea) for resource recovery from EoL c-Si solar cell scraps. The processing of the cell scrap collected from the EoL PV panels has been defined as a function, where the 2 kg (i.e., 1 kg per one cycle) of PV cell scraps for recovering precious metals (silver and aluminum) is defined as a functional unit. The system boundary in this chapter includes acid leaching, filtration, electrolysis, neutralization, and landfilling for final disposal. The environmental impact of processing 2 kg of PV cell scrap using HNO_3, I_2-KI, and thiourea solution was compared within the system boundaries (Fig. S1).

The input and output data for electrolysis, neutralization, and final disposal were adopted from Latunussa et al., (2016). The LCA studies were performed using OpenLCA (ver. 1.10.2) (Ciroth, 2007) with the Ecoinvent v3.5 database. The impact assessment was performed by means of the ReCiPe endpoint and midpoint methods provided in OpenLCA. In this study, the commonly used 11 midpoint indicators as reported in the previous studies (Corcelli et al., 2018) were investigated.
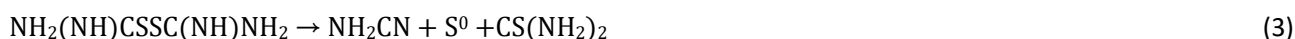
3. RESULTS AND DISCUSSION

3.1 Characterization of the EoL c-Si solar cell

Table S1 shows the composition of the solar cell used in this study. A total amount of tracer metals was extracted using 70% nitric acid. This is consistent with the previous study that used the same solar cell while more detailed preparation process including topological images can be found as well (Chung et al., 2021). Note that silver and aluminum contents were 5.973 mg/g and 75.605 mg/g, respectively, both of which are known to be the most valuable resources that can be extracted from EoL c-Si PV panel (Peeters et al., 2017).

3.2 Optimization of thiourea leaching

The Eh-pH diagram (i.e., Pourbaix diagram) has been often used to predict the stable phase of chemical species under specific pH and potential Eh, which can be useful in predicting the feasibility of leaching reaction. So et al., (2018) proposed the possible reaction pathways and stable form of various silver-thiourea compounds under broad range of Eh and pH. Thiourea forms cationic complexes with silver according to the following dissolution-complexation reactions (eq. (1)) (Gašpar et al., 1994). However, thiourea can be oxidized in an acidic solution to produce a primary decomposition product called formamidine disulfide ($NH_2(NH)CSSC(NH)NH_2$) as shown in Equation (2), which can reduce the overall performance via equation (3) (Calla-Choque and Nava-Alonso, 2020).

$$Ag + 3CS(NH_2)_2 = Ag[CS(NH_2)_2]_3 + e^- \tag{1}$$

$$2CS(NH_2)_2 = NH_2(NH)CSSC(NH)NH_2 + 2H^+ + 2e^- \tag{2}$$

$$NH_2(NH)CSSC(NH)NH_2 \rightarrow NH_2CN + S^0 + CS(NH_2)_2 \tag{3}$$

The stable, dissolved form of the cationic silver-thiourea complexes were found in the acidic region with an appropriate Eh value of 0-300 mV (as indicated by the shaded region in Fig. 1). Note that the pH and Eh conditions in the following series of leaching experiment results also fell within this range, indicating tha the stable TU-Ag complexes is formed in leachate.
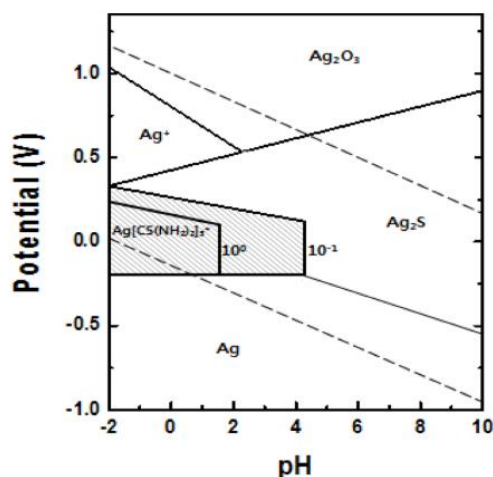

Figure 1. Pourbaix diagram of TU-Ag-H_2O system at 25 ℃, 1 atm (modified from So et al., 2018).

3.2.1 Effect of thiourea and ferric ion concentration on thiourea leaching system

Fig. 2 (a). shows the effect of thiourea concentration on the leaching of silver and aluminum, repectively. It is apparent from this figure that the leaching of silver increases with increasing TU concentration up to 0.3 M. The effect of TU concentration on the additional silver dissolution is not so efficacious when it is higher than 0.5 M, rather, it has an adverse effect in both cases beyond the optimal points, which might be attributed to the production of the formamidine compinds (So et al., 2018). In

addition, the additive Fe^{3+} concentration was also optimzed at 0.7 M under this condition (Fig. S2). Therefore, 0.5 M and 0.7 M are chosen as an optimum point which will be further used in the following LCA section.

Considering the aforementioned metals contents (5.973 mg /g and 75.605 mg/g for silver and aluminium, respectively), Ag leached out more than 90% whereas the leached Al freaction was only ca. 1%. This is presumably due to the different affinities of TU-metal complexations under the dwsignated pH-Eh, which could be beneficial to the selective recovery.

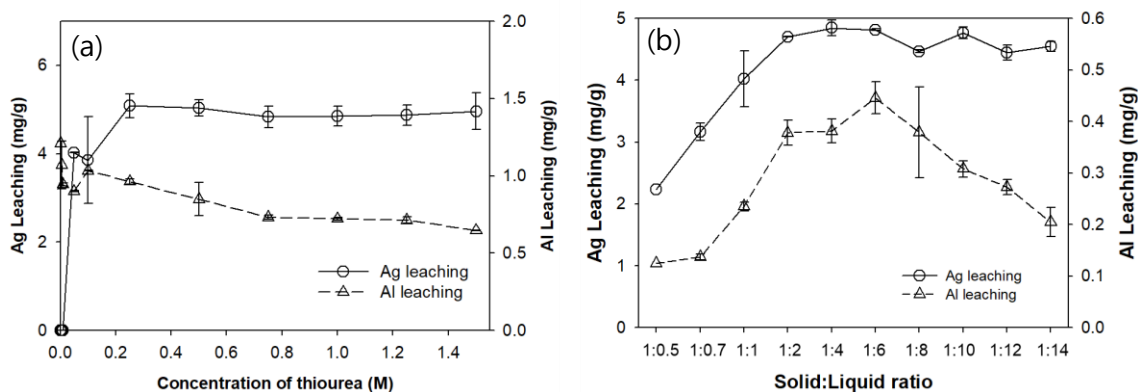### 3.2.2 Effect of S:L on thiourea leaching system

Figure 2. (a) Effect of thiourea concentration on the leaching amount of Ag and Al from 0.5 g of PV cell in TU-Fe system (initial concentration of Fe^{3+} 0.07 M, S:L 0.5 g:5 ml), (b) Effect of S:L on the equilibrium leaching amount of Ag and Al from 1 g of solar cell in TU system (initial concentration of thiourea and Fe^{3+} are 1.00 M and 0.07 M, respectively)

S:L ratio is another important parameters for batch type reactor that controls the mass transfer between reactants (Chen et al., 2015). Fig. 2b shows the changes in Ag and Al leaching from solar cell in different S:L ratio. In the case of Ag, a rapid increase in leaching amount is observed between 1:0.5 and 1:2, followed by gradual increase. On the other hand, that of Al also increased up to 1:6, and then rapidly dropped presumably due to the lowered contact opportunity from the passivation of surface with an excessive amount of applied lixiviant (Xiu et al., 2015).

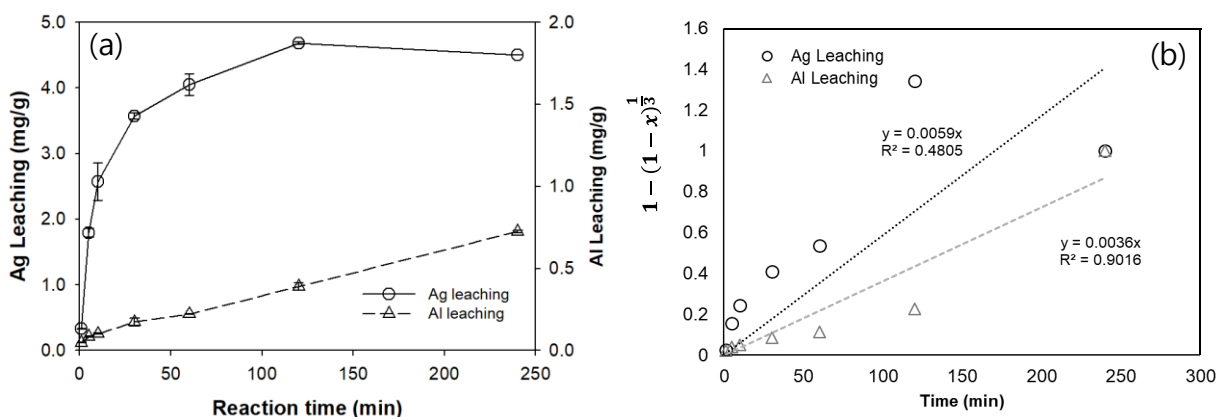### 3.2.3 Effect of reaction time on thiourea leaching system

Fig. 2 Effect of reaction time on the leaching kinetics of Ag and Al from 0.5 g of PV cell in TU- Fe^{3+} system (initial TU, Fe^{3+} and S:L ratio are 1.00 M, 0.07 M, and 0.5 g: 5ml, respectively)

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Results from the kinetic experiment is important for handling of large amount of PVwaste and often provides an insight to reaction mechanisms. Fig. 3 shows the leaching kinetics of Ag and Al within 240 min of the reaction time. Ag showed a dramatic reaction rate within the first 10 minutes, and then followed by a gradual increase up to 120 minutes, whereas Al is leached at a comparatively constant rate for 240 minutes of reaction time. The kinetic data were then fitted to the popular leahing model, shrinking core model (SCM) to determine which process controlls the leaching.

$$kt = 1 - (1 - x)^{\frac{1}{3}} \tag{4}$$

$$kt = 1 - \frac{2}{3}x - (1 - x)^{\frac{2}{3}} \tag{5}$$

Where x [-] is the leached fraction, k [T−1] is the leaching rate constant, and t (T) is the time. If the kinetic data is fitted to eq (4), chemcal reaction controlls the overall leaching, while if eq (5) fits the data better, the process is knwon to be controlled by diffusion (Othusitse and Muzenda, 2015). Ag leaching in this study is observed to be controlled by reaction with much faster rate (k = 0.011 min^{-1}; R^2 = 0.95) than Al (k = 0.002 min^{-1}; R^2 = 0.89).

3.3 Evaluation of life cycle assessment of c-Si PV cell leaching: HNO_3 vs iodine vs thiourea

The life cycle inventory data for each process and material have been derived from the Ecoinvent database. The input and output data for the LCA are presented in Table 1. Unlike the HNO_3 leaching method, the use of I_2-KI and thiourea does not require the neutralization process. In this chapter, it was assumed that the amount of Si scrap to be recovered was identical for all three different leaching agents. The amount of water supplied in the leaching process using HNO_3 and thiourea and I_2-KI is 20 kg and 10 kg, respectively. The only certain amount of iodine (I_2) was additionally supplied in the second cycle of the continuous process without supplying an additional water and potassium iodide (KI) input. Since the inventory of KI was not available in the Ecoinvent database, the inventory of potassium chloride was used in this study. In this study, the amount of lixiviants and the recovered silver and aluminum were adopted from the experiment and published paper (Chung et al., 2021).

Table 1. Input and output data of chemical treatment for c-Si PV cell scrap using different leaching agents.

	HNO$_3$ Leaching		I$_2$-KI Leaching		Thiourea Leaching	
	Amount	**Uncenrtainty**	**Amount**	**Uncenrtainty**	**Amount**	**Uncenrtainty**
Input						
PV cell[**]	2.000 kg	Lognormal dist. (σ_g, 1.0)	2.000 kg	Lognormal dist. (σ_g, 1.0)	2.000 kg	Lognormal dist. (σ_g, 1.0)
HNO$_3$[**]	2.520 kg[**]	Lognormal dist. (σ_g, 1.0)	-	-	-	-
I$_2$	-	-	1.620 kg[**]	Lognormal dist. (σ_g, 1.0)	-	-
KI	-	-	1.330 kg[**]	Lognormal dist. (σ_g, 1.0)	-	-
Thiourea	-	-	-	-	0.761 kg[***]	Lognormal dist. (σ_g, 1.0)
Fe$_2$(SO$_4$)$_3$	-	-	-	-	2.799 kg[***]	Lognormal dist. (σ_g, 1.0)
Electricity	2.580 kwh[*]	Lognormal dist. (σ_g, 1.2)	2.580 kwh[*]	Lognormal dist. (σ_g, 1.29)	2.580 Kwh[*]	Lognormal dist. (σ_g, 1.29)
Ca(OH)$_2$	12.992 kg[*]	Lognormal dist. (σ_g, 1.2)	-	-	-	-
Water	32.992 kg[*]	Lognormal dist. (σ_g, 1.2)	15.04 kg[*]	Lognormal dist. (σ_g, 1.29)	24.84 kg[*]	Lognormal dist. (σ_g, 1.29)
Output						
Limestone sludge	45.406 kg[*]	Lognormal dist. (σ_g, 1.2)	-	-	-	-
Inert sludge	-	-	12.504 kg[#]	Lognormal dist. (σ_g, 1.56)	19.905 kg[#]	Lognormal dist. (σ_g, 1.56)
Metal sludge	2.656 kg[*]	Lognormal dist. (σ_g, 1.2)	3.188 kg[*]	Lognormal dist. (σ_g, 1.29)	4.068 kg[*]	Lognormal dist. (σ_g, 1.29)
NO$_x$	0.712 kg[*]	Lognormal dist. (σ_g, 1.2)	-	-	-	-
Al recovered	0.143 kg[**]	Normal dist. (σ, 0.0089)	0.092 kg[**]	Normal dist. (σ, 0.00106)	0.00119 kg[***]	Normal dist. (σ, 0.00047)
Si recovered	1.576 kg[*]	Lognormal dist. (σ_g, 1.2)	1.576 kg[*]	Lognormal dist. (σ_g, 1.29)	1.576 kg[*]	Lognormal dist. (σ_g, 1.29)
Ag recovered	0.011 kg[**]	Normal dist. (σ, 0.00149)	0.008 kg[**]	Normal dist. (σ, 0.00017)	0.00953 kg[***]	Normal dist. (σ, 0.00033)

*Numerical values from Latunussa et al. (2016), **from Chung et al. (2021), *** this study, # assumed.

3.3.1 Life cycle impacts of chemical treatment process

The results of endpoint analysis for the chemical treatment process employing three different leaching agents are presented in Fig.4. The results represented only the environmental impacts, excluding the environmental disburdens associated with material recovery. The results revealed that the highest and lowest environmental impact was given by the leaching agent of HNO_3 and Thiourea, respectively. The highest impact regarding HNO_3 is because the of $Ca(OH)_2$ used for neutralization and NO_x emission during the electrolysis of HNO_3.
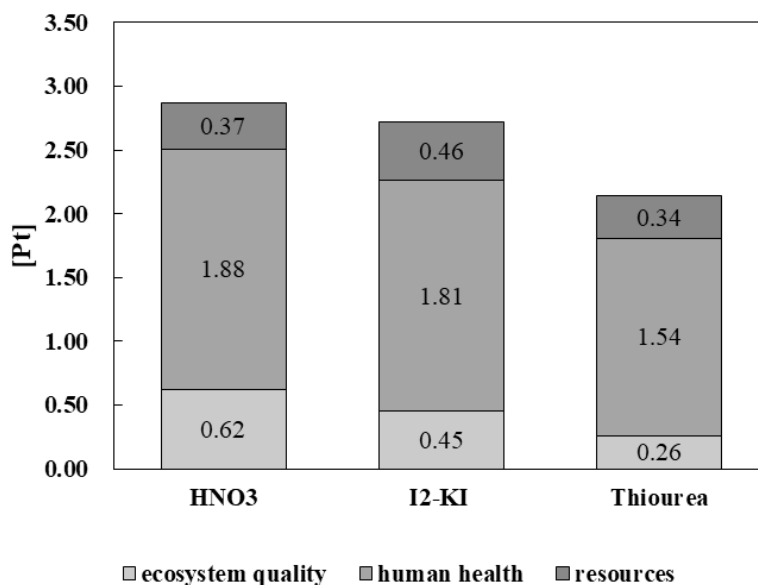

Figure 4. Environmental impacts associated with the chemical treatment of 2 kg of PV cell with three different leaching agents according to ReCiPe endpoint method

Table S1 shows the environmental impact results associated with the chemical treatment with three different leaching agents according to the ReCiPe midpoint method. The contribution of each step during the treatment is presented in Fig. 5, with relative values to the highest impacts scaled to 100%. For the impact categories of ODP, FEP, HTP, TETP, METP, and FDP, the chemical treatment using I_2-KI showed higher environmental impacts compared with the other two processes. These results are ascribed to the environmental burdens related to the utilization of different leaching agents. However, in the case of GWP, TAP, and POFP, the environmental impacts of the chemical treatment using HNO_3 as a leaching agent were found to be the highest compared with others. This was mainly ascribed to the NO_x emission during the electrolysis.
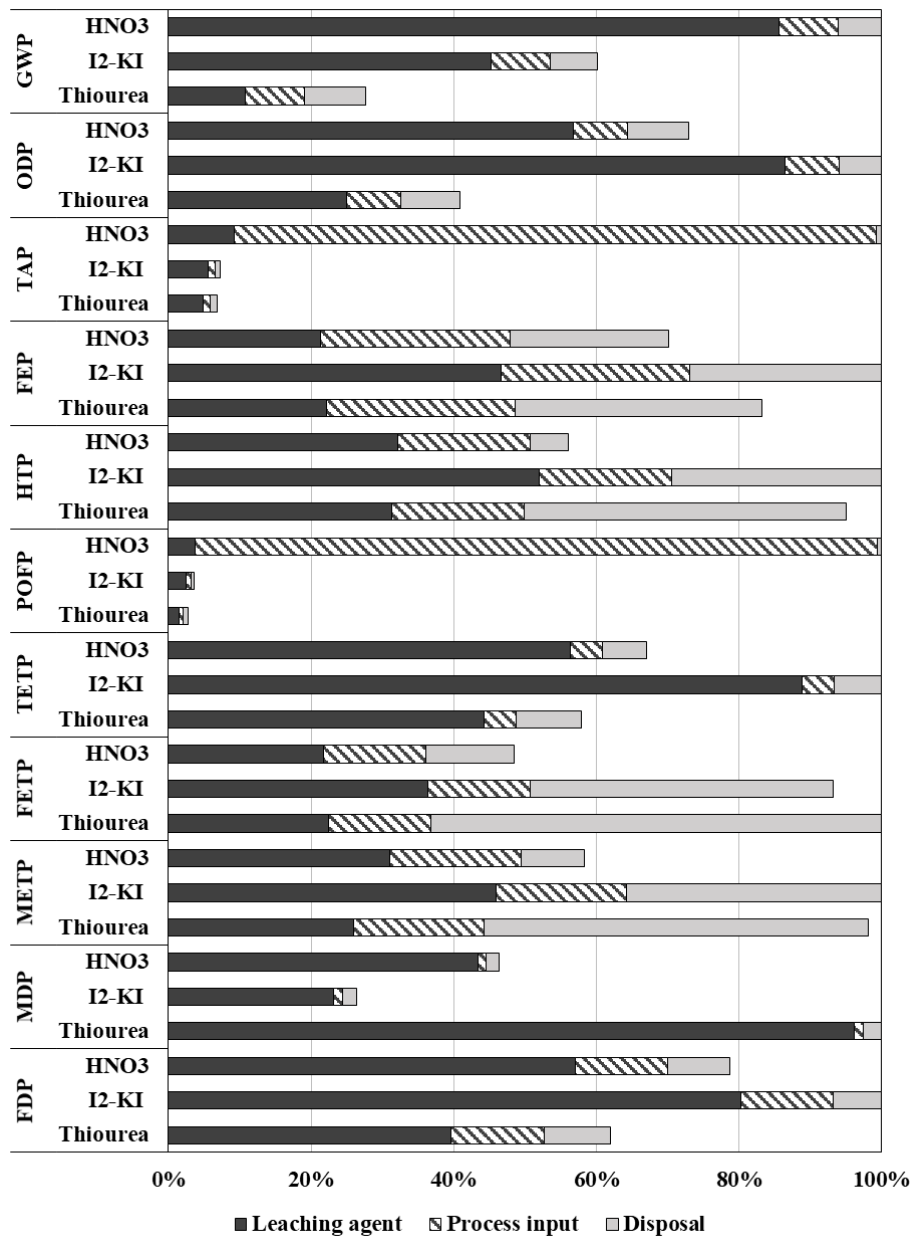
Figure 5. Relative environmental impact results of three different leaching agents for each midpoint indicator at individual process step; the maximum result is set to 100%.

3.3.2 Life cycle impacts of chemical treatment considering benefits by material recovery

In this section, the system boundaries have been extended to the production of secondary raw materials to assess whether and how far potential environmental benefits related to recycling exceed the environmental burden and disburden of the material recovery processes. Fig. 6 depicts the ReCiPe endpoint impacts of the material recovery using different leaching agents. The results showed that the environmental benefits associated with the material recovery of using HNO_3 as a leaching agent were higher than those with the other leaching agents. Among the recovery of aluminum, silicon, and silver, silicon recovery was founded to be mostly contributed to the avoided impact.
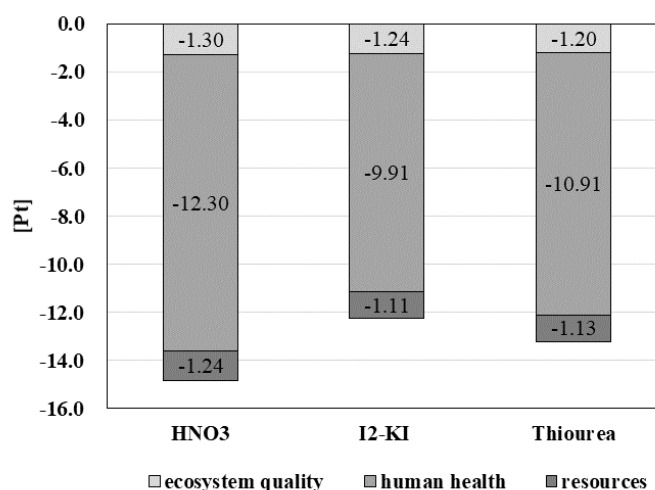
Figure 6. Environmental benefits associated with the material recovery of 2 kg of PV cell with three different leaching agents according to ReCiPe endpoint method.

The overall environmental impacts associated with chemical treatment, including the impacts from the material recovery, are shown according to the endpoint method. The score (net benefit) of HNO_3 leaching was higher than the other cases. The entire environmental benefit resulted from the material recovery was 5.2 times higher than the environmental impact by chemical treatment with HNO_3 leaching, whereas it was approximately 4.5 and 6.2 times higher in I_2-KI and thiourea leaching process, respectively.

Table S2 shows the results for the ReCiPe midpoint method. Except for ODP and TETP of I_2-KI process and TAP and POFP of HNO_3 process, the environmental benefits exceeded the environmental burdens by factors of 1.1 to 19.8 with HNO_3 leaching, by 1.5 to 25.3 with I_2-KI leaching, and by 1.4 to 7.6 with thiourea leaching depending on the categories. Among the LCIA indicators with three different leaching agents employed in chemical treatment, the values of GWP, POFP, HTP, and TETP are shown in Table S2, which illustrated the contribution of each process step and the recovered material to the overall environmental burdens and benefits analyzed by ReCiPe midpoint indicators. For the impact category of ODP and TETP using I_2-KI, the environmental impact from the entire recovery process was larger than the avoided impact. Among the avoided impacts related to material recovery, the Si recovery was found to be highest. For the impact categories of HTP, FETP, METP, FEP, and MDP, the avoided impacts related to material recovery were remarkably higher than the environmental burdens. For these impact categories, the environmental disburdens associated with silver recovery accounted for more than 77% of the total avoided impact.

3.3.3 Uncertainty analysis of chemical treatment

In this section, an uncertainty analysis of the environmental impacts for the chemical treatment of c-Si PV cell scraps using three different leaching agents was performed. Uncertainty analyses were carried out using Monte-Carlo simulation with 1,000 runs, which is one type of simulation that calculates the probability of the results based on random sampling (Groen et al., 2014). In this chapter, the estimated data adapted from the previous literatures (Latunussa et al., 2016; Chung et al., 2021) was assumed to have a log-normal distribution with geometric standard deviations, σg, which were chosen by Pedigree matrix (Weidema and Wesnaes, 1996). With respect to the input data related to the HNO_3 leaching process, 1.20 of σg was used, while 1.29 and 1.56 of σg were chosen for the material consumed and the amount of disposal, respectively, for I_2-KI and thiourea leaching process. On the other hand, the recovered amount was assumed to have a normal distribution with standard deviations adapted from the experimental results. The standard deviations for each material are shown in Table 1.

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Fig. 7 shows the uncertainty analysis results, which have revealed that the uncertainty values were found to be lower than the differences of environmental impacts for each chemical treatment method. On the other hand, the uncertainty values of 5% and 95% percentile for the material recovery were found to be within the differences in the environmental impacts between each result, as shown in Fig. 8. For the statistical analysis, the results were obtained by performing ANOVA tests. The results revealed that the impact variances from different leaching agents showed meaningful differences (p-Value < 0.05), as shown in Table S3 and S4.
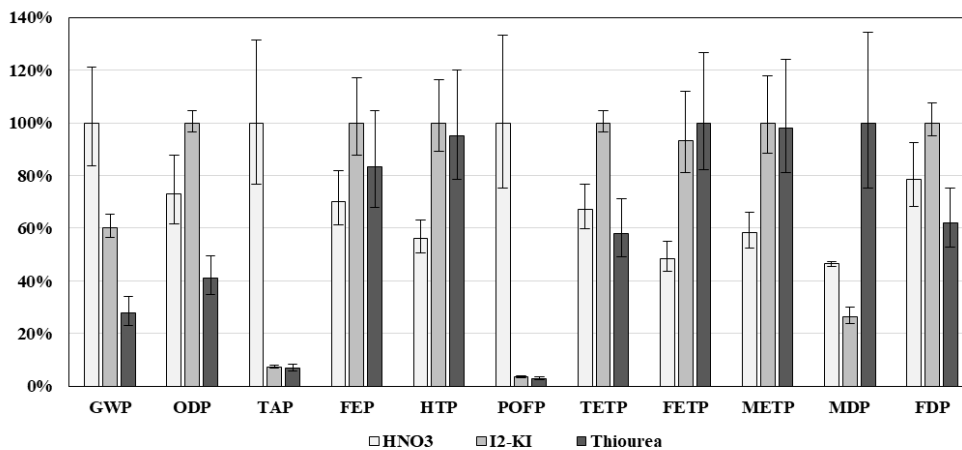

Figure 7. Environmental impacts comparison of chemical treatment using three different leaching agents with uncertainty analysis. The error bar indicates 90% confidence intervals.
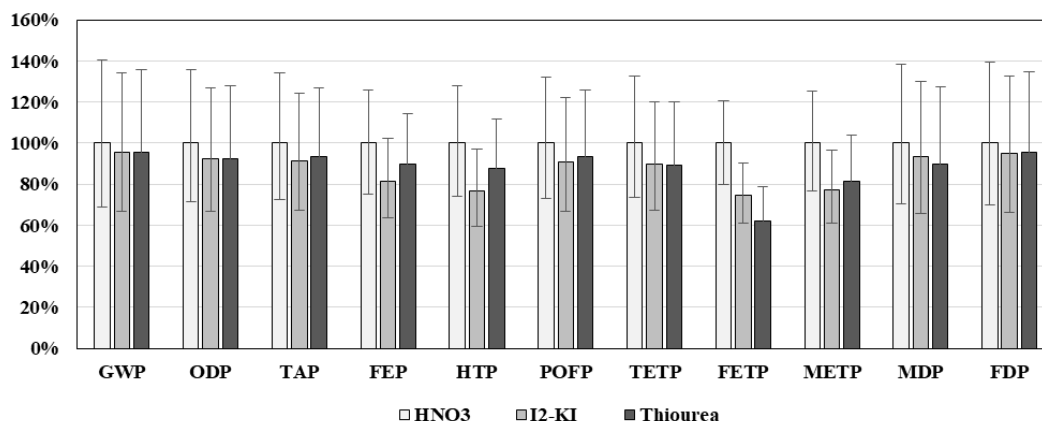

Figure 8. Environmental impacts comparison of material recovery using three different leaching agents with uncertainty analysis. The error bar indicates 90% confidence intervals.

4. CONCLUSION

This study gives a first attempt to evaluate the feasibility of TU-Fe system to replace the conventional leaching system using HNO_3 followed by neutralization process. Under the optimum conditions, the leaching capacity of TU-Fe system was nearly comparable to that of HNO_3, while the stoichiometric information from the experiment is successively substituted to the LCA to minimize the uncertainty from using arbitrary values from the literature.

The goal of this LCA was to compare the potential environmental impacts related to three different leaching agents (i.e., HNO_3, I_2-KI, and thiourea) for material recovery from EoL c-Si PV waste. The system boundary in this study included acid leaching, filtration, electrolysis, neutralization and landfilling for final

disposal. The environmental impacts produced from the high-rate recycling 2 kg PV cell scrap using HNO_3, I_2-KI, and thiourea solution were compared within the system boundary.

The ReCiPe results revealed that the highest environmental impacts were produced by chemical treatment using HNO_3 as a leaching agent, but the lowest was produced by thiourea leaching for the endpoint levels. The overall environmental impacts associated with the chemical treatment, including the impacts from the material recovery, revealed that the score (net benefit) of HNO_3 leaching was higher than the other two cases, resulted from more amount of the recovered materials, i.e., aluminum, silver, and silicon. In addition, the proposed alternatives for recycling of waste solar panels is expected to effectively reduce the recovery of resources (i.e., SRMs), energy consumption, and greenhouse gas emissions.

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Credit authorship contribution statement

Bora Seo: Conceptualization, Methodology, Writing - Original Draft. **Miyeon Lee**: Methodology, Writing- Original draft. **Bogyung Park**: Methodology, Visualization, Validation. **Jae Young Kim**: Methodology, Supervision, Writing - Review & Editing. **Jaeshik Chung**: Supervision, Writing - Review & Editing, Funding acquisition.

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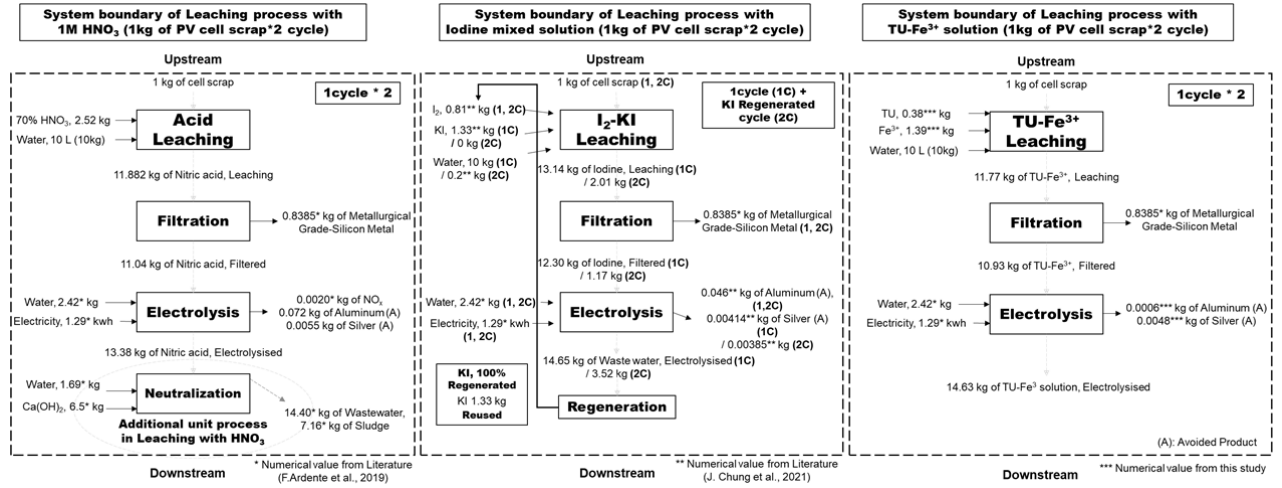
Figure S1. System boundaries of leaching process with 1 M HNO_3, 0.35 M I_2-0.7 M KI (two cycle comparison), and TU-Fe^{3+} solutions.
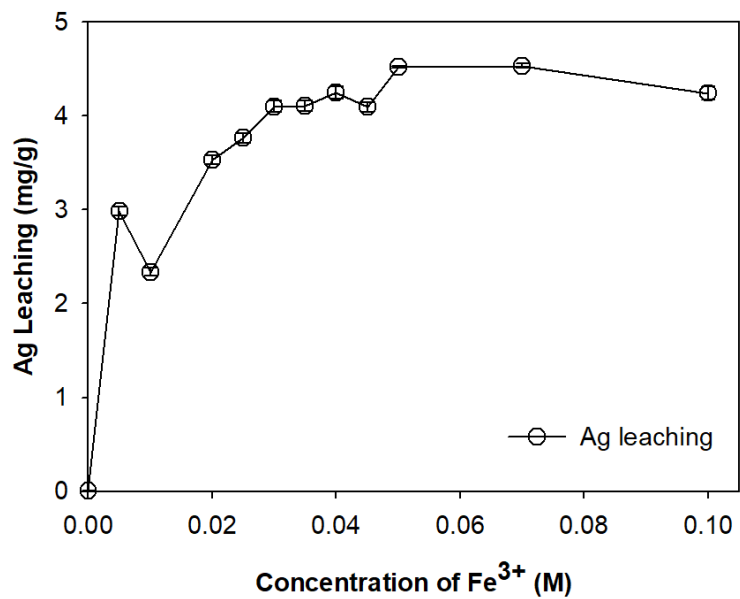
Figure S2. Effect of Fe^{3+} concentration on the equilibrium leaching amount of Ag and Al from 1 g of PV cell in TU-Fe^{3+} system (initial concentration of thiourea 1 M and S:L 0.5 g: 5 ml).

Table S1. Environmental impacts of each indicator associated with the chemical treatment of 2 kg of PV cell with three different leaching agents according to ReCiPe midpoint method.

Category	Unit	HNO$_3$ Leaching	I$_2$-KI Leaching	Thiourea Leaching
GWP	kg CO$_2$-Eq	1.94E+01	1.17E+01	5.36E+00
ODP	kg CFC-11-Eq	1.15E-06	1.58E-06	6.47E-07
TAP	kg SO$_2$-Eq	5.68E-01	4.15E-02	3.87E-02
FEP	kg P-Eq	3.07E-03	4.38E-03	3.65E-03
HTP	kg 1,4-DCB-Eq	1.14E+02	2.03E+02	1.93E+02
POFP	kg NMVOC	7.50E-01	2.74E-02	2.12E-02
TETP	kg 1,4-DCB-Eq	8.24E-03	1.23E-02	7.11E-03
FETP	kg 1,4-DCB-Eq	9.06E-02	1.74E-01	1.87E-01
METP	kg 1,4-DCB-Eq	1.01E+02	1.74E+02	1.71E+02
MDP	kg Fe-Eq	5.82E-01	3.30E-01	1.25E+00
FDP	kg oil-Eq	2.94E+00	3.74E+00	2.32E+00

Table S2. Environmental impacts of each indicator associated with the chemical treatment of 2 kg of PV cell with three different leaching agents according to ReCiPe midpoint method.

Impact category (Unit)	Leaching method	Leaching agent	Process input	Disposal	Al recovery	Ag recovery	Si recovery	Total
GWP (kg CO2-Eq)	HNO_3	1.66E+01	1.63E+00	1.18E+00	-6.93E-01	-3.35E+00	-1.87E+01	-3.36E+00
	I_2-KI	8.76E+00	1.63E+00	1.26E+00	-4.46E-01	-2.44E+00	-1.87E+01	-9.94E+00
	Thiourea	2.08E+00	1.63E+00	1.65E+00	-5.74E-03	-2.90E+00	-1.87E+01	-1.63E+01
ODP (kg CFC-11-Eq)	HNO_3	9.00E-07	1.20E-07	1.35E-07	-5.80E-08	-2.81E-07	-8.78E-07	-6.22E-08
	I_2-KI	1.37E-06	1.20E-07	9.39E-08	-3.73E-08	-2.04E-07	-8.78E-07	4.64E-07
	Thiourea	3.96E-07	1.20E-07	1.31E-07	-4.81E-10	-2.43E-07	-8.78E-07	-4.74E-07
TAP (kg SO2-Eq)	HNO_3	5.22E-02	5.11E-01	4.64E-03	-3.99E-03	-4.12E-02	-9.67E-02	4.26E-01
	I_2-KI	3.20E-02	5.43E-03	4.07E-03	-2.56E-03	-3.00E-02	-9.67E-02	-8.77E-02
	Thiourea	2.77E-02	5.43E-03	5.52E-03	-3.30E-05	-3.57E-02	-9.67E-02	-9.38E-02
FEP (kg P-Eq)	HNO_3	9.36E-04	1.16E-03	9.74E-04	-2.70E-04	-1.81E-02	-8.62E-03	-2.39E-02
	I_2-KI	2.04E-03	1.16E-03	1.18E-03	-1.74E-04	-1.32E-02	-8.62E-03	-1.76E-02
	Thiourea	9.70E-04	1.16E-03	1.52E-03	-2.24E-06	-1.57E-02	-8.62E-03	-2.07E-02
HTP (kg 1,4-DCB-Eq)	HNO_3	6.53E+01	3.77E+01	1.10E+01	-1.37E+01	-1.38E+03	-2.78E+02	-1.56E+03
	I_2-KI	1.06E+02	3.77E+01	5.99E+01	-8.83E+00	-1.00E+03	-2.78E+02	-1.09E+03
	Thiourea	6.38E+01	3.77E+01	9.18E+01	-1.14E-01	-1.19E+03	-2.78E+02	-1.28E+03
POFP (kg NMVOC)	HNO_3	2.86E-02	7.17E-01	4.85E-03	-2.41E-03	-3.72E-02	-7.21E-02	6.38E-01
	I_2-KI	1.88E-02	4.53E-03	4.03E-03	-1.55E-03	-2.71E-02	-7.21E-02	-7.34E-02
	Thiourea	1.11E-02	4.53E-03	5.52E-03	-2.00E-05	-3.23E-02	-7.21E-02	-8.32E-02
TETP (kg 1,4-DCB-Eq)	HNO_3	6.91E-03	5.59E-04	7.66E-04	-7.59E-04	-3.37E-03	-7.00E-03	-2.88E-03
	I_2-KI	1.09E-02	5.56E-04	8.19E-04	-4.88E-04	-2.45E-03	-7.00E-03	2.35E-03
	Thiourea	5.44E-03	5.57E-04	1.11E-03	-6.29E-06	-2.92E-03	-7.00E-03	-2.81E-03
FETP (kg 1,4-DCB-Eq)	HNO_3	4.06E-02	2.70E-02	2.30E-02	-3.64E-01	-5.96E-01	-2.03E-01	-1.07E+00
	I_2-KI	6.79E-02	2.69E-02	7.95E-02	-2.34E-01	-4.34E-01	-2.03E-01	-6.96E-01

	Thiourea	4.19E-02	2.70E-02	1.18E-01	-3.02E-03	-5.17E-01	-2.03E-01	-5.35E-01
METP (kg 1,4-DCB-Eq)	HNO$_3$	5.41E+01	3.19E+01	1.54E+01	-1.04E+02	-8.74E+02	-2.44E+02	-1.12E+03
	I$_2$-KI	7.98E+01	3.19E+01	6.22E+01	-6.67E+01	-6.36E+02	-2.44E+02	-7.73E+02
	Thiourea	4.51E+01	3.19E+01	9.37E+01	-8.59E-01	-7.58E+02	-2.44E+02	-8.32E+02
MDP (kg Fe-Eq)	HNO$_3$	5.43E-01	1.55E-02	2.27E-02	-3.11E-02	-1.13E+01	-1.56E-01	-1.09E+01
	I$_2$-KI	2.91E-01	1.54E-02	2.39E-02	-2.00E-02	-8.22E+00	-1.56E-01	-8.07E+00
	Thiourea	1.20E+00	1.54E-02	3.31E-02	-2.58E-04	-9.79E+00	-1.56E-01	-8.70E+00
FDP (kg oil-Eq)	HNO$_3$	2.13E+00	4.84E-01	3.26E-01	-1.71E-01	-1.04E+00	-4.71E+00	-2.98E+00
	I$_2$-KI	3.00E+00	4.84E-01	2.54E-01	-1.10E-01	-7.53E-01	-4.71E+00	-1.84E+00
	Thiourea	1.48E+00	4.84E-01	3.50E-01	-1.42E-03	-8.97E-01	-4.71E+00	-3.29E+00

Al: aluminum; Si: silicon; Ag: silver

Table S3. Uncertainty analysis of environmental impacts related to chemical treatment using three different leaching agents with ANOVA test results.

Impact Category	unit	HNO$_3$			I$_2$-KI			Thiourea			F-Value	p-Value
		Mean	5% Percentile	95% Percentile	Mean	5% Percentile	95% Percentile	Mean	5% Percentile	95% Percentile		
GWP	kg CO2-Eq	1.94E+01	1.62E+01	2.35E+01	1.17E+01	1.10E+01	1.27E+01	5.36E+00	4.49E+00	6.60E+00	1919.9	0.000
ODP	kg CFC-11-Eq	1.15E-06	9.75E-07	1.39E-06	1.58E-06	1.53E-06	1.67E-06	6.47E-07	5.54E-07	7.81E-07	9792.6	0.000
TAP	kg SO2-Eq	5.68E-01	4.36E-01	7.46E-01	4.15E-02	3.93E-02	4.50E-02	3.87E-02	3.28E-02	4.82E-02	1867.5	0.000
FEP	kg P-Eq	3.07E-03	2.68E-03	3.58E-03	4.38E-03	3.83E-03	5.28E-03	3.65E-03	2.97E-03	4.59E-03	558.0	0.000
MEP	kg N-Eq	1.14E+02	1.03E+02	1.28E+02	2.03E+02	1.75E+02	2.60E+02	1.93E+02	1.60E+02	2.44E+02	2822.2	0.000
HTP	kg 1,4-DCB-Eq	7.50E-01	5.65E-01	1.00E+00	2.74E-02	2.54E-02	3.06E-02	2.12E-02	1.82E-02	2.56E-02	2472.6	0.000
POFP	kg NMVOC	8.24E-03	7.33E-03	9.43E-03	1.23E-02	1.20E-02	1.28E-02	7.11E-03	6.05E-03	8.74E-03	1169.9	0.000
TETP	kg 1,4-DCB-Eq	9.06E-02	8.14E-02	1.03E-01	1.74E-01	1.44E-01	2.35E-01	1.87E-01	1.54E-01	2.37E-01	6229.4	0.000
FETP	kg 1,4-DCB-Eq	1.01E+02	9.13E+01	1.15E+02	1.74E+02	1.48E+02	2.27E+02	1.71E+02	1.41E+02	2.16E+02	2660.0	0.000
MDP	kg Fe-Eq	5.82E-01	5.72E-01	5.95E-01	3.30E-01	3.21E-01	3.45E-01	1.25E+00	9.44E-01	1.68E+00	1736.5	0.000
FDP	kg oil-Eq	2.94E+00	2.55E+00	3.46E+00	3.74E+00	3.55E+00	4.04E+00	2.32E+00	1.97E+00	2.82E+00	3350.9	0.000

Table S4. Uncertainty analysis of environmental benefits related to material recovery using three different leaching agents with ANOVA test results.

Impact Category	unit	HNO$_3$			I$_2$-KI			Thiourea			F-Value	p-Value
		Mean	5% Percentile	95% Percentile	Mean	5% Percentile	95% Percentile	Mean	5% Percentile	95% Percentile		
GWP	kg CO2-Eq	-2.3E+01	-3.3E+01	-1.6E+01	-2.2E+01	-3.1E+01	-1.5E+01	-2.2E+01	-3.1E+01	-1.5E+01	8.55	0.000
ODP	kg CFC-11-Eq	-1.2E-06	-1.7E-06	-8.9E-07	-1.1E-06	-1.6E-06	-8.3E-07	-1.1E-06	-1.6E-06	-8.2E-07	12.53	0.000
TAP	kg SO2-Eq	-1.4E-01	-1.9E-01	-1.0E-01	-1.3E-01	-1.8E-01	-9.7E-02	-1.4E-01	-1.8E-01	-9.9E-02	6.79	0.001
FEP	kg P-Eq	-2.8E-02	-3.5E-02	-2.1E-02	-2.2E-02	-2.8E-02	-1.7E-02	-2.5E-02	-3.1E-02	-1.9E-02	70.67	0.000
MEP	kg N-Eq	-1.7E+03	-2.2E+03	-1.3E+03	-1.3E+03	-1.7E+03	-1.0E+03	-1.5E+03	-1.9E+03	-1.1E+03	209.23	0.000
HTP	kg 1,4-DCB-Eq	-1.1E-01	-1.5E-01	-8.3E-02	-1.0E-01	-1.4E-01	-7.6E-02	-1.1E-01	-1.4E-01	-7.8E-02	10.97	0.000
POFP	kg NMVOC	-1.1E-02	-1.5E-02	-8.4E-03	-1.0E-02	-1.4E-02	-7.6E-03	-1.0E-02	-1.4E-02	-7.4E-03	18.32	0.000
TETP	kg 1,4-DCB-Eq	-1.2E+00	-1.4E+00	-9.5E-01	-8.8E-01	-1.1E+00	-7.2E-01	-7.3E-01	-9.3E-01	-5.7E-01	969.84	0.000
FETP	kg 1,4-DCB-Eq	-1.2E+03	-1.6E+03	-9.5E+02	-9.6E+02	-1.2E+03	-7.6E+02	-1.0E+03	-1.3E+03	-7.8E+02	251.76	0.000
MDP	kg Fe-Eq	-7.5E-04	-1.0E-03	-5.3E-04	-7.0E-04	-9.8E-04	-5.0E-04	-6.8E-04	-9.6E-04	-4.6E-04	361.58	0.000
FDP	kg oil-Eq	-6.0E+00	-8.4E+00	-4.2E+00	-5.7E+00	-8.0E+00	-4.0E+00	-5.8E+00	-8.1E+00	-4.0E+00	4.72	0.010