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Comparative environmental impacts analysis of technologies for recovering critical metals from copper anode slime: Insights from LCA

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ABSTRACT

Copper anode slime (CAS) is a byproduct produced during copper electrorefining process. It contains metals such as gold, silver, copper, selenium, tellurium etc. Without proper treatment, CAS posed significant environmental hazard due to its toxic components. Recovering critical metals from CAS not only mitigates environmental risks but also serves as an important source of these valuable materials. Recycling of critical metals can significantly enhance metal recycling efficiency and support the advancement of a circular economy. However, this process could introduce potential environmental impacts due to the increased consumption of energy, chemical material, and water. The process requires comprehensive assessment. In this study, life cycle assessment is employed to evaluate the potential environmental impact of the four resource recovery processes for copper anode slime: pyrometallurgy, hydrometallurgy, semi-hydrometallurgy, and combining bio-hydrometallurgy and semi-hydrometallurgy (CBS). The functional unit is 1 kg of copper anode slime. 5 metals are recycled during the process named: cupper (Cu), tellurium (Te), selenium (Se), gold (Au), and silver (Ag). Six impact categories—climate change, freshwater ecotoxicity, marine ecotoxicity, terrestrial ecotoxicity, human toxicity (cancer), and human toxicity (non-cancer)—were assessed and compared across the four recycling technologies. The LCA results show that CBS has the lowest environmental impact among all the assessed impact categories. CBS process demonstrated superior metal recovery rates. Hydrometallurgy has the lowest energy and material costs. CBS incurs higher total costs due to the use of expensive chemicals like potassium iodide.

1. Introduction

With the objective of carbon neutrality, the interdependence between metals and low-carbon energy conversion technologies has become increasingly close. Metals play a crucial role in these technologies [1,2]. Metals, especially critical metals are essential to support China's clean energy development and low-carbon transition. However, with the increasing competition and the growing demand for low-carbon energy technology, the sustainable supply of metal resources presents new challenges [3]. To support China's the "dual carbon" goals of carbon emissions peaking before 2030 and carbon neutrality before 2060, renewable energy technology deployment will drive a significant increase in the demand for critical metal [4]. It estimated that the demand for 20 types of metals, including selenium, tellurium, copper, platinum, and rare earths, will increase for 8.6-fold [5]. China, being a major consumer of many critical metal mineral, must develop sustainable strategies to meet this demand. Recycling critical metals and building an efficient recycling system is vital in supporting the low-carbon development.

Copper pyrometallurgical solid slag (CPSW) is primarily landfilled in China, with total landfills exceeding 1 billion tonnes. Copper anode slime (CAS), a significant byproduct of this process, represents an important resource for the recovery of precious metals like gold, silver, platinum, and palladium. However, this resource is largely underutilized. Globally, three conventional methods are used to treat anode slime: traditional pyrometallurgy, hydrometallurgy, and semi-

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hydrometallurgy [6]. Pyrometallurgical processes mainly include oxidative roasting [7], soda roasting [8], Kaldo furnace processes [9], and sulfuric acid roasting [10]. The traditional pyrometallurgical-electrolytic process remains in use today, but it faces significant challenges such as high return slag, low equipment utilization, and harmful smoke and dust emissions. Hydrometallurgical processes, such as acidic leaching [11], alkaline leaching [12], and chlorination leaching [13] offer several advantages over pyrometallurgy, including shorter production cycles and lower energy consumption. However, hydrometallurgical process often struggles with unstable production outcomes. Semi-hydrometallurgy combines the strengths of both pyrometallurgy and hydrometallurgy, reducing environmental impacts and enhancing resource recovery efficiency [14]. Despite these advantage, high investment costs and the need for extensive wastewater treatment remain major challenges. Bioleaching, an emerging low-carbon technology for mineral processing and non-ferrous metal extraction, is pivotal in remediation of heavy metal-contaminated mining sites [15,16]. Bioleaching involves the use of microorganisms to recover valuable metals by catalytic oxidation, dissolving them into a leachate in ionic form, or removing harmful elements from the minerals [17]. This method is characterized by low energy consumption, simple equipment requirements, and pollution-free operations. It has been industrialized for copper, gold, cobalt, nickel, zinc, uranium, and other metals, and has been promoted in more than 50 countries [18,19].

Building on the advantages of bioleaching technology and semi-hydrometallurgy, a novel recovering method, combining bio-hydrometallurgy and semi-hydrometallurgy (CBS), integrates the high efficiency of semi-hydrometallurgy with eco-friendly benefits of bio-hydrometallurgy. A comprehensive environmental and cost analysis of CBS recovering technology is conducted using life cycle assessment (LCA) and life cycle cost methodologies. The assessment results are compared with those of three conventional recovery methods, highlighting CBS's performance in terms of sustainability and cost-effectiveness.

Life cycle assessment (LCA) has become increasingly popular as a tool for evaluating the potential environmental impact of products and materials throughout their lifecycle [20]. The application of the LCA method in metal recovery has been widely studied in the academic community. For instance, researchers have conducted a complete LCA of aluminum recycling [21], the recovery of valuable metals from discarded printing plates [22], and batteries [23]. Recently, there has been extensive research on LCA in the copper slag recycling industry, focusing particularly on comparing the environmental benefits of primary copper mining [24], and secondary copper production [25]. Additionally, studies have explored the implementation of full life cycle analysis for the disposal of copper tailings [26], and copper slag [27]. The research team has also focused on the recovery for valuable metals from CPSW and conducted a life cycle assessment of the three conventional recovering technologies [27]. Furthermore, they evaluated technology of recovering critical metals from smelting slag residue, flue gas, and copper anode sludge generated by copper pyrometallurgical smelting. The findings indicate that electricity consumption is the primary contributor to global warming potential (GHG). For example, research shows that the impacts of GHG of recycling slag are 86 %, 52 %, and 79 % lower in the production of valuable metal copper concentrates, primary copper hydrometallurgy, and primary gold respectively [28].

The objective of this study is to perform a quantitative assessment of a novel recovery method, CBS, from environmental and economic perspective, and to compare the results with those of three conventional recovery methods. The article is structured as below, in section 2, the methodologies of LCA and life cycle costing (LCC) are presented alongside an analysis of three commonly used processes for recovering critical metals from copper anode slime: pyrometallurgy, hydrometallurgy, and semi-hydrometallurgy, as well as CBS. In section 3, the Life cycle impact results are presented and compared among these four technologies, as well as economic analysis. Sensitivity and uncertainty analysis is also presented in this section.

2. Method

2.1. Copper anode slime composition

CAS was collected from a hazardous slag recycling metal factory located in China, combined with relevant literature to analyze various technologies for metal recovery. The composition of the copper anode slime varies due to differences in copper refining technologies and the treatment of distinct copper products. The particle size of CAS typically ranges from 0.147 to 0.074 mm, with a light gray to gray-black color. The elemental content varies significantly, and its physical composition is complex. CAS generally contains Au, Ag, Pb, Se, Te, As, Sb, Bi, Fe, S, Sn, Ni, Cu, SiO_2, Al_2O_3, platinum group elements, and water (Cook et al., 2024). Therefore, this study examines the specific metal compounds and their concentrations in CAS, which are present in Table 1. Copper in CAS primarily exists in metallic form, with some copper compounds present. Silver is mainly found as monomorphic silver, silver selenide, and silver telluride, while gold primarily exists as monomorphic gold, gold selenide, and gold telluride [29]. Anode slime derived from copper concentrate smelting typically contains higher levels of Cu, S, Ag, Pb, Te, and small amounts of Au, Sb, Bi, As, and other minerals, along with minor platinum group elements. In contrast, CAS from copper-nickel sulfide ores is richer in Ni, Cu, S, Se, and platinum group metals but contains lower levels of Au, Ag, and Pb. Meanwhile, CAS from the electrolysis of miscellaneous copper contains more Pb and Sn [14].

2.2. Goal and scope

2.2.1. Goal and functional unit

The objective of this study is to assess the potential environmental impact of CAS recycling technologies using LCA. The climate change and other potential environmental impacts from CAS recycling technologies are comprehensively assessed, and quantified, and compared considering the variability of energy, and the recovery rate among different technologies. The results can provide scientific support for the construction of a critical metal recycling system under the low-carbon transition both in China and around the world.

Five metals are recycled from CAS in varying quantities depending on the recovery technique used. The functional unit is 1 kg copper anode slime. This functional unit provides a consistent and united basis for comparing potential environmental impacts of the four recovery technologies.

2.2.2. System boundary

The system boundary starts from the copper anode slime which is a byproduct generated during refining of copper and ends in recovering five critical metals, namely Au, Te, Se, Au and Ag. The system boundary captures all major inputs (materials and energy), processes, and output directly related to recovering CAS and ensures a comprehensive assessment and comparison of potential environmental impact of each technology. Detail descriptions for each recovering technologies and

Table 1
Composition of copper anode slime.

Metal composition	Percentage of Metal	Primary Metal Compounds (Percentage)
Cu	15	$CuSO_4$ (80 %), CuTe (2 %), CuSe (2 %), CuAgSe (12 %), Cu (4 %)
Au	0.2	Au (75 %), Au—Pb (25 %)
Ag	9.1	Ag_2Se (48.22 %), CuAgSe (33.25 %), AgTe (18.53 %)
Se	4	CuAgSe (55.34 %), CuSe (40.12 %)
Te	2	CuTe (14.74 %), AgTe (50 %), PbTe (35.26 %)

technical flows are presented in the section below.

2.2.2.1. Pyrometallurgy. Pyrometallurgy is a high-temperature smelting process used to recover precious metals from various sources, including CAS. It is one of the most used technologies for precious metal recovery due to its efficiency and scalability. The Kaldo furnace technology is integral to this process for CAS treatment (Fig. 1 A). Initially, pressurized oxidation acid leaching is employed to remove copper from the CAS called copper and tellurium leaching. In this step, copper anode slime undergoes pressure oxidation leaching in an acid solution at room temperature, effectively reducing the copper and tellurium content. The leachate generated can be directly recycled back into the copper electrolysis system to recover copper and tellurium. The decompressed slag is then smelted in the Kaldo furnace. During smelting, solvents and other additives are introduced to facilitate the enrichment of gold and silver in the anode slime, forming a gold-silver alloy. The gold and silver are subsequently extracted and separated using electrolytic refining. During the melting process, gold and silver accumulate as a gold-silver alloy, which undergoes electrolysis to isolate the two metals. Additionally, soot produced during the melting process is captured using a Venturi dust removal system. Crude selenium is then extracted from the recovered soot through wet leaching [30].

2.2.2.2. Hydrometallurgy. Hydrometallurgy is a chemical process that uses acids to extract precious metals from CAS. The primary steps of this recycling technology are present in Fig. 1 B. Copper anode slime undergoes a four-step leaching process, including copper sulfate leaching-electrolysis, gold chlorination, silver recovery through sodium treatment, and selenium-tellurium leaching from the gold reduction solution. Sulfuric acid is used to leach copper from CAS, converting it into copper sulfate. The resulting copper sulfate solution undergoes electrolysis, producing pure copper metal [11]. The copper slag is treated with

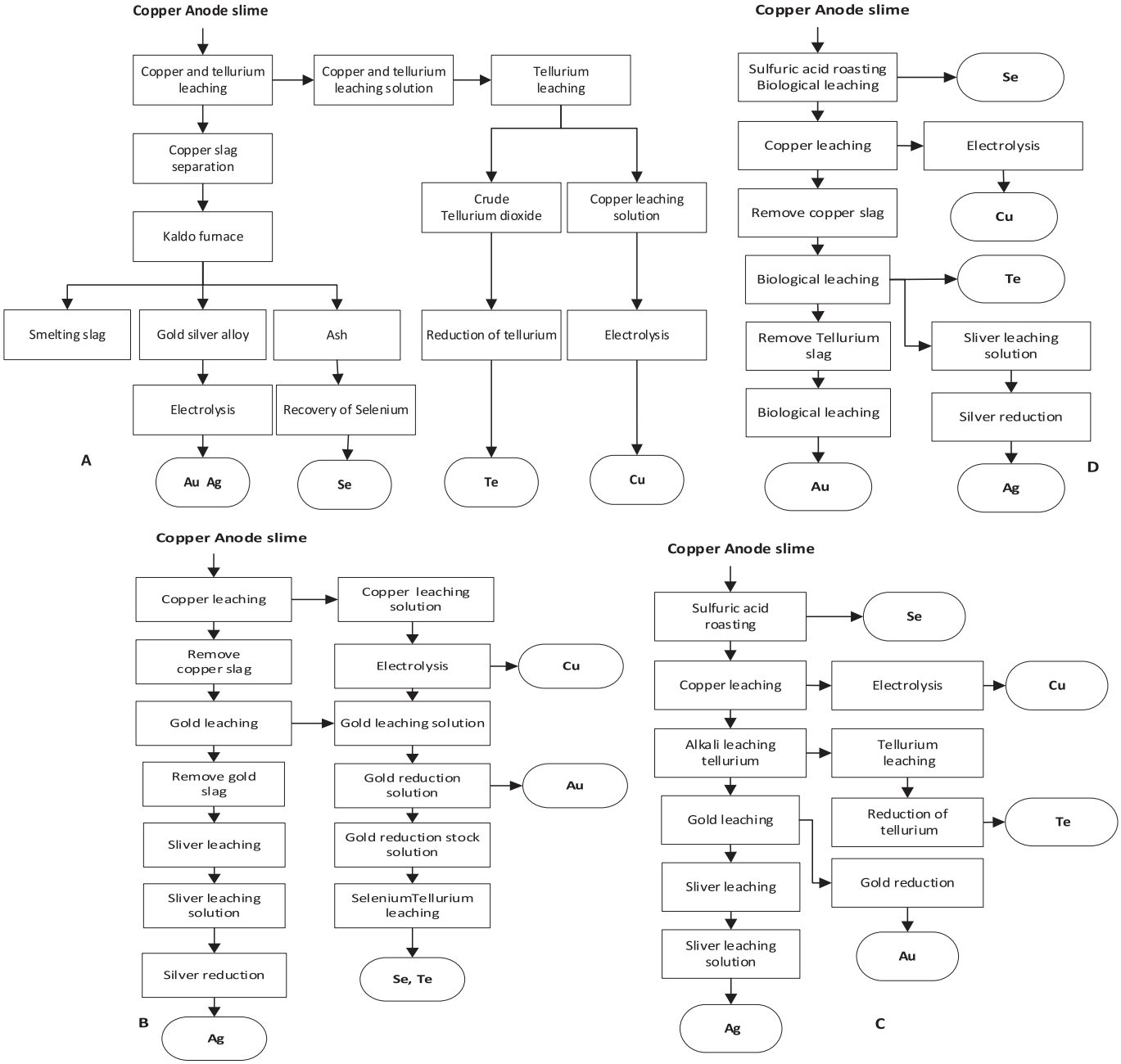


Fig. 1. Flow diagram for the recycling process for the assessed technology (A is pyrometallurgy, B is hydrometallurgy, C is semi-hydrometallurgy, D is CBS).

chlorination to extract gold. The gold solution is then reduced using sodium chlorate, iron sulfite, and other reagents, producing crude gold powder. The remaining gold slag is treated to extract silver using sodium salt, and the silver solution is reduced with formaldehyde (HCHO) to produce crude silver powder. Selenium and tellurium, present in the gold reduction solution, are leached out using hydrazine hydrate and other reagents [31]. This process results in the recovery of multiple valuable products, including copper, crude gold powder, crude silver powder, selenium, and tellurium, each separated and refined for further use [32].

2.2.2.3. Semi-hydrometallurgy. Semi-hydrometallurgy combines elements of both pyrometallurgy (high-temperature processing) and hydrometallurgy (chemical leaching) to recover metals from CAS. This approach integrates the strengths of pyrometallurgy and hydrometallurgy, increasing recovery rate of multiple metals. The primary steps of this recycling technology are present in Fig. 1 C. Copper anode slime undergoes sulfating roasting, during which selenium is oxidized to selenium dioxide. The selenium dioxide is volatilized and absorbed into an aqueous solution within the absorption tower, forming sodium selenite. This is subsequently reduced to elemental selenium using sulfur dioxide from furnace gases [8]. Selenium slag is treated with a low-acid solution to extract copper. The resulting copper sulfate solution undergoes electrolysis to produce copper in its metallic form. Copper slag is subjected to alkaline leaching to extract tellurium. The copper slag is neutralized using sodium hydroxide to obtain a tellurium leaching solution. This is then treated with sulfuric acid, hydrogen chloride, and other reagents to isolate elemental tellurium. Tellurium slag undergoes chlorination to extract gold. The gold solution is reduced using sodium chlorate, iron sulfite, and other reagents, resulting in crude gold powder. The gold slag is treated with sodium sulfite to extract silver. The resulting silver solution is reduced with formaldehyde (HCHO) to produce crude silver powder. Residual slag containing small amounts of gold and silver can be sold for further processing. Crude gold and silver powders are refined through casting and electrolysis to produce high-purity gold and silver powders, which are cast into final products [33].

2.2.2.4. Combining bio-hydrometallurgy and semi-hydrometallurgy. Combining bio-hydrometallurgy and semi-hydrometallurgy (CBS) is considered an innovative and sustainable treatment process for CAS, integrating the strengths of bioleaching and semi-hydrometallurgy (Fig. 1 D). The optimized CBS method integrates the efficiency of semi-hydrometallurgy with the eco-friendly advantages of bio-hydrometallurgy, providing a comprehensive solution for critical metal recovery from CAS. Bio-hydrometallurgy is a technique that leverages naturally occurring microorganisms to extract metal components from ores. Known for its environmentally friendly, low-carbon approach and high resource recovery efficiency, it holds significant potential for industrial applications. The sulfuric acid roasting and selenium evaporation technique is refined by replacing the sulfur dioxide reduction process with biodegradation. Bacteria strain QZB-1 is used under aerobic conditions to convert NaSeO3, formed at high temperatures in acidic environments, into elemental selenium [34]. The traditional hydrometallurgy recovery process for tellurium is replaced by a bio-leaching method. In the bioleaching process, *Pseudomonas mendocina* MCMB-180, using sucrose and diammonium phosphate as nutrients, directly reduces tellurium into its elemental form within the bacteria, replacing the reduction of tellurium dioxide. Pseudomonas Mendocino MCMB-180, in the presence of sucrose, and diammonium hydrogen phosphate as nutrients, reacted within the bacteria to directly reduce to tellurium monomers, and achieved a 99 % leaching rate of tellurium extracted from NaTeO3 solution. Excess sodium bisulfite, used in both bio-leaching for tellurium and the existing silver leaching process, converts metallic silver into complex ions in the leaching solution. After tellurium bio-leaching, formaldehyde reduces silver to its elemental state, shortening the original process. In the final leach residue, gold exists in its elemental form and is extracted using iodide-oxidizing bacteria (IOB). This replaces the conventional gold chloride leaching process, achieving efficient gold recovery [14,29].

2.3. Life cycle inventory

The inventory encompasses input and output data for materials and energy within the defined system boundary. Functional unit-based inputs and outputs are derived by gathering relevant data and performing necessary calculations [35]. Primary data on material usage and energy consumption for each recovery technology are detailed in Table 2. The detail input-output data for the four CAS recovering technologies are supplied in Appendix A. To conduct the life cycle analysis, the recycling technologies are divided into various sub-systems. For each sub-system, the resources' input and output (materials and energy) and the associated environmental disturbances are evaluated to generate a comprehensive quantitative result. Detailed data for each sub-system of the recycling technologies are provided in the Supplementary File. The database supporting this analysis integrates field research and literature data with reputable, validated databases, including the Ecoinvent database. This globally recognized LCA resource includes over 18,000 reliable datasets across industries like metals and mining, with substantial updates incorporating Chinese-specific data in 2022. The data collected on various environmental disturbances are organized into a life cycle inventory [36]. Besides Ecoinvent database, average electricity data were used as the electricity input from GaBi database from Sphera [37]. In this study, Chinese. Marginal electricity across China's regional grids was also identified for enhanced accuracy [38]. The primary objective is to compare the environmental and energy performance of various recycling technologies.

The purpose of technology cost analysis is to help businesses understand their cost structure, identify key cost components, evaluate cost-effectiveness, and develop strategies for cost control. This analysis supports pricing decisions and boosts competitiveness in production technologies. Additionally, technology cost analysis enables businesses to incorporate cost factors accurately when pricing products, ensuring that prices cover production costs while achieving profitability. Ultimately, the primary goal of technology cost analysis is to facilitate effective and efficient cost management, supporting sustainable growth and maintaining a competitive edge in the market [39]. Operating costs consist of variable costs and fixed costs. Variable production costs include power, direct materials, and labor, while fixed costs are mainly composed of depreciation expenses and capital investment [40]. Depreciation and capital investment costs are considered in this analysis. The cost data for the materials are the market price, and electricity cost is the cost for the average Chinese electricity cost for industrial consumer [41].

2.4. Life cycle impact assessment

A quantitative approach is used to analyze the critical metal recovery technologies of copper anode slime using the characterized ReCiPe2016midpoint(H) model. We selected six indicators to calculate the environmental impact of the process:climate change, freshwater ecotoxicity, marine ecotoxicity, terrestrial ecotoxicity, human toxicity, cancer and human toxicity, non-cancer (Table 3).

To assess climate change, the climate change is selected as the midpoint characterization factor, expressed in kilograms of CO_2 equivalents (kg CO_2-eq), which quantifies the infrared radiative forcing of greenhouse gases. At the midpoint level, the fate and effects of chemicals are characterized using 1,4-dichlorobenzene equivalents (1,4DCB-eq), covering human toxicity and various ecological toxicities (freshwater, marine, and terrestrial). The calculations are based on the global multimedia fate, exposure, and effect model USES-LCA 2.0, updated with data from the USEtox database to handle dissociating chemicals.

Table 2
Input and output of material and energy consumption and their associated costs.

	Material/ Energy	Unit price	Pyrometallurgy		Hydrometallurgy		Semi-Hydrometallurgy		CBS	
		Yuan/kg(Yuan/kwh)	Quantity	Cost	Quantity	Cost	Quantity	Cost	Quantity	Cost
			kg	Yuan	kg	Yuan	kg	Yuan	kg	Yuan
Material cost	H_2SO_4	0.36	0.77	0.28	1.55	0.56	1.96	0.71	0.92	0.33
	NaOH	3.80	0.84	3.19			0.15	0.57	0.15	0.57
	Na_2SO_3	2.80			0.46	1.27	0.48	1.33	0.37	1.04
	HCl	0.20	1.27	0.25						
	soda	1.80	0.05	0.09						
	lead oxide	19.18	0.03	0.56						
	quartz	2.50	0.01	0.03						
	Coke powder	1.27	0.02	0.03						
	HNO_3	1.67	0.50	0.84						
	SO_2	1.40	0.09	0.12			0.09	0.12		
	NaCl	0.60			0.15	0.09	0.17	0.10	0.06	0.03
	$NaClO_3$	4.20			0.16	0.66	0.15	0.62		
	H_2O_2	0.91			0.30	0.27				
	Formaldehyde	1.20			0.05	0.06	0.05	0.06	0.05	0.06
	Hydrazine hydrate	16.00			0.04	0.68				
	Fe	0.45			0.004	0.002	0.05	0.02		
	sugar	6.50							0.14	0.88
	$(NH_4)_2HPO_4$	7.00							0.07	0.47
	KI	500.00							0.25	126.00
	Water	0.004	4.64	0.02	13.30	0.05	20.47	0.08	123.50	0.51
Energy cost	Electricity	0.64	6.44	4.09	3.74	2.37	4.35	2.76	3.90	2.48
	diesel oil	7.04	0.12	0.84						
	Cu		0.15		0.15		0.15		0.15	
	Te		0.01		0.02		0.01		0.02	
Output	Au		0.002		0.002		0.002		0.002	
	Ag		0.15		0.15		0.15		0.15	
	Se		0.01		0.03		0.04		0.04	

Table 3
Life cycle impact assessment category.

Impact category	Unit
Climate change	kg CO2-eq
Freshwater ecotoxicity	kg 1,4-DCB
Marine ecotoxicity	kg 1,4-DCB
Terrestrial ecotoxicity	kg 1,4-DCB
Human toxicity, cancer	kg 1,4-DCB
Human toxicity, non-cancer	kg 1,4-DCB

The ecotoxicological effect factor represents the change in the probability density function of species due to a change in the environmental concentration of a chemical. The human toxicological effect factors separately evaluate carcinogenic and non-carcinogenic effects, reflecting the change in lifetime disease incidence due to a change in intake of the substance [42].

The calculation formula at the midpoint level primarily quantifies the contribution of each environmental impact category through characterization factors. The formula can be expressed as:

$$\text{Impact Category} = \sum (\text{Flow}^{*}\text{Characterization Factor})$$

Flow: Represents the emission or consumption of a specific substance, typically measured in units such as (kg), (kWh), or (m^3).

Characterization Factor: Used to convert the flow of a specific substance into its contribution to various environmental impact categories, with units of (impact units / emission units).

2.5. Sensitivity analysis method

The formula for calculating the Sensitivity Coefficient (SC_EI) is used to evaluate how sensitive a critical metal recovery system is to environmental impacts caused by variations in specific parameters. This approach identifies the system's response to key inputs like material and electricity consumption. A ± 10 % variation in the parameter value is applied to assess its influence on environmental impacts.

$$\text{SC_EI} = \left(\text{EI_}(-10\%) - \text{EI_}(+10\%)\right) / (\text{EI_base}) \text{“}\times\text{”} 100\%$$

SCEI:Sensitivity Coefficient , %,

EI-10 %:The environmental impact value obtained when the parameter value is reduced by 10 % , kg CO_2-eq,

EI + 10 %:The environmental impact value obtained when the parameter value is increased by 10 % , kg CO_2-eq,

EI_{base}:The environmental impact value obtained when the parameter is at its baseline value , kg CO_2-eq.

3. Results and discussion

3.1. Comparison of technology resource recovery rate

The performance of the four technologies is evaluated by the mass of metals recovered (Table 4) and recover rate (Table 5). Pyrometallurgy demonstrates excellent performance in recovering gold and silver, with recovery rates of 99.8 % and 98 %, respectively. However, its recovery rates for selenium and tellurium are significantly lower, at only 27.86 % and 37.3 %, respectively. Hydrometallurgy generally outperforms pyrometallurgy in terms of recovering Se and Te achieving recycling rate of 94.08 % and 76.32 %, respectively. Nevertheless, its gold recovery rate is relatively lower at 93.4 %. %. Semi-hydrometallurgy shows balanced recovery rates across all elements, with selenium recovery reaching an impressive 99.39 %, underscoring its potential for effective multi-metal recovery. CBS demonstrates significant advantages in resource recovery,

Table 4
Metal recovered from CAS unit kg.

	Pyrometallurgy	Hydrometallurgy	Semi-hydrometallurgy	CBS
Cu	1.49E-01	1.49E-01	1.47E-01	1.47E-01
Au	1.99E-03	1.87E-03	1.87E-03	1.87E-03
Ag	1.48E-01	1.48E-01	1.48E-01	1.48E-01
Se	1.30E-02	2.85E-02	4.00E-02	3.80E-02
Te	1.29E-02	1.53E-02	1.29E-02	1.99E-02

Table 5
Metal recovered rate from CAS.

	Au	Ag	Se	Te	Cu
Pyrometallurgy	99.80 %	98.00 %	27.86 %	37.30 %	98.00 %
Hydrometallurgy	93.40 %	98.88 %	94.08 %	76.32 %	99.28 %
Semi-Hydrometallurgy	98.80 %	99.09 %	99.39 %	76.70 %	99.10 %
CBS	98.10 %	98.10 %	98.55 %	96.20 %	99.10 %

especially for copper anode slime. It achieves high recovery rates for gold and silver (98 %), selenium (98.6 %), and copper (99.1 %). The CBS approach leverages microorganisms or their metabolic products to facilitate metal dissolution and recovery. The CBS method offers competitive recovery, especially for Te, despite having slightly lower recovery of Cu. Silver recovery is uniform across all methods, suggesting similar efficiency for this metal regardless of the process used. The amounts of recycled Cu are similar across all technologies, with hydrometallurgy slightly outperforming than others. Pyrometallurgy shows the highest recovery of Au. All methods achieve recovery of silver at 0.14847 kg. Semi-hydrometallurgy demonstrates the highest recovery of Se at 0.04 kg. followed by CBS, recovering 0.038 kg. CBS how the clear advantage over the other three technologies of recycling Te at 0.0199 kg. By improving recovery rates and resource utilization efficiency, CBS effectively extracts metals that are otherwise challenging to recover using conventional methods.

3.2. Environmental impact analysis of three technologies

3.2.1. Climate change

A quantitative approach is used to analyze the critical metal recovery technologies of copper anode slime using the characterized ReCiPe2016midpoint(H) model. The climate change (measured in CO_2-eq kg/FU) associated with four different metallurgical processes pyrometallurgy, Hydrometallurgy, Semi-hydrometallurgy, and CBS are compared. The CO_2 emissions are shown for each process, broken down by the specific contributions from five metals: Cu, Te, Se, Au, and Ag (Fig. 2). Pyrometallurgy is shown to have the highest CO_2 emissions of 9.31 kg, with the greatest contribution from silver (Ag), followed by copper (Cu). Hydrometallurgy shows a significant reduction in greenhouse gas (GHG) emissions compared to pyrometallurgy with CO_2 emissions of 5.28 kg. Copper remains a major contributor, but tellurium and selenium also contribute to emissions, though to a lesser extent. Semi-hydrometallurgy also results in reduced emissions, with copper and silver still having a noticeable impact. However, the CO_2 emissions of 5.26 kg, are lower than both pyrometallurgy and hydrometallurgy. CBS stands out with the lowest CO_2 emissions of 3.67 kg. This is due to the reduced reliance on energy-intensive processes, with contributions from copper, gold, silver, and selenium all being relatively minimal.

Electricity consumption is the largest contributor to the impact of clime change among all four technologies. Pyrometallurgical process, with the highest clime change impact, is more energy-intensive, due to the high temperatures required for smelting (Fig. 3). The high energy consumption is due to the leaching of CuTe with the melting technology of the Kaldo furnace and electrolysis of recovering Cu, Au, and Ag. Besides electricity, the consumption of HNO3 and NaOH during gold and silver electrolysis and tellurium leaching respectively, account for 21.2 % and 12.5 % of GHG emissions respectively.

Electricity accounts for 43.7 % of climate change in the hydrometallurgy process. Material consumption accounts for 53.0 % of total climate change which is the highest compared to the other three technologies. Chemicals like NaCl, Na_2SO_3, H_2SO_4, and Formaldehyde play significant roles in hydrometallurgy. During CBS process, electricity consumption, accounting for 65.5 % of total carbon emissions, caused 2.40 kg CO_2-eq, which is the lowest among assessed technology. Na_2SO_3 (sodium sulfite), formaldehyde, and $(NH_4)_2HPO_4$ (ammonium phosphate) contribute more to the CBS process, reflecting the innovative use of reagents in this method for bioleaching and resource recovery.

Fig. 2 and Fig. 3 demonstrate the substantial reliance on electricity across all processes, suggesting that transitioning to renewable energy sources could significantly reduce potential environmental impacts. Additionally, CBS appears to optimize chemical use while maintaining high reliance on electricity, underlining its innovative and sustainable approach in metal recovery. It suggests that CBS will be increasingly competitive with respect to GHG emissions as the transition to low-carbon electricity.

3.2.2. Ecotoxicity analysis

Ecotoxicity mainly studies the degree of pollutants or activity to

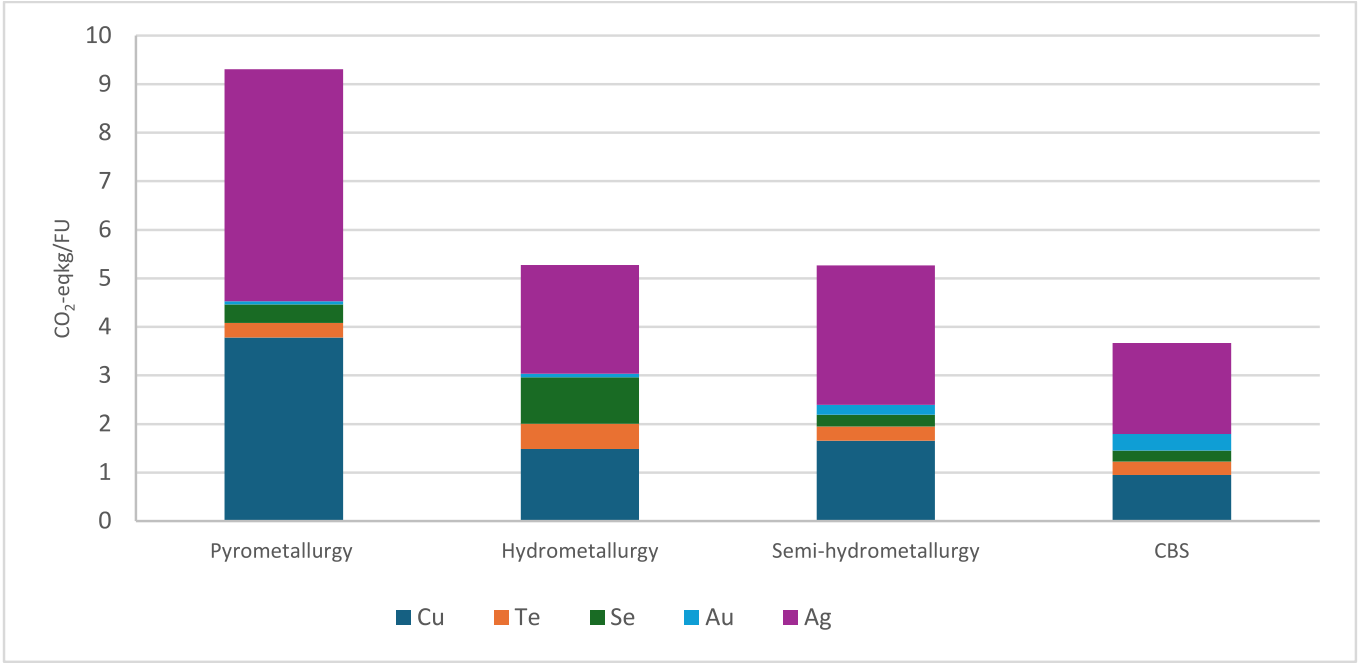


Fig. 2. Climate change from Pyrometallurgy, Hydrometallurgy, and Semi-Hydrometallurgy and CBS.

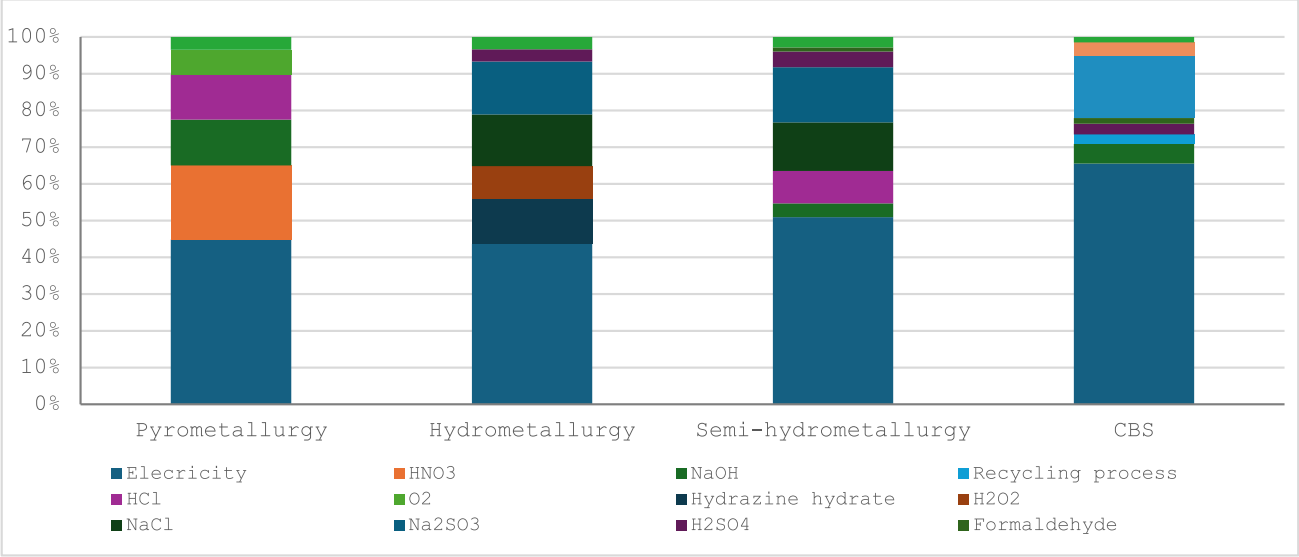


Fig. 3. Shares of key material and energy to climate change of the four recycling technologies.

harmful effects on environment and ecosystem. Terrestrial ecotoxicity potential, freshwater ecotoxicity potential, and marine ecotoxicity potential of the four CAS recycling technologies are analyzed (Fig. 4). From the figures, in terms of ecotoxicity, hydrometallurgy causes the greatest impacts of freshwater ecotoxicity potential of 0.40 kg 1,4-DCB and terrestrial ecotoxicity potential of 48.8 kg 1,4-DCB. Pyrometallurgy causes the greatest marine ecotoxicity potential impact of 0.20 kg 1,4-DCB. CBS recovering technology shows the lowest ecotoxicity impacts potential.

Hydrometallurgy exhibits the highest freshwater ecotoxicity potential and terrestrial ecotoxicity potential, primarily driven by selenium and copper contributions, nearly 5 times higher than the CBS technology of 0.07 kg 1,4-DCB and 0.09 kg 1,4-DCB respectively. Selenium accounts for a substantial share of the impact due to wet leaching of copper tellurium, with a contribution of 90 %, which is mainly the use of chemicals (sulfuric acid), in the case of hydrometallurgy. Pyrometallurgy has moderate freshwater ecotoxicity potential compared to the other processes. The dominant contributions are from copper and smaller proportions of tellurium and selenium due to wet leaching.

Semi-hydrometallurgy shows a reduced ecotoxicity impact compared to pyrometallurgy and hydrometallurgy. Copper is the primary contributor, with minor contributions from tellurium and selenium. CBS demonstrates the lowest freshwater ecotoxicity potential among the processes. Its lower environmental footprint highlights its

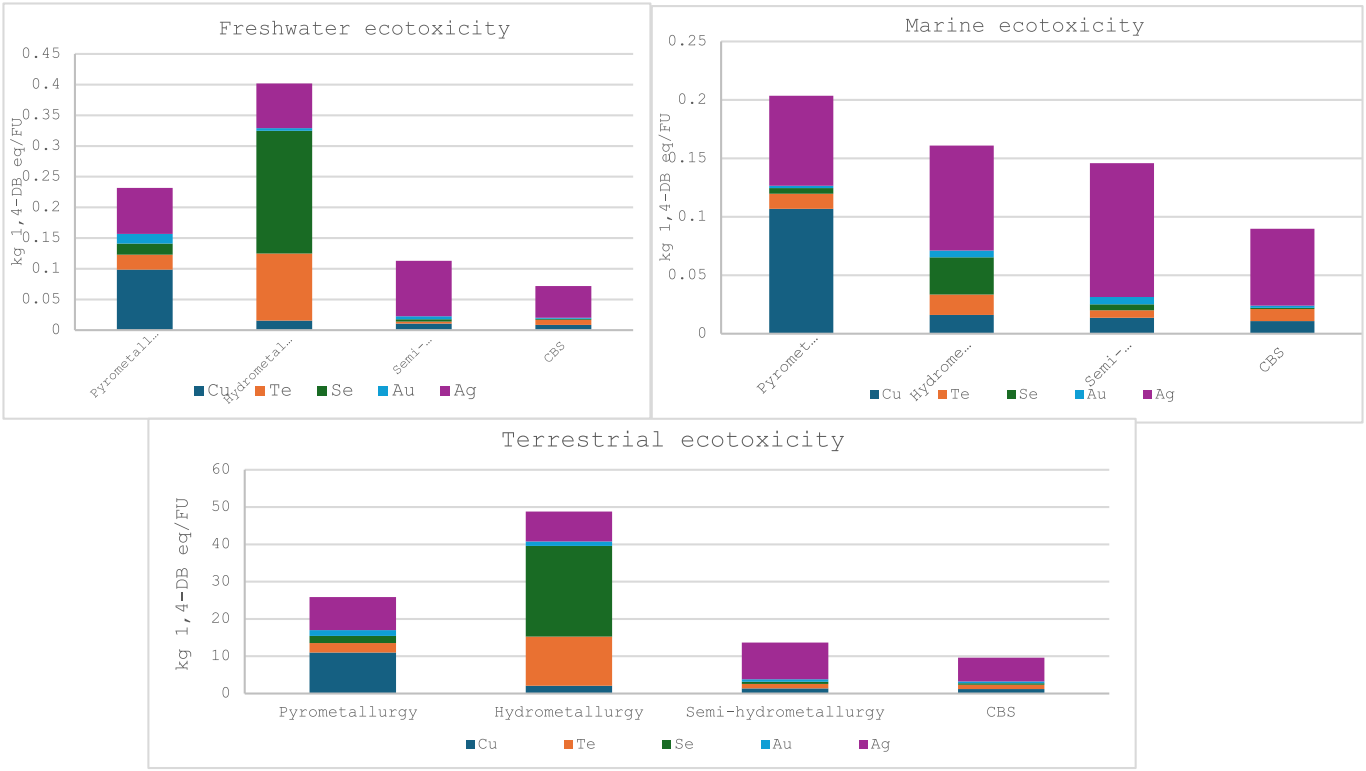


Fig. 4. Comparison of ecotoxicity of pyrometallurgy, hydrometallurgy, semi-hydrometallurgy, and CBS.

advantage in minimizing ecotoxicological impacts, especially for copper and selenium recovery.

A comparison of the ecotoxicity of the critical metals recovered by pyrometallurgy, hydrometallurgy, semi-hydrometallurgy, and CBS reveals that CBS has the lowest impact. The main cause of the high environmental impacts of pyrometallurgy, and hydrometallurgy involves the use of chemical material such as HNO_3 Na_2SO_3, and NaCl etc., which may cause toxicity to living organisms, and the accumulation of chemicals in living organisms, resulting in a reduction of biodiversity, and the impairment of ecological functions.

3.2.3. Human health assessment

From the perspective of human health, different CAS recycling technologies are mainly for human toxicity cancer, and human toxicity non cancer (Fig. 5).

In terms of human health, pyrometallurgy has the highest impact on human toxicity non-cancer, which is 5.09 kg 1,4-DCB. This is primarily due to the pressurized leaching of copper tellurium during high pressure leaching, where the contribution rate reaches 90 %, mainly driven by the use of chemical drugs such as sulfuric acid. For hydrometallurgy, the impacts on human toxicity cancer, and human toxicity non-cancer are 0.27 kg 1,4-DCB and 4.30 kg 1,4-DCB respectively. The primary cause of human toxicity cancer is the leaching copper, selenium, and tellurium attributed to high electricity use and chemicals such as sulfuric acid and hydrazine hydrate. Semi-hydrometallurgical shows impact 0.26 kg 1,4-DCB for human toxicity cancer, and 4.07 kg 1,4-DCB for human toxicity non-cancer. These effects are primarily caused by the leaching of copper and silver, largely due to high electricity usage and the application of chemicals such as sulfuric acid, sodium sulphatic, and sodium hydroxide. CBS demonstrates the lowest human toxicities, with 0.11 kg 1,4-DCB for human toxicity cancer, and 2.49 kg 1,4-DCB for human toxicity non-cancer - half the values observed for semi-hydrometallurgy. The main contributors to both human toxicity cancer and non-cancer are leaching silver and telluride, with sodium metabisulfite serving as the key chemical agent.

A comparison of the human health impacts of pyrometallurgy, hydrometallurgy, semi-hydrometallurgy, and CBS, CBS has the least impact on human health, while pyrometallurgy and hydrometallurgy show higher environmental impacts. Furthermore, the study reveals that the main cause of human health impact in pyrometallurgy, and hydrometallurgy, semi-hydrometallurgy, and CBS process is the use of sodium metabisulfite.

3.3. Economic analysis

An economic analysis was undertaken based on the power cost, and material cost of different technology types for processing one ton of CAS. It can be seen from the data (Table 6) that the power cost of the pyrometallurgy is the highest (4.93 yuan), followed by the CBS (2.48 yuan), while the power cost of the hydrometallurgy is the lowest (2.37 yuan).

Table 6
Economic analysis Unit: Yuan.

	Material cost	power cost	total cost
Pyrometallurgy	5.41	4.93	10.35
Hydrometallurgy	3.65	2.37	6.03
Semi-Hydrometallurgy	3.61	2.76	6.37
CBS	129.88	2.48	132.36

The material cost is 5.41yuan, 3.65 yuan, 3.61 yuan, and 129.88 yuan for pyrometallurgy, hydrometallurgy, semi-hydrometallurgy, and CBS respectively. CBS has an exceptionally high material cost, leading to the highest total cost among all processes. Despite its low power cost, the high cost of materials significantly drives up its overall expense, followed by pyrometallurgy, while the material cost of the hydrometallurgy is the lowest.

Hydrometallurgy and Semi-hydrometallurgy are the most economical options in terms of total cost, offering cost-effective solutions with moderate power and material expenses. CBS, while environmentally favorable, incurs an overwhelmingly high material cost, making it economically less viable in comparison. Pyrometallurgy, though less expensive than CBS, still faces higher costs than wet processes due to substantial power and material requirements.

3.4. Sensitivity analysis

For pyrometallurgy, the sensitivity analysis shows that under climatee change, electricity used during copper and tellurium leaching has the highest sensitivity, while H_2SO_4 (sulfuric acid roasting) exhibits the lowest (Fig. 6 A). In both terrestrial ecotoxicity and freshwater ecotoxicity, hydrazine hydrate used in selenium and tellurium leaching shows the highest sensitivity, while diesel oil used in the Kaldo furnace shows the lowest. For marine ecotoxicity, lead oxide used in the Kaldo furnace has the highest sensitivity, and diesel oil used in the Kaldo furnace has the lowest. In terms of human toxicity cancer, electricity during copper and tellurium leaching shows the highest sensitivity, and diesel oil in the Kaldo furnace shows the lowest. Similarly, for human toxicity non-cancer, diesel oil in the Kaldo furnace is the least sensitive.

The sensitivity analysis of hydrometallurgy reveals that under climate change, electricity consumption during copper leaching exhibits the highest sensitivity, while H_2SO_4 (sulfuric acid) used in copper leaching shows the lowest sensitivity (Fig. 6 B). In terms of terrestrial ecotoxicity and freshwater ecotoxicity, NaOH used in copper and tellurium leaching demonstrates the highest sensitivity, while electricity consumption during selenium and tellurium leaching shows the lowest. For Marine Ecotoxicity, H_2SO_4 (copper leaching) has the highest sensitivity, and electricity consumption in selenium and tellurium leaching has the lowest sensitivity. In the context of human toxicity cancer, electricity consumption during copper leaching exhibits the highest sensitivity, while H_2SO_4 (copper leaching) shows the lowest. Lastly, for human toxicity non-cancer, H_2SO_4 (copper leaching) displays the

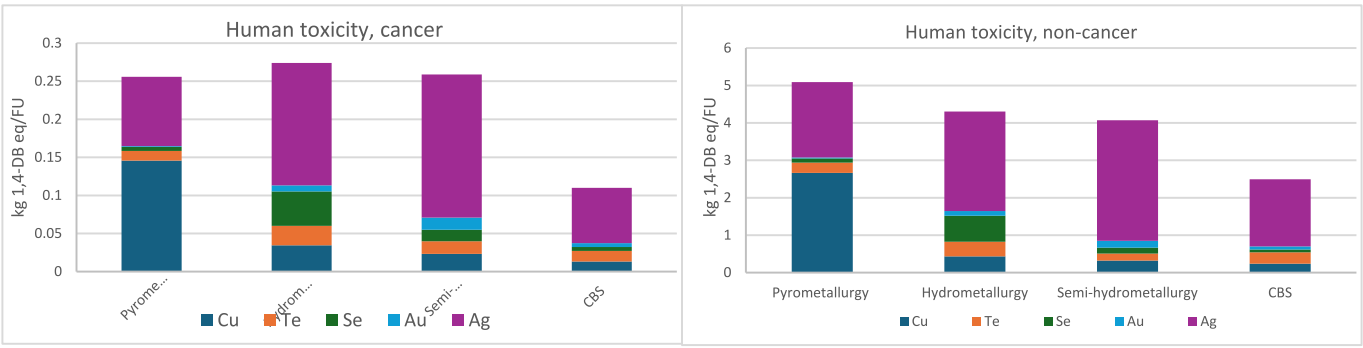


Fig. 5. Comparison of human health evaluation of pyrometallurgy, hydrometallurgy, semi-hydrometallurgy, and CBS.

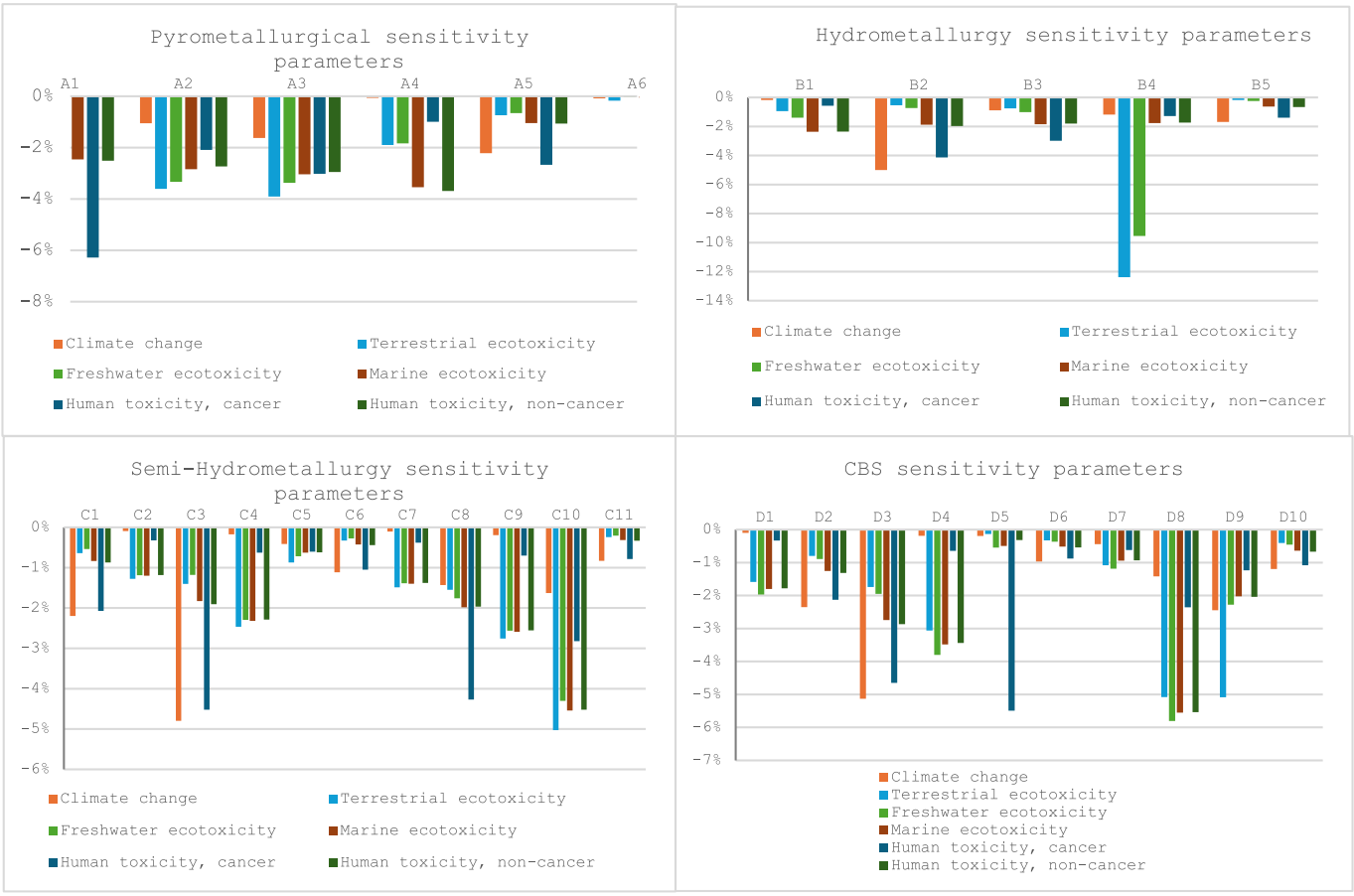


Fig. 6. Technology sensitivity analysis (A is the sensitivity analysis of pyrometallurgy, B is the sensitivity analysis of hydrometallurgy, C is the sensitivity analysis of semi hydrometallurgy, D is the sensitivity analysis of CBS).

highest sensitivity, and electricity consumption during selenium and tellurium leaching has the lowest sensitivity.

For semi-hydrometallurgy, the sensitivity analysis indicates that with climate change, electricity used in copper leaching shows the highest sensitivity, while electricity used in sulfuric acid roasting exhibits the lowest (Fig. 6 C). In terrestrial ecotoxicity, freshwater ecotoxicity, and marine ecotoxicity, Na_2SO_3 used in silver leaching demonstrates the highest sensitivity, while electricity used in silver leaching shows the lowest. In human toxicity cancer, electricity used in copper leaching shows the highest sensitivity, while H_2SO_4 in sulfuric acid roasting shows the lowest. Similarly, for human toxicity non-cancer, Na_2SO_3 (silver leaching) shows the highest sensitivity, and electricity (silver leaching) shows the lowest.

Finally, in CBS, under climate change, electricity used in copper leaching shows the highest sensitivity, while H_2SO_4 in sulfuric acid roasting and biological leaching shows the lowest (Fig. 6 D). In terrestrial ecotoxicity, KI in biological leaching of gold shows the highest sensitivity, while $(NH_4)_2HPO_4$ used in biological leaching of tellurium shows the lowest. For freshwater ecotoxicity and marine ecotoxicity, Na_2SO_3 in silver leaching exhibits high sensitivity, while electricity in biological leaching of tellurium shows the lowest sensitivity in freshwater and $(NH_4)_2HPO_4$ in biological leaching of tellurium shows the lowest in marine ecotoxicity. In human toxicity cancer, $(NH_4)_2HPO_4$ in biological leaching of tellurium shows the highest sensitivity, while H_2SO_4 in sulfuric acid roasting and biological leaching shows the lowest. For human toxicity non-cancer, electricity used in copper leaching shows the highest sensitivity, while $(NH_4)_2HPO_4$ in biological leaching of tellurium shows the lowest sensitivity.

To reduce emissions and minimize environmental pollution across the various metallurgical methods, a multi-faceted approach is necessary. First, efforts should be made to optimize electricity consumption, as it shows the highest sensitivity in terms of climate change and Human toxicity cancer. This can be achieved by integrating renewable energy sources, improving energy efficiency, and exploring alternative, low-energy leaching technologies. For chemicals like sulfuric acid (H_2SO_4) and sodium hydroxide (NaOH), which are less sensitive in some cases but still contribute to environmental and human health risks, alternatives or process improvements that reduce chemical use or enhance recycling should be explored. Additionally, the use of toxic substances such as hydrazine hydrate, KI, and Na_2SO_3 in various leaching processes should be minimized by investigating more environmentally benign chemicals or optimizing the leaching processes to reduce the quantities required. Overall, the adoption of cleaner technologies, energy-efficient practices, and safer chemical substitutes will help mitigate environmental and health impacts in metallurgical processes.

4. Conclusion

The objective of this study is to perform a quantitative assessment of a novel recovery method, CBS, from environmental and economic perspective, and to compare the results with those of three conventional recovery methods. The functional unit is 1 kg of copper anode slime. The findings provide a theoretical foundation and improvement trends for further establishing a green and sustainable recycling system for the recovery of key metals from copper anode slime.

This study evaluates the environmental impacts of various copper anode sludge recovery technologies using life cycle assessment, with 1 kg of recovered material as the functional unit.

Among the four technologies analyzed of pyrometallurgy, hydrometallurgy, semi-hydrometallurgy, and CBS), CBS, demonstrated the lowest potential environmental impact in terms of greenhouse gas emissions, ecotoxicity, and human health. This made it the most environmentally favorable process. The CBS process demonstrated superior metal recovery rates, particularly for gold, silver, selenium, tellurium, and copper. Hydrometallurgy has the lowest energy and material costs. CBS incurs higher total costs due to the use of expensive chemicals like potassium iodide. The sensitivity analysis shows chemicals are key factors affecting environmental performance and should be prioritized for optimization. Pyrometallurgy is most sensitive to hydrochloric acid. Hydrometallurgy is most sensitive to hydrazine hydrate. Both semi-hydrometallurgy and CBS are most sensitive to sodium sulfite. Electricity consumption shows lower sensitivity across all technologies.

In summary, optimizing the use of chemicals is an effective strategy to reduce environmental impacts and lowering costs. Future research should focus on improving the efficiency and stability of CBS recovering technology. To enhance the cost-effectiveness of the CBS process and promote its largescale application, further technological optimisation and cost control will be key directions for future development.

CRediT authorship contribution statement

Yu Li: Writing – original draft, Visualization, Software, Methodology, Formal analysis, Data curation. **Jenny Baker:** Writing – review & editing, Methodology, Conceptualization. **Yaxi Fang:** Writing – review & editing. **Haizhou Cao:** Data curation. **Cameron Pleydell-Pearce:** Writing – review & editing. **Trystan Watson:** Writing – review & editing, Conceptualization. **Sha Chen:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Conceptualization. **Guangling Zhao:** Writing – review & editing, Writing – original draft, Software, Methodology, Formal analysis.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Haizhou Cao is currently employed by Zhejiang Huijin Environmental Protection Technology Co., LTD. The other authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.enceco.2025.01.005.

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