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Optimizing Heavy Metal Reduction in Chemical Industry Waste: A Comprehensive Response Surface Methodology Approach for Enhanced Environmental Sustainability

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ABSTRACT. This study investigates the optimization of heavy metal reduction in solid waste from the chemical industry using Response Surface Methodology (RSM). The Box-Behnken design was employed to optimize the extraction parameters for removing Co, Cu, Pb, and Cr using NaCl as a solvent with Ethylenediaminetetraacetic acid (EDTA) as a chelating agent. The effects of EDTA: Fly Ash ratio, mixing duration, and extraction temperature were evaluated using induced coupled plasma (ICP) and Scanning Electron Microscope and Energy-Dispersive X-ray (SEM/EDS) analysis. NaCl proved effective in reducing Co, Cr, and Pb levels. Optimal conditions for NaCl treatment were identified as an EDTA:Fly Ash ratio of 2:1, 3 hours of mixing time, and an extraction temperature of 52.3 °C, resulting in a 92.3% total metal reduction. ANOVA results confirmed the statistical significance of the model, with high R^2 values (0.932 – 0.991) for all metals. The EDTA:Fly Ash ratio and its interaction with mixing duration were found to be the most influential factors in the process. The study demonstrates the effectiveness of RSM in optimizing heavy metal reduction processes and provides insights for improving waste management practices in the chemical industry. The findings highlight the importance of process parameter optimization in enhancing the efficiency of heavy metal removal from industrial solid waste.

INTRODUCTION

The chemical industry, a cornerstone of global economic development, is a major contributor to solid waste generation, presenting substantial challenges to societal well-being and environmental sustainability. This waste includes hazardous and non-hazardous materials like toxic chemicals, heavy metals, sludge, and manufacturing by-products (Abdel-Shafy and Mansour, 2018). Improper management can lead to soil, water, and air contamination, impacting ecosystems, biodiversity, and human health. Inadequate disposal or treatment can cause long-term environmental degradation, affecting nearby communities and beyond (Mor and Ravindra, 2023). Effective management strategies and regulatory frameworks are thus crucial for mitigating these impacts and promoting sustainable practices in the chemical industry.

Combining incineration with physical-chemical treatment effectively manages solid waste and reduces heavy metal content in ash residues. Post-incineration ash contains concentrated heavy metals, posing environmental risks if untreated (Anicetus, 2014). Solvent extraction using NaCl forms complexes with heavy metal ions, facilitating their separation through ion exchange (Chen *et al*., 2022). Ethylenediaminetetraacetic acid, disodium salt dihydrate (EDTA) further stabilizes heavy metals in solution, preventing reactivity or reprecipitation (Ralston *et al*., 2007). This combined approach significantly reduces heavy metal concentrations in ash waste, enabling safer disposal or potential reuse.

Various factors, such as the ratio of ash waste to solvent, mixing time, and extraction temperature, significantly influence the pH and solubility of heavy metals in extraction processes (Tang and Steenari, 2016; Tang *et al*., 2019; Pandey and Bhattacharya, 2019). A higher ash-to-solvent ratio can alter solution acidity or

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alkalinity (Zhang *et al*., 2021). Longer mixing time enhances contact between solvent and ash, facilitating heavy metal dissolution and pH equilibrium (Tang *et al*., 2021). Temperature affects heavy metal solubility by altering chemical kinetics and solvation energy, impacting pH based on solvent behavior (Chen *et al*., 2022).

Solvent extraction and collectors have been explored to enhance heavy metal reduction in waste streams. Rattanasak and Chindaprasirt (2009) optimized Al_2O_3 and SiO_2 extraction from fly ash using 10 M NaOH with a 1:1 sodium silicate to NaOH ratio. Sembiring *et al*. (2020) achieved a 50 – 85% reduction in transition metals from boiler ash using EDTA and organic solvents. However, many studies lack optimization methods like response surface methodology (RSM) to establish robust models correlating independent variables (solvent type, mass ratio, pH, temperature, and extraction time) with dependent variables (heavy metal removal efficiency). RSM enables systematic exploration and optimization of multiple variables simultaneously, improving efficiency in achieving desired

RSM optimizes processes using Box-Behnken and Central Composite designs (BBD and CCD). BBD requires fewer runs, which is ideal for reducing experiments in induced coupled plasma (ICP) and Scanning Electron Microscope and Energy-Dispersive X-ray (SEM/EDS) analyses (Goren and Kobya, 2021). CCD better explores the entire factor space, handling non-linear responses effectively but requiring more runs (Yeten *et al*., 2005; Myers *et al*., 2016). The choice depends on experimental goals and response surface characteristics. This study examines how the ash waste to EDTA ratio, extraction time, and temperature impact heavy metal removal efficiency.

Optimizing heavy metal removal processes is crucial for improving life cycle analysis (LCA) and reducing environmental emissions. By employing RSM, researchers can develop models that accurately predict removal efficiencies, leading to more efficient and cost-effective treatments (Ugwu *et al*., 2022; Buenaño *et al*., 2024). This optimization minimizes waste disposal impact, enables safe reuse of treated materials, and reduces energy and resource inputs (Aziz *et al*., 2023). Consequently, it prevents toxic element leaching, reduces landfilling needs, and minimizes air pollution from contaminated ash dust, resulting in better LCA outcomes and overall reduced environmental impact (Das *et al*., 2023).

RESEARCH METHODS

This study used the following equipment: glassware, laboratory shaker, hot plate, funnel, filter paper, centrifuge, petri dish, oven, ICP equipment, and SEM/EDS equipment. The materials used included coal ash waste from PT. Riau Andalan Pulp and Paper (RAPP), deionized water from AMIDIS, sodium chloride (NaCl), hydrogen chloride (HCl) from HIMEDIA, and ethylenediaminetetraacetic acid, disodium salt dihydrate (EDTA) from Merck Titriplex. The research procedures are shown in Figure 1 and explained in detail, starting from the subsection of 'Extraction of Heavy Metals from Ash Waste' to the Subsection of Data Analysis, to perform RSM modelling to achieve the optimal process.

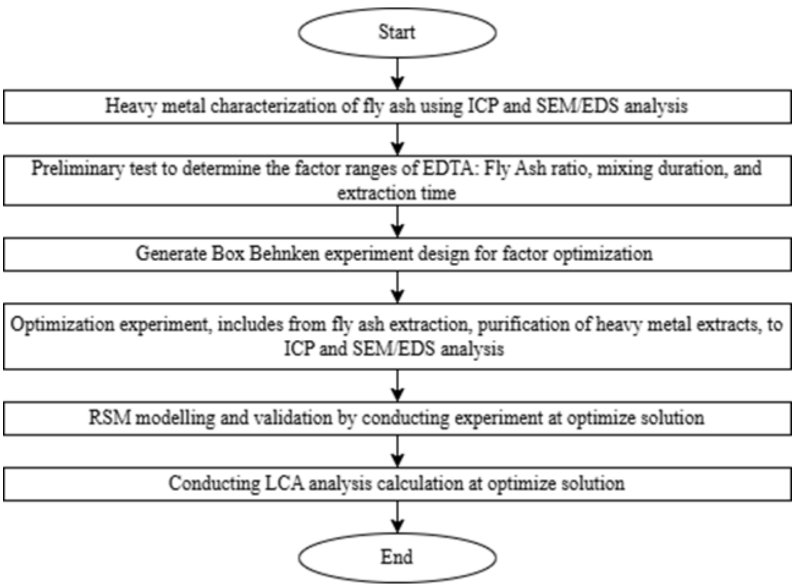

Figure 1. Research scheme diagram.

Extraction of Heavy Metals from Ash Waste

The procedure for extracting heavy metals from ash waste, as described by Sembiring *et al*. (2020), involves several steps. First, ash waste was dried at 100 °C for 2 – 3 hours to eliminate moisture, then sieved using a 400-mesh sieve for particle size uniformity. Some samples were set aside as blanks to determine initial heavy metal concentrations. Portions of 50 grams of ash waste were soaked in 500 mL of 1 M NaCl for 48 hours, with EDTA-to-ash waste ratios by mass of 1:1, 3:2, and 2:1 (w/w) (e.g., 50 g EDTA: 50 g ash waste for 1:1, 75 g EDTA: 50 g ash waste for 3:2, and 100 g EDTA: 50 g ash waste for 2:1). This mixture was shaken for 2 hours andheated up to 70 °C for 45 minutes. All samples were then shaken for either 1, 2, or 3 hours, followed by a 48-hour resting period.

Separation and Purification of Heavy Metal Extracts

The procedure for separating and purifying heavy metal extracts from ash waste, as detailed by Sočo and Kalembkiewicz (2007), involves the following steps. After allowing the samples to stand for 24 hours, three layers were formed: sludge at the bottom, solvent in the middle, and the extraction product on top. The extraction product was separated and oven-dried for 3 hours. The remaining layers are centrifuged at 2500 rpm for 20 minutes, resulting in sludge, which is then dried in a petri dish at 100 °C for 6 hours to evaporate the solvent. The dried sludge and extraction products are then mixed and analyzed to determine the remaining heavy metal content.

Induced Coupled Plasma (ICP) analysis

The ICP-OES analysis was conducted, as described by Bauer and Limbeck (2018). Samples were analyzed using the radial view iCAP 6500 ICP-OES spectrometers with the ETV 4000A system for sample introduction and evaporation, both utilizing PTFE tubes. A 20 µL sample was pipetted into a standard graphite boat, dried with an IR lamp to evaporate the solvent, and placed in the ETV 4000A furnace. The temperature was ramped to 400 °C for 60 seconds, held at 400 °C for 10 seconds, rapidly increased to 2150 °C, and held for 20 seconds, with Freon R12 gas modifier and argon gas flow. Signals were recorded and processed by Thermo iTEVA software, with peak area transient ETV data points collected every 0.5 seconds. This analysis enables the determination of the remaining metal concentrations in the treated ash waste.

Scanning Electron Microscope and Energy-Dispersive X-ray (SEM/EDS) Analysis

The procedure for SEM/EDS analysis, as detailed by Ni'matuzahroh et al. (2020), involves testing samples to study the morphological structure of boiler ash extracts before and after extraction treatment. Specimens were coated with gold ions under vacuum using the COXEM SPT-20 Ion Sputter Coater. High-quality images were obtained under high vacuum (approximately 10^{-6} Torr) with the HITACHI FlexSEM 1000 VP-SEM at 15.0 kV, 6.1 mm work distance, and four different magnifications (1000, 2000, 5000, and 10,000). An SE detector labeled on the data bar detected images, and one micrograph at 5000 magnification mapped elements C, O, N, Na, Al, Si, Pb, Mo, Cl, Ca, Cr, Co, and Cu.

Data Analysis

The Box-Behnken design, introduced by George E. P. Box and Donald Behnken in 1960, is a statistical experimental design within response surface methodology (RSM). It models data curvature and pinpoints factor settings for response optimization. Each factor was set at three equally spaced values, typically coded as -1, 0, and +1 (Bagheri *et al*., 2017). This design efficiently fits a quadratic model, including squared terms, factor products, linear terms, and an intercept.

Box-Behnken designs offer advantages over other RSM methods by often needing fewer experimental runs than central composite designs, saving resources. They avoid extreme factor settings, suiting processes with safe operating zones. Combining a two-level factorial design with an incomplete block design, some factors vary while others remain at central values; center points are crucial for precision (Goren and Kobya, 2021). Limitations include the inability to predict responses at design space corners and considering only three levels per factor, which might affect accuracy (Alaoui *et al*., 2015).

Design of Experiments

The Box-Behnken design requires the number of experiments according to Equation 1.

$$N = k^2 + k + cp \quad (1)$$

where k represents the number of factors and cp represents the number of center point replications (Alaoui *et al.*, 2015). In this study, three factors of ash waste to EDTA, mixing duration, and extraction temperature were optimized with one replication, resulting in 15 sets of experiments for each solvent. Each factor was tested at three levels: low (-1), high (+1), and a center point (0), to assess experimental errors (Bagheri *et al.*, 2017). Optimization ranges for the variable factors were determined based on preliminary test results, detailed in Table 1.

Table 1. Optimization factor ranges for NaCl solvent.

Variable factor	Factor	Level		
		-1	**0**	**1**
EDTA: Ash ratio	X	1:1	3:2	2:1
Mixing duration (/h)	Y	1	2	3
Extraction time (°C)	Z	25	47.5	70

To model the relationship between the predicted response and optimization variables using a second-order polynomial regression equation for three factors, the general equation was expressed as shown in Equation 2 (Polat *et al.*, 2019).

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_3^2 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 \quad (2)$$

where x_1, x_2, x_3 are the variable factors (e.g., EDTA: ash ratio, mixing duration, extraction temperature), y is the response variable or output (e.g., percentage reduction of heavy metals Cu, Cr, Co, and Pb), β_0, β_1, β_2, β_3, β_{11}, β_{22}, β_{33}, β_{12}, β_{13}, β_{23} are regression coefficients to be estimated. The Minitab software version 18 was used to generate a second-order polynomial equation for three factorial factors, analyze optimization results using RSM, and perform statistical analysis to determine model significance.

RESULTS AND DISCUSSION

Morphological Structure and Elemental Composition of Ash Waste

The morphological structure and elemental composition of boiler ash undergo significant changes during the extraction process with NaCl and EDTA. Initially, untreated boiler ash typically exhibits a heterogeneous structure with diverse particle sizes and shapes. SEM images would likely reveal irregular, angular particles, spherical cenospheres characteristic of fly ash, agglomerated clusters, and porous structures with rough surfaces (Figure 2a).

After NaCl extraction, the ash particles generally show reduced size due to the dissolution of soluble components, resulting in a more uniform particle distribution (Figure 2b). The porosity increases as soluble salts are removed, and new crystalline structures may form due to NaCl interaction. The subsequent EDTA extraction further alters the morphology, leading to even smaller particle sizes, smoother surfaces due to metal ion chelation, increased porosity, and potential collapse of some particle structures as structural metals are removed such as iron (Fe), aluminium (Al), calcium (Ca), magnesium (Mg), and heavy metals (e.g., lead (Pb), cadmium (Cd), copper (Cu), zinc (Zn)), which are often bound in the ash matrix or adsorbed on the particle surface.

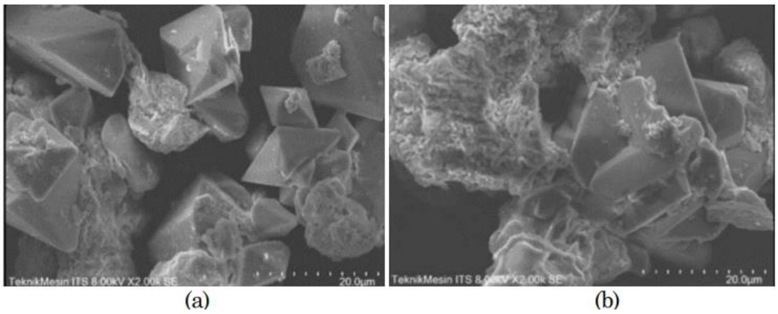

Figure 2. Morphological structure of fly ash (a) before and (b) after treatment.

As revealed by SEM-EDS analysis, the elemental composition also changes significantly throughout the extraction process. Initially, the ash contains high Si, Al, Ca, and Fe concentrations, with trace amounts of Pb, Mo, Cr, Co, and Cu. Oxygen is abundant due to oxide forms, and detectable levels of Na and Cl from soluble salts are present (Rodriguez-Casariego *et al.*, 2018). The Na and Cl concentrations increase after NaCl treatment, while the Ca and other soluble elements decrease. The Si and Al concentrations relatively increase proportionately in the total composition. The EDTA extraction then causes a significant reduction in metal concentrations (Ca, Pb, Cr, Co, Cu), increased C and N signals from EDTA complexes, and a further relative increase in Si and Al concentrations.

Element-specific observations show that Si and Al remain relatively stable throughout the process, forming the ash matrix. Ca undergoes a significant reduction after both NaCl and EDTA treatments. Heavy metals like Pb, Mo, Cr, Co, and Cu show a marked decrease after EDTA extraction due to chelation (Thrivierge *et al.*, 2021). Na increases after NaCl treatment but may decrease following EDTA washing. The Cl initially increases after NaCl treatment but decreases after washing. Oxygen remains relatively stable, possibly slightly increasing due to exposed oxide surfaces (Rodriguez-Casariego *et al.*, 2018). Carbon and nitrogen increase after EDTA treatment due to the presence of the organic chelator (Thrivierge *et al.*, 2021).

Model Prediction and Optimization by RSM Analysis

The predicted response models for Co, Cu, Pb, Cr, and the total reduction percentage using the NaCl and EDTA mixture were represented by Equations 3 – 7. The heavy metals recorded by the ICP analysis in the ash waste predominantly consist of Co, Cu, Pb, and Cr, with the remainder being negligible.

$$\%Red_{Co} = 91.8 - 7.3X - 9.3Y + 0.23Z + 2.47X^2 + 0.74Y^2 - 0.0022Z^2 + 4.43XY + 0.021XZ - 0.018YZ \quad (3)$$

$$\%Red_{Cu} = 94.6 - 67.6X - 7.7Y + 0.42Z + 24\,X^2 - 1.42Y^2 + 0.0045Z^2 + 5.12XY - 0.089XZ + 0.075YZ \quad (4)$$

$$\%Red_{Pb} = 86.7 - 46.2X + 16.9Y - 0.01Z + 17.1X^2 - 2.86Y^2 + 0.0004Z^2 + 1.42XY - 0.06XZ + 0.035Y \quad (5)$$

$$\%Red_{Cr} = 89.8 - 2.8X - 5.6Y - 0.043Z + 3.28X^2 + 0.54Y^2 - 0.00036Z^2 + 1.27XY - 0.02XZ + 0.018YZ \quad (6)$$

$$\%Red_{Total} = 91.3 - 41.6X + 5.25Y - 0.18Z + 15.5X^2 - 1.1Y^2 + 0.002Z^2 + 3.2XY - 0.056XZ + 0.04YZ \quad (7)$$

The highest percentage reductions of heavy metals Co, Cu, Pb, and total were achieved at ratios by mass of 2:1 (w/w), durations of 3 hours, and temperatures of 47.5 °C, amounting to 98.1%, 85.9%, 93.2%, and 90.2%, respectively. Meanwhile, the highest reduction of Cr was attained at a ratio of 2:1, duration of 1 hour, and temperature of 47.5 °C, reaching 93.1%. The Analysis of Variance (ANOVA) results examine the adequacy and significance of the model coefficients for the optimized reductions of Co, Cr, Pb, and total metals are summarized in Table 2.

Table 2. ANOVA and optimization of the solution for NaCl solvent.

ANOVA parameter	**ANOVA result for the heavy metal of**				
	Co	**Cu**	**Pb**	**Cr**	**Total**
F-value (p<0,05)	0.019	0.006	0.018	0.000	0.008
Lack of fit (p>0,05)	0.256	0.638	0.503	0.226	0.606
R^2 coefficient	0.932	0.960	0.933	0.991	0.954
Significant model term	X, XY	X, Y, X^2	X, Y, X^2	X, Y, X^2 , XY	X, Y, X^2
Optimized response at the factor value of	**Optimization fit**				
	%Co fit	%Cu fit	%Pb fit	%Cr fit	%Total fit
X = 2:1 Y = 3 h^{-1} Z = 52.3 °C	97.63	89.20	96.09	92.49	92.25

The F-value assesses the overall significance of the model, with a low value and a significant p-value ($p < 0.05$) indicating that the model terms (factors and interactions) explain a substantial portion of the variance in all response variables (percentage reduction of heavy metals) (Ohale *et al.*, 2017). The lack of fit assesses the model's adequacy in fitting the data, showing non-significant results across all response variables, confirming the model's suitability (Mai *et al.*, 2021). The R^2 (coefficient of determination) measures how well the model explains variability, with high values indicating a strong fit and capturing significant variability in responses (Bennett *et al.*, 2013). Significant model terms highlight influential factors and interactions in optimizing heavy metal reduction (Bayuo *et al.*, 2022). For Co, these include X (main effect of ash ratio), XY (interaction between ash ratio and mixing duration), and similar terms apply to Cr, Cu, Pb, and Total. At the optimal conditions (EDTA: Fly Ash ratio 2:1, mixing duration 3 hours, extraction temperature 52.3 °C), the predicted response values are: total metal reduction 92.3%, Co reduction 97.6%, Cr reduction 92.5%, Cu reduction 89.2%, and Pb reduction 96.1%.

The 3D surface plot showing the relationship among the variables' ratio-temperature, duration-temperature, and ratio-duration for the response variable %total metal reduction is depicted in Figure 3. The relationship between EDTA:Ash ratio (X) and mixing duration (Y) shows a curved surface with the highest total reduction (around 90%) achieved at higher ratios (2:1) and longer durations (3 hours). The curvature indicates a quadratic effect, where the metal reduction initially decreases at lower ratios before increasing significantly at higher ratios.

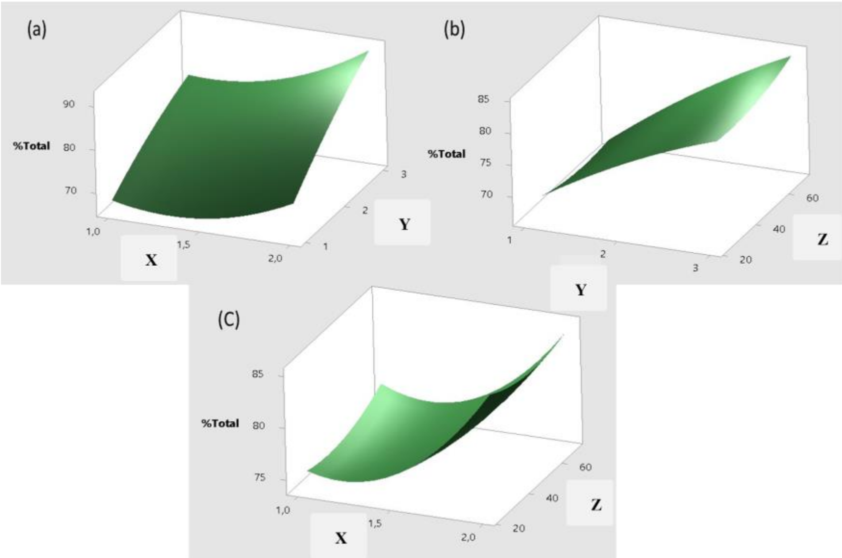

Figure 3. The 3D surface plot for NaCl solvent, (a) EDTA: ash ratio(X) and mixing duration(Y), (b) mixing duration(Y) and extraction time(Z), and (c) EDTA: ash ratio(X) and extraction time(Y).

The surface plot demonstrates a linear positive relationship between mixing duration (Y) and temperature (Z). The total metal reduction improves as both duration and temperature increase, reaching maximum values (around 85%) at 3 hours duration and higher temperatures. The relatively flat surface suggests a less pronounced interaction between these variables. However, this plot shows a complex curved relationship between EDTA:Ash ratio (X) and temperature (Z). The surface exhibits a saddle-like shape, indicating that optimal metal reduction (around 85%) is achieved at a higher ratio (2:1) and moderate temperatures (around 47.5 °C). The curvature suggests that a very low ratio and a very high temperature may be less effective for metal reduction.

Life Cycle Assessment (LCA) Analysis of the NaCl/EDTA-Treated Ash System

Life cycle assessment (LCA) is a method used to measure technological, economic, and environmental impacts (Sari *et al.*, 2012). Based on the optimum results from the RSM analysis, we conduct an LCA analysis for the NaCl-EDTA extraction process of heavy metals from ash waste. The LCA analysis was performed in openLCA version 1.11.

Energy Consumption

The total energy consumption is estimated at 125 kJ/kg of ash waste treated. This included 85 kJ/kg for drying, 10 kJ/kg for mixing and shaking, and 30 kJ/kg for the heating process. This relatively low energy requirement, especially compared to high-temperature treatment methods, suggests reduced greenhouse gas emissions and a lower carbon footprint for the process.

Chemical Usage

The process utilizes 160 g/kg of NaCl and 20 g/kg of EDTA per kg of ash waste. While chemical usage is significant, the high efficiency of metal removal justifies its use from an environmental perspective.

Water *Consumption*

The process requires approximately 10 L of water per kg of ash waste treated. This moderate water usage indicates a need for efficient water management strategies in the overall process.

The high removal efficiencies achieved through this process significantly contribute to sustainability and environmental protection in multiple ways. By substantially reducing the heavy metal content in treated ash waste, the risk of toxic elements leaching into soil and groundwater is minimized, thereby safeguarding ecosystems and human health (Kazi *et al*., 2023). Furthermore, reducing hazardous waste volume decreases the burden on landfills and minimizes long-term environmental risks (Velusamy *et al*., 2021).

Compared to high-temperature treatments, the process's low energy requirement of 125 kJ/kg reduces overall energy consumption and emissions (Velusamy *et al*., 2021). It aligns with circular economic principles by transforming waste into a potential resource stream, reducing demand for virgin materials (Hessel *et al*., 2021). The treated ash becomes safer for disposal or reuse in applications like construction materials, enhancing waste management practices (Kazi *et al*., 2023). Moreover, the significant removal of heavy metals lowers the ash waste's ecotoxicity potential, minimizing potential harm to ecosystems in case of environmental release. For instance, greenhouse gas emissions were lowered to 79%, and waste sent to landfills was reduced by up to 90% throughout this circular economy model.

CONCLUSION

In conclusion, this study demonstrates the effectiveness of the NaCl-EDTA extraction process for heavy metal removal from ash waste, achieving impressive reduction rates of up to 97.63% for Co, 96.09% for Pb, 92.49% for Cr, and 89.20% for Cu, with a total metal removal of 92.25%. The optimized process conditions (EDTA: Ash ratio 2:1, mixing duration 3 hours, extraction temperature 52.3 °C) and low energy requirement of 125 kJ/kg highlight its potential as an environmentally friendly and efficient treatment method. Future research should focus on scaling up the process, exploring the recovery and reuse of extracted metals, and investigating the potential applications of the treated ash in construction materials to enhance its sustainability and economic viability further.

CONFLICT OF INTEREST

The corresponding author, on behalf of all authors, declares that there are no financial or personal relationships with other individuals or organizations that could inappropriately influence or bias the work presented in this article.

AUTHOR CONTRIBUTION

MPS: Conceptualization, Methodology, and Manuscript Drafting; DH, CC, and SRM: Supervision, Manuscript Review, and Editing; MPET: Software, Data Analysis, Manuscript Review, and Editing.

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