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Research Paper

Assessment And Characterization of Individual and Combined Chelant/Surfactant for Remediation of Mixed Toxic Metals and Radionuclides in Contaminated Tin Mine Tailings

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Abstract

The study assessed the levels, public health risks associated with mixed contaminants specifically toxic metals and radionuclide present in tin mine tailing dumpsites across the Jos Plateau State, Nigeria. Characterization of these contaminants was performed using Atomic Absorption Spectrophotometry and Gamma Photometry. Bench-scale washing experiments were conducted to evaluate the effectiveness of individual chelants and surfactants, as well as their binary blends, in removing the contaminants from tailings. Tailings samples were collected from Zawan A, Kuru A, Sharubutu, Rim dumpsites, and nearby agricultural farmlands located in Jos South and Riyom Local Government Areas. These samples were analyzed physiochemically to determine the concentrations of toxic metals (Cd, Cr, Pb) and radionuclides (U, Th, K). Batch and column washing experiments were conducted to test the efficiency of individual washingagentssodium dodecyl sulfate (SDS) and ethylene diamine tetra acetic acid (EDTA)and their binary blends in removing mixed contaminants. The experimental conditions included varying initial concentrations (8–64 mM), pH (2–10), soil-to-liquid ratios (1:1–1:40 g/mL), washing durations (25–200 minutes), and weight percentages of the components in the blend. Before washing experiments, the chelant's critical stability constants (CSC) were determined: EDTA with Cd = 16.5, Pb = 18, Cr = 23.8, U = 22.3, Th = 23.3, and K = 1. The critical micelle concentration (CMC) of SDS was found to be 8 mM. Metal concentrations (mg/kg) were: Cd (1.81–2.98), Pb (11.45–50.53), Cr (1.21–6.50); radionuclide concentrations (Bq/kg) were: U (3.92–30.98), Th (4.71–159.02), K (0.001–0.47). Principal Component Analysis (PCA) suggested a correlation between these contaminants and anthropogenic sources. Batch washing results showed that contaminant removal efficiency increased with higher initial concentrations of chelant and surfactant. SDS removal rates: Cd (43.73–74.00%), Pb (19.67–74.37%), Cr (40.60–64.16%), U (32.89–82.94%), Th (12.79–79.59%). EDTA removal rates: Cd (50.15–78%), Pb (24.05–77.84%), Cr (45.36–66.92%), U (29.77–76.43%), Th (9.54–74.49%). As pH increased (2–10), removal efficiency decreased. SDS: Cd (74.31–28.11%), Pb (80.87–16.48%), Cr (77.69–27.82%), U (85.43–34.22%). EDTA: Cd (79.82–33.64%), Pb (87.22–10.92%), Cr (80.54–32.83%), U (82.73–22.62%), Th (77.57–12.79%). Lowering the solid/liquid ratio improved removal rates. SDS: Cd (41.59–77.06%), Pb (28.41–73.42%), Cr (40.85–69.92%), U (40.31–88.56%), Th (10–82.06%). EDTA: Cd (46.79–78.29%), Pb (37.62–79.96%), Cr (48.12–74.68%), U (33.10–82.94%), Th (8.93–78.16%). Increased washing time also enhanced removal. SDS: Cd (30.89–83.18%), Pb (24.18–88.76%), Cr (25.81–71.68%), U (17.00–82.68%), Th (9.55–80.21%). EDTA: Cd (34.25–86.85%), Cr (30.32–73.43%), U (24.52–85.86%), Th (10.90–82.21%). Column experiments showed decreasing removal rates over time. SDS: Cd (86.85–38.23%), Pb (89.84–39.89%), Cr (80.45–46.87%), U (82.73–17.06%), Th (95.09–20.17%). EDTA: Cd (89.29–53.21%), Pb (92.29–48.39%), Cr (85.46–49.12%), U (77.54–13.98%), Th (91.76–18.86%). Binary blend experiments (10–90% w/w) showed varying removal: Cd (96.33–41.59%), Pb (95.36–10.86%), Cr (95.74–28.57%), U (94.70–13.88%), Th (987–13.55%). Column results for blends showed leachate concentrations: Cd (1.73–0.44%), Pb (1.64–0.63%), Cr (1.31–2.44%), U (0.79–0.52%), The study concludes with recommended preventive and remedial actions, as well as areas for future research. Th (0.60–0.20%). Statistical analysis confirmed that individual and blended chelant-surfactant systems were effective in contaminant removal. The synergistic effect of the blends yielded better results than the individual agents.

Keywords: *Tin Tailings, Toxic Metals, Radionuclides, Chelants, Surfactants*

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I. Introduction

1.1 Background of the Study

Due to discharge of toxic metals and radionuclides into the nearby ecosystems, mining operations, especially tin mining, have significantly contributed to environmental contamination. The left over after tin ore is extracted, known as tin mine tailings, frequently include high concentrations of these pollutants, which can be harmful to the environment and public health [22].

Waste fractions of an ore or mineral body that are thrown away during mining operations without being treated are known as Tin mining tailings. Both magnetic (iron ore, columbite) and non-magnetic (cassiterite, monozite, silica, and vast amounts of zircon sand) minerals can be found in tin tailings [1]. They are employed in producing refractory goods, fertilizer, animal feed, road construction, and tailing storage facility backfill. Nigeria was one of the top producers of tin in the world in the 1960s, after the 20th century, production had reduced. Nonetheless, mining operations continue in some regions. Tin tailings, a by-product of the ore, are produced during tin mining in addition to the main aim, tin.

According to [2], the amount of tailings generated varied between 20 and 50 percent for other minerals and 90 to 98 percent for copper ores.

The inhabit in the host rocks, radionuclides like uranium and thorium isotopes can be found in high amounts in tin mine tailings. These radionuclides can release ionizing radiation, which is known to have harmful effects on human health, which includes risk of cancer [20].

Similarly, toxic metals like lead, cadmium, arsenic, and mercury, which are frequently present in tin mine tailings, are recognized as hazardous to both humans and ecosystems. These metals have the potential to bioaccumulate in the food chain, resulting in long-term health consequences such as neurological disorders, kidney damage, and developmental abnormalities [7].

Considering the possible dangers linked to the presence of radionuclides and toxic metals in tin mine tailings, effective remediation strategies are necessary to reduce the exposure of these contaminants to humans and the environment. Chelants and surfactants have been extensively researched as potential remediation agents because of their capacity to improve the mobilization and removal of radionuclides and heavy metals from contaminated soils and sediments [3].

Chelants are organic substances that create stable complexes with metal ions through chelation, effectively diminishing their bioavailability and mobility in the environment [11]. They can attach to heavy metals and radionuclides, forming soluble complexes that can be more readily eliminated through soil washing or leaching methods [5]. Commonly utilized chelants include ethylenediaminetetraacetic acid (EDTA), citric acid, and diethylenetriaminepentaacetic acid (DTPA).

Surfactants, however, are amphiphilic substances that can interact with both water and organic matter. They can enhance the solubility of hydrophobic contaminants such as organic pollutants, radionuclides, and heavy metals by creating micelles or complexes [12]. Surfactants can facilitate the mobilization and removal of contaminants from the soil matrix through techniques like soil washing or enhanced desorption [9]. Commonly used surfactants consist of sodium dodecyl sulfate (SDS), Tween 80, and Triton X-100.

The joint introduction of chelating agents and surfactants has demonstrated potential in the cleanup of radionuclides and toxic metals from tin mining waste. By utilizing these substances, the pollutants can be activated and later extracted, thereby lowering their bioavailability and possible threats to public health [20]

Research Methodology

Tailings and farmland soils were sampled from Zawan A, Kuru A, Sharubutu, and Rim in Jos south and Riyom Local Government Areas of Plateau state, Nigeria. Analytical methods included atomic absorption spectrophotometry and gamma photometry. Soil washing experiments were performed at varying pH, concentrations, washing times, and soil/liquid ratios. Blended washing agents were tested for synergistic interactions.

Plateau state is located in the middle belt of Nigeria's with an area of 30.91 km (11936 sq mi), the state has an estimated population of about three million people. It is located between latitude 9051'30"N to 1002'00"N and longitude 8048'00"E to 9051'00E. The name Plateau state emerged due to its mountainous topography with captivating rock formations. The mountains attitude ranges from around 1,200 meter (about 400 feet) to a peak of 1,829 meters above sea. The predominant occupation of the people here is mining and subsistence farming, 700 g of each of the sample was taken.

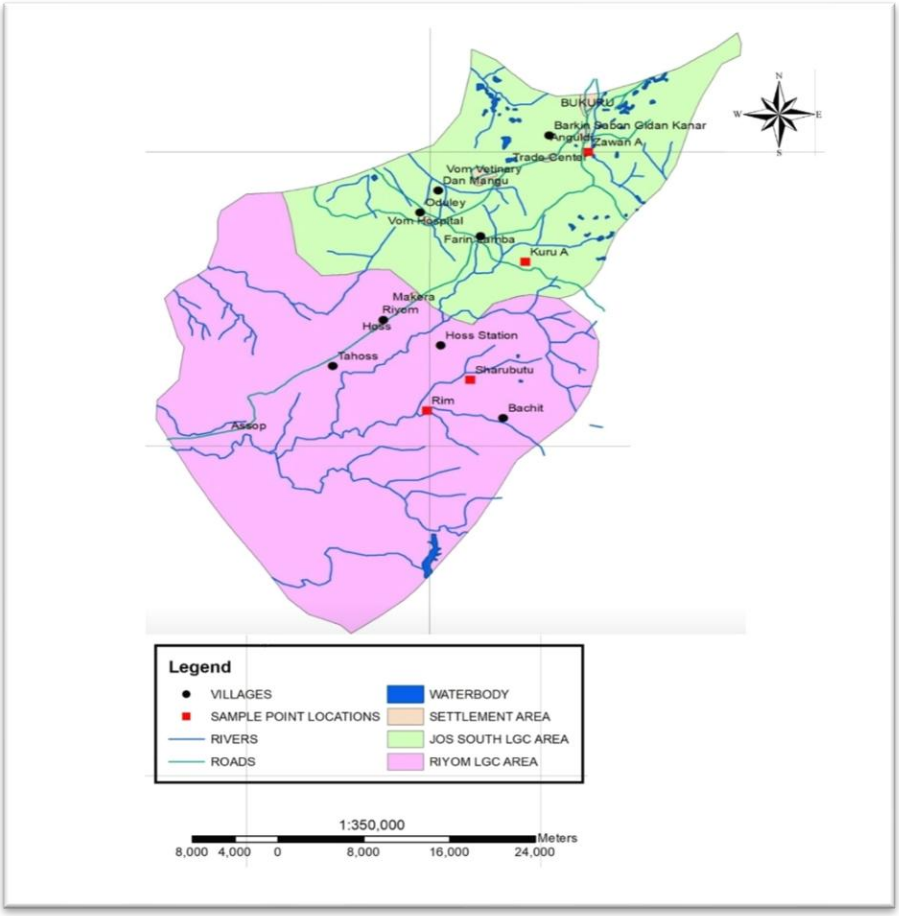
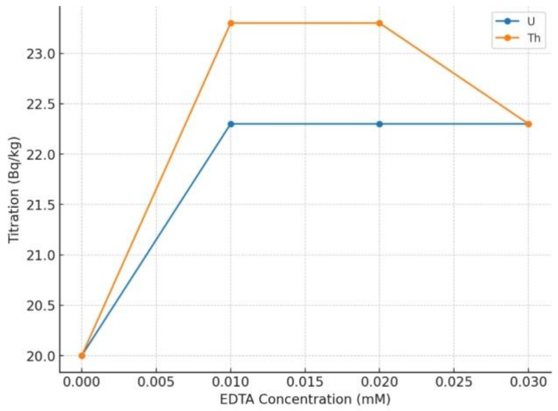


Figure 1: Map Showing the Location of the Study Area
Source: Cartographic, GIS and Remote Sensing Studio Department of Geology University of Jos, 2023.

Figure 1 showed the where the tailing samples were taken from Jos south and Riyom L.G.A. of Plateau State. The Geographical Positioning System (GPS) was used to locate the sampling area to ensure consistency. Each sample was taken from the mining site in Zawan A (ZS1) and Kuru A (KS2) of Jos-South Local Government Area, Sharubutu (SS3) and Rim (RS4) and adjacent farmland areas as control of Riyom Local Government Area of Plateau State, Nigeria.

Washing agents
Chelant: Ethylene diamine tetraacetic acid (EDTA) and Surfactant: Sodium dodecyl sulfate (SDS). All reagents were analytical grades and were received without further purification.
The efficacy of the washing agents (EDTA) and SDS was carried out before the washing experiment
Determination of Chelant's critical Stability Constant (CSC)
The contaminant removal mode was conducted with the help of chelant (EDTA). It is important to determine the CSC of the chelant before washing procedure so as to be guided on the washing solution to be prepared. Using conductivity principle, Figure 2 shows a plot of conductivity against concentration of the contaminant gives the CSC (Cd = 16.5, Pb = 18, Cr = 23.8 U = 22.3, Th = 23.3, K = 1) at 25^0c.The value compare favourably those reported in literature for EDTA [11]; [19]. Figure 2 at concentration (conductivity) due to availability of stability and after CSC conductivity was increasing with increasing concentration.



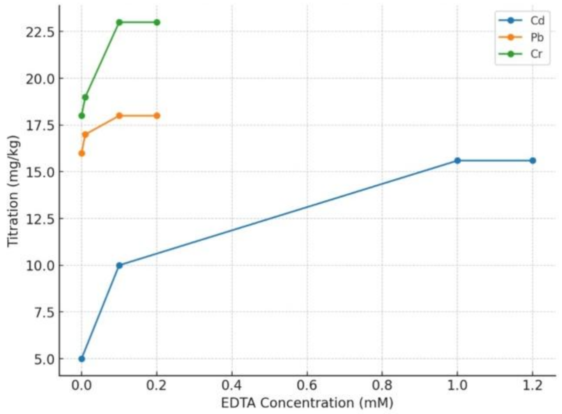


Figure 2a: Chelant (EDTA) CSC Determination for Radionuclides Figure 2b: Chelant (EDTA) CSC Determination for Toxic Metals

Determination of Surfactant Critical Micelle Concentration(CMC)

The contaminant removal experiment was conducted using an anionic surfactant, sodium dodecyl sulfate (SDS). It was essential to determine the critical micelle concentration (CMC) of the surfactant before proceeding with the soil washing process in order to accurately prepare the washing solution.

Following the dilution principle, Figure 3 presents a plot of experimental data showing the relationship between surfactant concentration and conductivity for the determination of the CMC of SDS. The plot displays a linear trend with a change in slope, and the concentration at which this change occurs (0.008 mol/dm³ or 8 mM) is identified as the CMC of SDS at 25°C.

This value aligns well with those reported in the literature, where CMC values for SDS range between 8.0 mM and 8.6 mM [18]. In Figure 3, it is observed that below the CMC, conductivity increases with concentration due to the availability of free surfactant molecules. However, after the CMC is reached, conductivity increases at a slower rate because micelle formation dominates, and further dilution leads to a sharp decrease in conductivity.

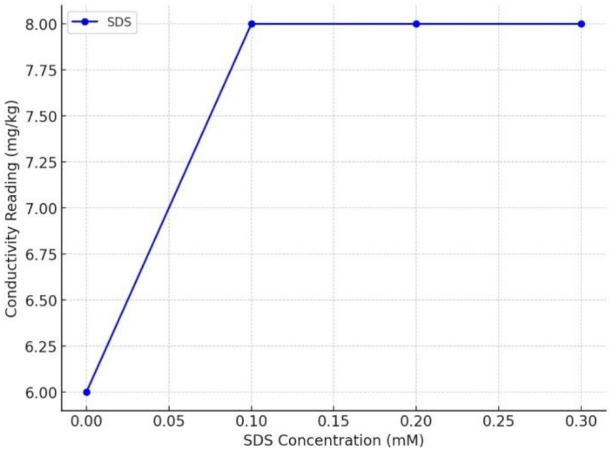


Figure 3: Determination of Critical Micelle Concentration (CMC) of SDS

Batch Mode

The batch washing experiment was conducted following the method outlined by [13]. The washing solution for the single dose surfactant (SDS) and single dose chelant (EDTA) was prepared using 8 mM, 16 mM, 32 mM, and 64mM, with variations from 3, 6, 9, and 12. The soil/liquid ratio varied at 1:1, 1:10, 1:20, 1:30 and 1:40. The washing time was modified from 25min, 50 min, 75min, 100 min, 125min, 150 min, 175 min, to 200 min. Mine tailings (3 g) were equilibrated with 20 mL of EDTA (3 mmol) and a surfactant (30 g) SDS in a 50 mL centrifuge tube for the washing agent experiment. Equilibration was accomplished through sonication of the soil suspension for 10 min using an ultrasonic homogenizer (XL 2020 Sonic Dismembrator, Misonix, Farmingdale, NY). An extended horn of 25 cm in length and 1. 2 cm in diameter, tuned to a frequency of 20 kHz, delivered ultrasonic energy (240 W) in a pulsed mode with a fixed vibration amplitude setting of 6. The extraction comprised pulsed bursts of power delivered for 3 s followed by a 2 s resting phase to the soil suspension held in a plastic tube placed in an ice bath. Subsequently, samples will be centrifuged at 4000 rpm (10 min) to retrieve the supernatant fraction. Aliquots of the supernatants were preserved for assessing the concentration of radionuclides and toxic metals in this fraction.

The percentage of each metal removed was computed using an equation presented by [1], as follows:

$$\text{Contaminant removal } \% = \frac{\text{contaminant mass in supernant (CLVL)}}{\text{initial contaminant in soil (CsMs)}} \text{ x } 100\% \qquad (1)$$

Where:
CL: Concentration of contaminant in supernatant (mg/L)
Cs: Concentration of contaminant in soil (mg/kg)
VL: volume of supernatant (L)
Ms: dry mass (kg)

II. Results and Discussion

The concentration of Pb decrease from 31.68±0.02mg/kg to 1.84±0.001mg/kg implying a percentage recovery of 94%, the concentration of Cd decreases from 3.27±0.02mg/kg to 0.01±0.002mg/kg implying a percentage recovery of 99.7% and the concentration of Cr decreases from 3.99±0.02mg/kg to 0.02±0.002mg/kg implying a percentage recovery of 99.5

Metals	Concentration of Sample before Washing experiment	Concentration of Sample after washing experiment	Recovery%
Pb	31.68±0.02	1.84±0.001	94
Cd	3.27±0.02	0.01±0.002	99.7
Cr	3.99±0.02	0.02±0.002	99.5

Table 1: Mean Concentration (mg/kg) of Toxic Metals in Mine Tailings Sample before and after Washing with Surfactant (SDS) and Chelant (EDTA)

The concentration of U decrease from 18.88±1.48mg/kg to 5.00±3.32mg/kg implying a percentage recovery of 74%, the concentration of Th decreases from 75.70±4.19mg/kg to 9.10±31.10mg/kg implying a percentage recovery of 88% and the concentration of Cr decreases from 0.47± 0.03mg/kg to 0.11± 0.01mg/kg implying a percentage recovery of 77.

Radionuclides	Concentration of Sample before Washing experiment	Concentration of Sample after washing experiment	Recovery%
U	18.88± 1.48	5.00±3.32	74
Th	75.70± 4.19	9.10±31.10	88
K	0.47± 0.03	0.11± 0.01	77

Table 2: Mean activity Concentration (Bq/Kg) of Radionuclides in Tin Mine Tailing before and after Washing with Surfactant (SDS) and Chelant (EDTA)

Analysis of Toxic Metals (TMs)

Lead (Pb), cadmium (Cd), and chromium (Cr) concentrations varied across the sampling sites, with Pb concentrations ranging from 11.45 ± 0.01 mg/kg to 50.53 ± 0.03 mg/kg. The highest Pb concentration was recorded at Kuru A, while Rim had the lowest. Cadmium and chromium concentrations followed similar patterns. These metals likely stem from mining activities, improper waste disposal, and agricultural practices. Pb concentrations observed at Zawan A were lower than those reported by [3], but higher than those in [6].

Radionuclide Analysis

Radionuclide activity concentrations in the tin mine tailings were assessed by measuring levels of ^{238}U, ^{232}Th, and ^{40}K. The activity concentrations at 100 meters from the mining dumpsites were generally higher than those at control sites, potentially due to variations in altitude or temperature. Notably, ^{232}Th contributed the most to specific activities in all the samples analyzed. The highest concentration of ^{238}U (30.98 ± 2.41 Bq/kg) was found at the Sharubutu site, ^{232}Th (159 ± 8.82 Bq/kg) at the Kuru site, and 40K (1.88 ± 0.10 Bq/kg) at the Rim site. Activity concentrations in the mine tailings were generally higher than those at agricultural farmlands, which is likely due to the mining operations generating dust that may increase exposure to Naturally Occurring Radioactive Materials (NORM), including radon gas [17].

The activity concentrations of ^{238}U, 232Th, and ^{40}K in the study area were lower than the global averages of33 Bq/Kg, 36 Bq/kg, and 474 Bq/kg, respectively [21]. The absorbed dose rates (D) and annual effective doses (AED) were calculated for the various sites, with values at the tin mine tailings generally lower than global averages. For instance, the mean absorbed dose rate for the tin mining sites was 58.18 nGy/h, and the mean annual effective dose was 71.38 Sv/y, both of which are lower than the global average dose rate of 60 nGy/h [21]. The Radium equivalent (Ra eq) for the study areas was below the recommended maximum permissible value of 370 Bq/kg [21], and the Hazard Indices (Hex and Hin) were also within acceptable limits [10].

Batch Washing Mode of Tin Mine Tailing

Effect of Initial Surfactant and Chelant Concentration

The impact of varying concentrations of EDTA and SDS on the removal of contaminants from tin tailings was examined, as shown in Figure 4. As the concentration of the chelant EDTA increased, the removal efficiency for contaminants also rose. Specifically, for Cd, the removal at 8 mM, 16 mM, 32 mM, and 64 mM was 1.64 mg/kg (50.15%), 2.02 mg/kg (61.77%), 2.37 mg/kg (72.48%), and 2.56 mg/kg (78.29%), respectively. A similar trend was observed for Pb, where the removal at 8 mM, 16 mM, 32 mM, and 64 mM were 7.62 mg/kg (24.05%), 13.89 mg/kg (43.84%), 20.11 mg/kg (63.48%), and 24.66 mg/kg (77.84%), respectively. This suggests that higher chelant concentrations facilitated the removal of more contaminants, with the order of removal efficiency for the contaminants being Cd > Cr > Pb > Th > U at all tested EDTA concentrations.

The difference in structure between the chelant (EDTA) and surfactant (SDS) may have influenced their efficiency. EDTA, with a critical saturation concentration (CSC) of 0.25 mM, is more likely to form a complex and precipitate contaminants, whereas SDS, with a critical micelle concentration (CMC) of 0.012 mM, forms micelles that trap contaminants. Previous studies suggest that once micelles are formed at higher concentrations of surfactant, contaminants trapped within these micelles are not present in the supernatant, and hence not detectable. Despite this, the percentage of contaminant removal increased as both chelant and surfactant concentrations rose, aligning with trends observed in other studies [13][23].

At surfactant concentrations above the CMC, SDS enhanced the solubility of contaminants by partitioning them into micelles, which reduced surface tension and improved contaminant solubilization. Below the CMC, SDS remained as soluble macromolecules and was less effective at interacting with contaminants. The trend of increased contaminant removal with higher SDS concentrations is consistent with findings from [23], who used nonionic surfactants in a similar experiment.

A t-test applied to the percentage removal of contaminants indicated significant negative variance for Cd ($p = 2.20E-07$) and Pb ($p = 8.48E-04$) as the initial surfactant concentration increased. However, for Pb, the difference in variance was not significant ($p > 0.05$).

Figure 4 illustrates the removal of mixed contaminants from the tailing sample at varying SDS concentrations. At 8 mM, 1.43 mg/kg of Cd (43.73%) was removed, and at 16 mM, 2.34 mg/kg (58.72%) was removed. As the concentration of SDS increased to 32 mM and 64 mM, 2.76 mg/kg (62.39%) and 2.42 mg/kg (74.00%) of Cd were removed, respectively. Similar trends were observed for Pb, where the removal was 6.23 mg/kg (19.67%) at 8 mM, 12.62 mg/kg (39.84%) at 16 mM, 18.46 mg/kg (58.27%) at 32 mM, and 23.89 mg/kg (74.37%) at 64 mM. Cr removal followed a similar trend, with 1.62 mg/kg (40%) at 8 mM, 2.03 mg/kg (50.88%) at 16 mM, 2.32 mg/kg (58.15%) at 32 mM, and 2.56 mg/kg (64.16%) at 64 mM. The differences in removal efficiency for Cr and Pb were consistent with findings by [23].

Significance tests on the performance of EDTA and SDS revealed that EDTA showed positive variance for Cd and Pb, while negative variance was observed for Cr. For the radionuclide contaminants, the removal followed the order: U > Th > K, with p-values greater than 0.05, suggesting no significant difference in removal efficiency among these contaminants.

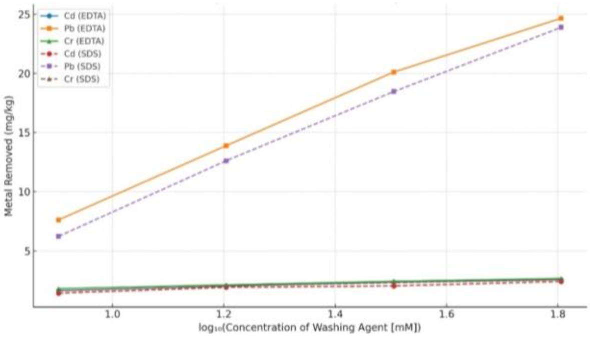
The percentage of contaminant removal was calculated using the following equation:

$$\text{Percent contaminant removed (\%)} = \frac{\text{Amount Removed}}{\text{Initial Concentration}} \times 100 \qquad (2)$$

Where:

Amount Removed refers to the mass of the contaminant extracted from the soil after washing (measured in mg/kg).

Initial Concentration refers to the original mass of the contaminant in the soil before washing (measured in mg/kg) [5].



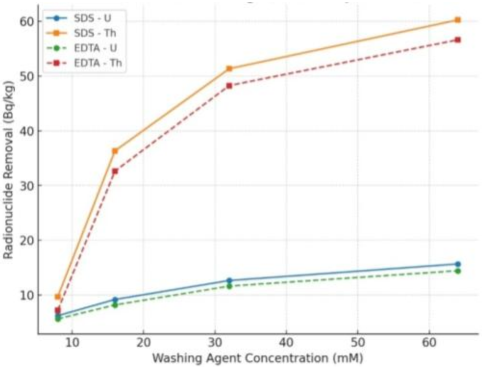


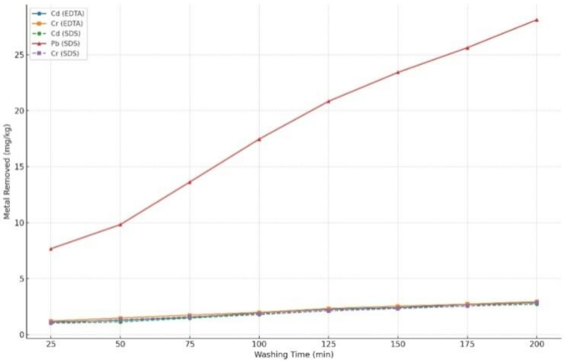
Figure 4a: Effects of concentration (mM) of Washing Agent in Toxic Metal Figure 4b: Effects of Washing Agent Concentration (mM) on Radionuclide Removal from Removal by EDTA and SDS Tin Mine Tailings (Batch Experiment)

Duration of Washing Time

Figure 5a and 5b illustrates the effectiveness of contaminant removal with varying washing times using an EDTA solution. The data indicate that the removal efficiency increased with washing time, with 1.22 mg/kg of Cd removed (34.25%) at 25 minutes. At 50 minutes, the removal rose to 1.26 mg/kg (38.53%), and by 200 minutes, 2.84 mg/kg (86.85%) of Cd was removed. A similar trend was observed for Cr, where removal increased from 1.21 mg/kg (30.32%) at 25 minutes to 2.93 mg/kg (73.43%) at 200 minutes. EDTA outperformed SDS under similar conditions in contaminant removal.

The overall trend demonstrated a quick initial increase in contaminant removal, with equilibrium achieved within 25 minutes. After this, the removal efficiency before slightly increasing, likely due to enhanced interactions between the surfactant and contaminants as washing time progressed. These observations align with previous studies [6][5]. A significant test revealed positive variance for Cr ($p < 0.05$) with SDS, though a negative variance was observed for Pb.

Figure 5a and 5b further depicts contaminant removal using SDS. For Cd, the removal ranged from 1.01 mg/kg (30.89%) at 25 minutes to 2.72 mg/kg (83.18%) at 200 minutes. Cr removal ranged from 1.03 mg/kg (25.81%) at 25 minutes to 2.86 mg/kg (71.68%) at 200 minutes, and Pb removal showed a consistent increase from 7.66 mg/kg (24.18%) at 25 minutes to 28.12 mg/kg (88.76%) at 200 minutes. The contaminants followed the order: Cd > Cr > Pb > U > Th > K in removal efficiency.



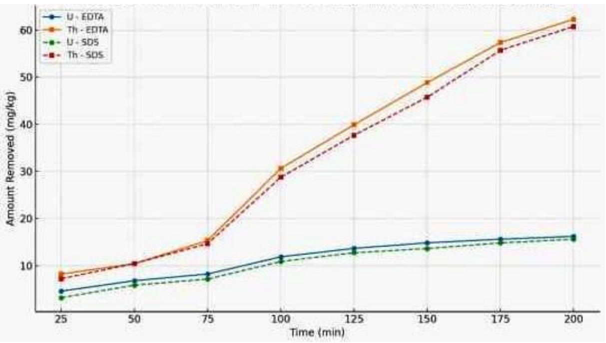


Figure 5a.Effects of Washing Time (min) on Toxic Metals Removal from Tin Mine Tailings

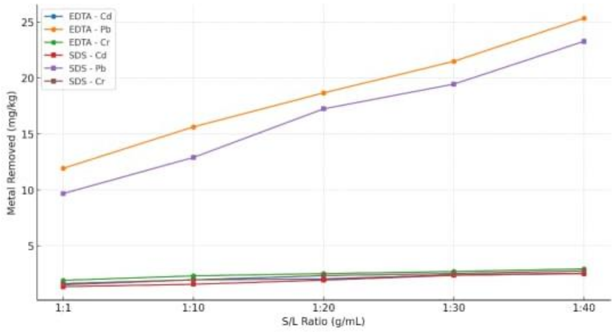
Figure 5b: Effects of Washing Time on Radionuclide Removal from Tin Mine Tailings

Effect of Soil/Liquid Ratio

The effect of soil/liquid ratio on contaminant removal is shown in Figure 6, where EDTA was used. At a ratio of 1:1, 1.53 mg/kg (46.79%) of Cd was removed, and this increased as the ratio expanded, with 2.56 mg/kg (78.29%) removed at 1:40. Cr and Pb removal followed a similar trend, with the highest removal observed at 1:40 (Cr: 2.94 mg/kg or 73.68%, Pb: 25.33 mg/kg or 79.96%).

In contrast, Figure 6a and 6b presents results using SDS, showing a similar pattern where the removal efficiency increased as the soil/liquid ratio decreased. The removal of Cd, Cr, and Pb followed the same trend: 1:1 had the lowest removal rates, while the highest removals occurred at 1:40.

These findings suggest that a higher soil/liquid ratio hinders contaminant removal efficiency, possibly due to decreased interaction between the surfactant and contaminants.



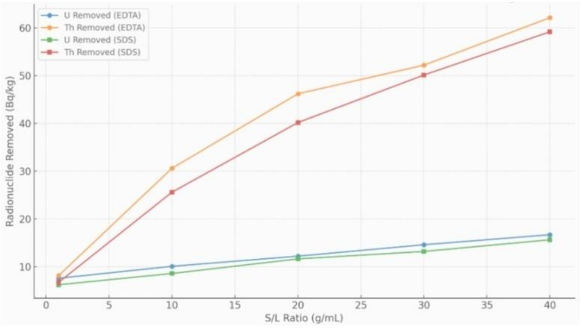
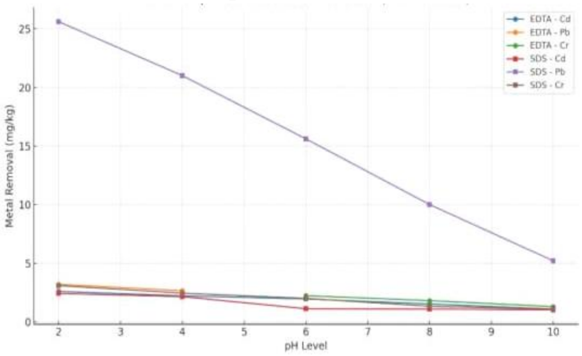


Figure 6a: Effects of Solid to Liquid Ratio on Toxic Metal Removal (mg/kg) from Tin Mine Figure 6b: Effects of Solid to Liquid (g/mL) Ratio on Radionuclide Tailings (Batch Experiment)

Effect of Initial Chelant and Surfactant pH

Figure 7a and 7b highlights the effect of pH on contaminant removal with EDTA. The best performance for Cd, Cr, and Pb occurred at pH 2, where 79.82% of Cd, 80.54% of Cr, and 87.22% of Pb were removed. As pH increased, the removal efficiency decreased. This trend is consistent with previous studies, which suggest that lower pH increases the solubility of contaminants, facilitating their removal from soil [13];[14].

Figure 7a and 7b shows similar results using SDS, where pH 2 and 4 yielded the highest removal rates for all contaminants. Pb showed the highest removal rate of 80.87% at pH 2, followed by Cr and Cd. The results confirmed that lower pH values were more effective for contaminant removal.



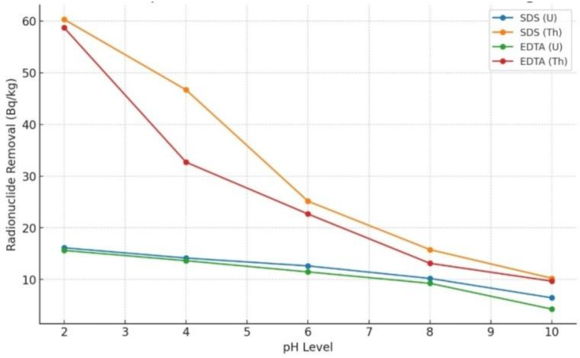
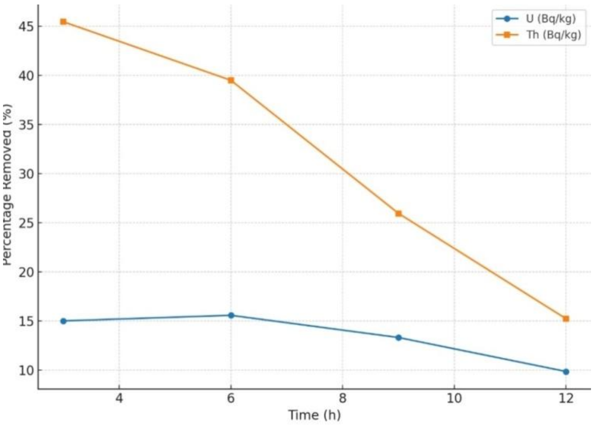


Figure 7a: Effects of pH on Toxic Metal Removal (EDTA and SDS Figure 7b: Effects of pH on Radionuclide Removal from Tin Mine Tailing

Column Washing Experiment

Effect of Blended Washing Agents with Respect to Time

Column washing experiments with SDS and EDTA solutions revealed that the amount of contaminants removed decreased as the leaching time increased figure 8a and 8b. For example, in the SDS column experiment, 86.85% of Cd was removed at 3 hours, and 38.23% at 12 hours. Similarly, for Cr and Pb, the removal efficiency decreased as time increased, reflecting the decreasing concentration of contaminants available for leaching.



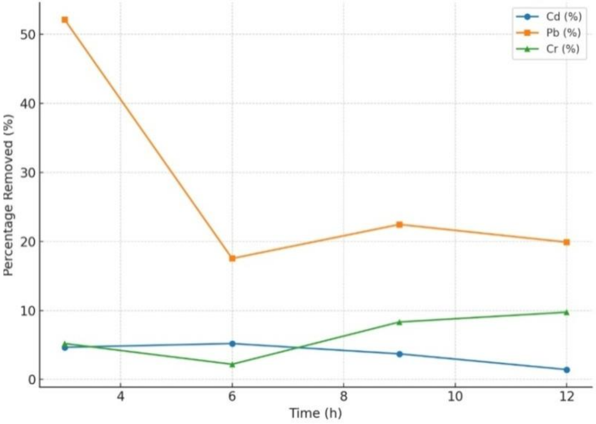


Figure 8a: Effects of Chelant and Surfactant Blend on Toxic Metals (Column Mode)

Figure 8b Effects of Chelant and Surfactant Blend on Radionuclides (Column Mode) in Tin Mine Tailings in Tin Mine Tailings

Chelant and Surfactant Blend Column Mode

Effect of Blend Percentage

Figure 9 presents the effect of varying percentages of EDTA and SDS blends on contaminant removal. At a 90% EDTA and 10% SDS blend, 96.33% of Cd, 95.74% of Cr, and 95.36% of Pb were removed, showing that higher EDTA percentages led to better contaminant removal efficiency. The contaminant removal efficiency decreased as the SDS percentage increased.

Chelant and Surfactant Blend Column Washing Mode

Further column experiments using the 90% EDTA and 10% SDS blend showed high efficiency in removing contaminants, particularly Cd, Pb, and Cr. At 3 hours, Cd removal was 96.33%, and Pb removal was 95.36%, indicating the superior performance of this blend over individual washing agents.

These results figure 9 underscore the effectiveness of combining chelants and surfactants, with the EDTA-SDS blend outperforming the individual agents in contaminant removal efficiency. This synergy is supported by significant improvements in contaminant removal from tailings, highlighting the potential of combined washing agents for environmental remediation.

Correlation Results of the Blended Surfactant and Chelant

Table 3 illustrate the performance of individual surfactant (SDS), chelant (EDTA), and their binary blend in removing contaminants during column experiments. The results show that, in general, the binary blend of SDS and EDTA was more effective in contaminant removal than either of the individual components alone. However, there were cases where the individual surfactant outperformed the blend, suggesting that the binary combination may exhibit both synergistic and antagonistic effects depending on specific conditions.

To further assess the nature of the interaction between SDS and EDTA in the blend, the percentage of contaminants removed by each individual component and their binary blend was analyzed using the Synergism Index (SI). The SI was calculated using the following formula [4][8]:

$$SI = \frac{(Es + Ee)}{(Es + e)} \qquad (3)$$

Where:

SI is the Synergism Index.

Es is the percentage of contaminants removed by the individual surfactant (SDS).

Ee is the percentage of contaminants removed by the individual chelant (EDTA).

Es + e is the percentage of contaminants removed by the binary blend of SDS and EDTA.

If the SI value is less than 1, the effect is considered antagonistic, whereas an SI greater than 1 indicates a synergistic effect. The results of the SI calculation are summarized in Table 3.

In the control experiment, distilled water was used as a blank washing solution. The contaminant removal performance with distilled water was as follows: 13.21 mg/kg of Th (1.74%), 0.49 mg/kg of Pb (1.55%), 0.04 mg/kg of Cd (1.22%), 0.04 mg/kg of Cr (1.00%), and 0.15 mg/kg of U (0.70%). Potassium (K) showed a negative variance in the analysis. These results confirm that the blend of surfactant and chelant outperformed distilled water in the removal of contaminants, highlighting the importance of the SDS-EDTA combination in achieving enhanced remediation.

Contaminant	**Percolation Period**	**Percent Contaminant removed**				**Synergism Index (SI)**	**Correlation**
		Es	**Ee**	**Es +Es**	**Es +e**		
	3	28.46	29.24	57.70	52.11	1.11	Synergistic
	6	22.44	24.63	47.07	17.53	2.68	Synergistic
Pd	9	18.46	20.62	39.08	22.46	1.74	Synergistic
	12	12.64	15.33	27.93	19.89	1.40	Synergistic
	3	2.84	2.92	5.76	4.67	1.23	Synergistic
	6	2.36	2.54	2.90	5.21	0.94	Antagonistic
Cd	9	1.82	2.13	3.95	3.72	1.06	Synergistic
	12	1.25	1.74	2.99	1.44	2.08	Synergistic
	3	3.21	3.41	6.62	5.21	1.27	Synergistic
	6	2.62	2.81	5.43	2.21	2.45	Synergistic
Cr	9	2.03	2.22	4.25	8.33	0.51	Antagonistic
	12	1.87	1.96	3.83	9.75	0.39	Antagonistic

	3	15.62 14.64 30.26 15.01	2.02	Synergistic
	6	13.46 12.76 26.22 15.58	1.68	Synergistic
U	9	9.64 8.22 17.86 13.32	1.34	Synergistic
	12	3.222 2.64 5.86 9.86	0.59	Antagonistic
	3	71.98 69.46 141.19 45.47	3.10	Synergistic
	6	52.62 50.21 102.83 39.51	2.60	Synergistic
Th	9	20.69 15.62 36.31 25.96	1.39	Synergistic
	12	15.27 14.28 29.55 15.23	1.94	Synergistic
K		BDL BDL BDL BDL	BDL	

BDL; Below detection limit

Table 3: Correlation between SDS, EDTA and their Binary (Ee=EDTA Es= SDS

Comparison of Results

Lead (Pb): 11.45–50.53 (mean ~31.7) — below FAO/WHO guideline of ~50.

Cadmium (Cd): 1.81–5.11 (mean ~2.48) — below ~5.

Chromium (Cr): 1.21–6.50 (mean ~3.99) — well within the ~50 guideline.

Highest Pb: Kuru A; Lowest: Rim

Cd highest in Rim; lowest in Sharubutu

Cr highest in Kuru A; lowest in Rim

Comparison with Global Averages

Global averages WHO/FAO (2001) **Pb=50mg/kg Cd=3mg/kg Cr= 50mg/kg**

Values obtained: lower than global average for all three toxic metal in both tin mine tailings and agricultural farm lands

Radium equivalent activity (Ra_eq): up to ~127 Bq/kg (< 370 Bq/kg limit).

External hazard index (H_ex): 0.07–1.23 (mean ~0.61); internal index (H_in): 0.06–1.85 (mean ~0.88)—both generally < 1, within safe thresholds.

Absorbed gamma dose rate: 30–114 nGy/h (mean ~58 nGy/h)—comparable to the ~60 nGy/h global average.

Annual effective dose: 5.97–140 µSv/y (mean ~71.4)—well below the ~6000 µSv/y global average.

Comparison with Global Averages

Global averages (UNSCEAR 2000): **^{238}U ≈ 35 Bq/kg; ^{232}Th ≈ 30Bq/kg; ^{40}K ≈ 420 Bq/kg**

Values obtained: lower than global average for all three radionuclides

III. Conclusion

The study demonstrated that the combined use of a chelant and surfactant is effective for soil washing applications. Batch experiments revealed that contaminant removal efficiency generally increased with the higher and initial concentration of both agents. Additionally, lower pH levels and reduced soil-to-liquid ratios significantly enhanced removal efficiency, with contaminant extraction also improving over extended washing durations.

The study also revealed that EDTA is more effective in removing toxic metals than radionuclides while SDS is more effective in removing radionuclides than EDTA.

In contrast, the column experiment showed a decline in contaminant removal over time. Statistical analysis using a t-test ($p < 0.05$) confirmed the significance of these findings.

Blends of chelant and surfactant at varying ratios were tested, and the results indicated that their combined application outperformed the use of either agent alone. The most effective formulation was a blend of 90% EDTA and 10% SDS. Synergism analysis further confirmed that the observed enhancement in performance was primarily due to synergistic interactions rather than antagonistic effects.

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