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Synthetic Precipitation Leaching Behavior of As, Cd, Cr, Pb and Zn in Contaminated Soil Stabilised and Solidified (S/S) using Cement and Sugarcane Bagasse Ash

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Abstract *- Soil remediation uses the Stabilisation and Solidification (S/S) method is widely used because of its higher rate of treatability using cement and other sustainable alternative materials. one of the important parameters when measuring the effectiveness of remediation is through the leaching test. Therefore, this study is conducted to evaluate the Synthetic Precipitation Leaching Procedure behavior (SPLP) of contaminated As, Cd, Cr, Pb, and Zn soil. The SPLP testing is conducted according to US EPA Method 1312 in SW-846. The samples containing solids and liquids are handled by separating the liquids from the solid phase, and the solids are then extracted with a diluted sulfuric acid/nitric acid solution. A liquid-to-solid ratio of 20:1 by weight is used for an extraction period of 18±2 hours. After extraction, the solids are filtered from the liquid extract and analyzed using ICP-MS. Results indicated that when SCBA was added to OPC content in soil samples, less heavy metal was leached from the S/S sample. On average, the satisfying result was shown by samples containing 10% OPC + 10% SCBA where reduction of heavy metals in final leachate is more than 90% for As, Cd, Cr, Pb, and Zn. In conclusion, a significant study shows that the combination of cement and SCBA can improve leachability quality through effective remediation methods.*

Keywords – *Stabilisation and solidification, contaminated soil, cement, SCBA, SPLP*

I. INTRODUCTION

Stabilization/Solidification (S/S) is a term used to describe the technology that involves mixing contaminated medium and binding reagents to reduce hazardous substances into non-hazardous substances which are environmentally acceptable for current land disposal [1]. Even though stabilization and solidification are similar terms, the binding reagent effect on waste is different. Stabilization refers to a process that reduces the chemical reaction by converting waste into a less hazardous substance. Meanwhile, solidification is a more specific process that treats material to increase its solidity and structural integrity [2]. Additionally, solidification does not remove nor degrade contaminants but prevents or eliminates their mobility.

The S/S method mainly consists of mixing contaminated material with suitable stabilizers. Lime, cement and other cementitious industrial waste materials are commonly used in S/S treatments. Among the types of binders mentioned, cement-based systems are the most widely used due to their relatively low cost, wide availability, and versatility [3]. However, the manufacture of cement often leads to environmental pollution. The CO_2 emitted from the manufacturing process has a major influence on climate change due to the greenhouse effect [4]. At present, cement is slowly being replaced by renewable binders such as agricultural byproducts that are more sustainable, cost-effective, and can improve the leaching characteristics of contaminated soils. Besides, the need for safe and environmentally friendly methods for eliminating heavy metals from contaminated soil has necessitated research on agricultural waste byproducts such as sugarcane bagasse ash, rice husk ash, sawdust, coconut husk ash, oil palm shells, and so on [5].

The utilization of agricultural byproducts in the production of cement-bonded materials offers an attractive alternative. Hence, in this research, sugarcane bagasse ash (SCBA) has been investigated for its suitability as a cement replacement in the S/S remediation method. The usage of sugarcane bagasse ash (SCBA) may help solve disposal problems and provide a cost-effective cement replacement material. On the other hand, sugarcane production was recorded at 1.8 billion tonnes in 2012 and is expected to increase yearly. Malaysia possesses nearly 37,000 acres of sugarcane plantations. Therefore, it is fairly easy to collect sugarcane bagasse with the establishment of sugarcane collection centers. For instance, the Federal Agriculture

Marketing Authority (FAMA) in Malaysia has set up a Sugarcane Collection Center or Pusat Pengumpulan Tebu (PPT) in Batu Pahat, Johor, for export purposes. Therefore, the use of agricultural wastes, particularly SCBA, would help solve agricultural waste disposal problems and provide a sustainable cement replacement material. To meet the specific objectives for evaluating the effectiveness of the remediation techniques, more than 100 leaching tests have been developed. One of the important leaching tests amongst all is Synthetic Precipitation Leaching Procedure (SPLP). Therefore, this study investigated the synthetic precipitation leaching behavior of soil contaminated by As, Cd, Cr, Pb, and Zn after remediation by cement and bagasse ash.

II. MATERIALS AND METHODS

A. Collection and preparation of soil and bagasse ash (BA)

This study was conducted at the Research Center for Soft Soil, Universiti Tun Hussein Onn, Malaysia. The contaminated soil has been taken from a Landfill site located in Bukit Bakri, Johor. During the sampling process, the soil on the top with a depth of 1 meter was removed to avoid the intake of humus, waste, and plant roots before placed in a polystyrene container. The sample is then taken to the laboratory to be dried in the oven at 105^{o}C for 24 hours. After drying, the samples were sieved using a 2 mm sieved and stored in the polyethylene plastic.

In this study, sugarcane bagasse was collected from juice hawkers around the Parit Raja area, Batu Pahat, Johor. The sugarcane bagasse was then taken to the laboratory for processing purposes. Sugarcane bagasse is washed with clean water to remove dirt and other substances during the collection process. Afterward, the sugarcane bagasse was dried in the sunlight at an uncontrolled temperature. The dried sugarcane bagasse is burned using a furnace at 650^{o}C for an hour to produce ash. According to [6], the burning process will reduce the carbon content by at least 4.9%. Through the X-Ray Fluorescent test, the sugarcane bagasse ash (SCBA) used in this study contains a chemical composition dominated by silicon dioxide (SiO_2) with 20.37% of the total mass. After the burning process is completed, the SCBA is cooled to room temperature before grinding using a grinder machine until it produces a fine powder with a size of 90 µm.

B. Production of stabilizing soil samples

In general, the study was conducted to partially replace a percentage of SCBA into cement as a binder in contaminated soil. All materials are mixed based on 4 groups, namely soil samples only (control samples), soil samples with cement only, soil samples with cement and SCBA, and soil samples with SCBA only. The distribution of samples in this group is seen as a process of finding the best combination of materials to evaluate the tests' effectiveness. A pre-determination of the mixed ratio has been made where the percentage of each sample percentage was shown in Table 1. To evaluate the study's effectiveness and accuracy, each sample group was triplicated for each curing period. Samples are also mixed in bulk to ensure homogeneity.

TABLE 1. PROPORTION AND LABEL

Sample Mixed	**Label**	**Percentage of Binder (%)**		
		Soil	**OPC**	**BA**
Soil	A	100	0	0
soil + cement	B	95	5	0
	C	90	10	0
	D	85	15	0
	E	80	20	0
soil + cement + bagasse ash (BA)	F	95	2.5	2.5
	G	90	5	5
	H	85	7.5	7.5
	I	80	10	10
soil + bagasse ash (BA)	J	95	0	5

In this study, the mixture of a sample depends on the water-cement ratio, where it is determined according to the optimum moisture content (OMC) from the compaction test, which is 0.2 to 0.4. Afterward, all the raw materials such as soil, cement, and SCBA are mixed using a mixer to ensure the sample's homogeneity. Then, the stabilized samples are compacted in a split mold to form a sample of 38 mm in diameter and 76 mm in height. A specially designed miniature hand compacting tool was used to compact the mixture into 4 layers with 50 blows. A hand compactor was used for each layer to flatten the mixture to a level surface before the sample was tamped at 50 times. The extruded specimens were wrapped and stored for 7, 14, and 28 days before toxicity characteristic leaching testing. The curing process was done under room temperature in a container that contains a small quantity of water to regulate the natural moisture.

C. Result and Analysis

a) the pH of Synthetic Precipitation Leaching Extraction:

Figure 1 shows the sample's pH changes after being extracted for 18 hours versus various stabilized samples mix designs after being treated for 28 days. The control of pH is a crucial factor in assessing the leachability of S/S samples, especially for heavy metal contaminated soil. The pH values of leachates of samples B, C, D, E, F, G, H, and I are essentially alkaline at 8.56, 9.94, 11.29, 11.95, 9.08, 10.17, 11.31, and 12.02, respectively. The pH of the leachate of sample A was slightly acidic at 6.26. These values indicate a trend whereby an increase in cement and SCBA content in the mix design also increases the treated samples' leachate pH. This result indicates that the mix ratio influences the pH value as sample A contained soil without any additives.

On the other hand, although samples B, C, D, and E contained a high percentage of OPC, their pH values were still lower than that of samples F, G, H, and I (incorporated with SCBA). This is due to the ability of SCBA to enhance pH development when it is incorporated with OPC as an additive. This result is consistent with the XRF analysis of the cement and BA, whereby very low calcium content may have contributed to the highest leachate pH.

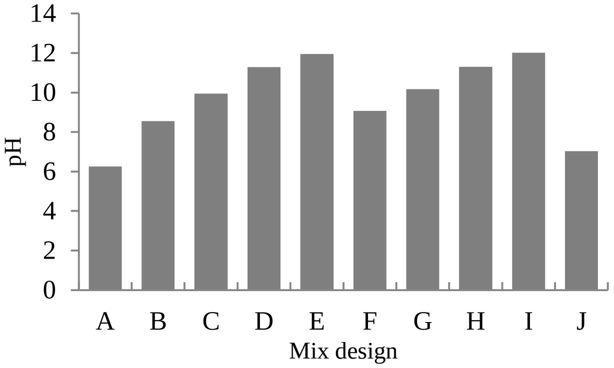

Fig 1: pH of stabilized soil samples after extraction

b) Leachability of Arsenic (As): Arsenic is a natural component of the earth's crust and is widely distributed throughout the environment in the air, water, and land. It is highly toxic in its inorganic form. Arsenic is one of the metals which is of major public health concern. The World Health Organisation (WHO) stated that the concentration of As to be released in landfill leachate should not exceed 0.05 mg/L. Table 2 shows that all samples that were stabilized and solidified by cement and bagasse ash successfully reduced the arsenic concentration by more than 99% after 18 hours of extraction. Samples made up of cement only contained arsenic in concentrations of 0.002 mg/L, 0.0012 mg/L, 0 mg/L, and 0 mg/L for samples B, C, D, and E, respectively. Better results were shown by samples containing cement incorporated with bagasse ash where samples F, G, H, and I successfully bound up to 100% of arsenic. This could be because bagasse ash is an adsorbent material that can reduce arsenic leachability [7].

Figure 2 shows that arsenic only leached out of soil samples (control samples) with a concentration above 0.05 mg/L, the drinking water standard regulated by WHO. However, this control sample still shows satisfying results because it was still able to bind to more than 98% of arsenic (1.309 mg/L) compared to the initial concentration of 96 mg/L after 18 hours of extraction. This significant result occurred due to arsenic adsorption, which is bound to the soil's organic elements.

TABLE 2. REDUCTION OF ARSENIC AFTER S/S TREATMENT

Sample	**Initial Concentration (mg/L)**	**Average Concentration of SPLP at (18 Hour) (mg/L)**	**Reduction (%)**
A	96	1.3090	98.64
B	96	0.0020	100
C	96	0.0012	100
D	96	0	100
E	96	0	100
F	96	0	100
G	96	0	100
H	96	0	100
I	96	0	100
J	96	0.0020	100

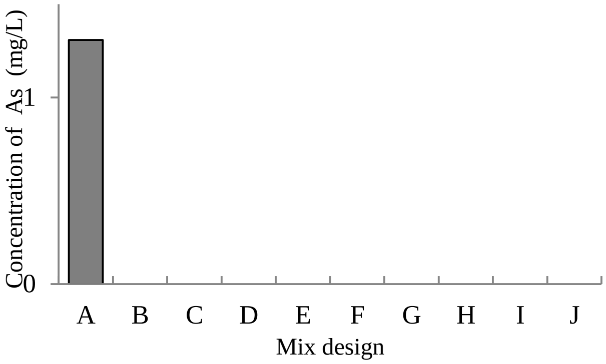

Fig 2: Concentration of arsenic after S/S treatment

Figure 3 shows the pH and As leachability of SPLP leachate. The pH values of leachate samples B, C, D, E, F, G H, I, and J were alkaline at 8.56, 9.94, 11.29, 11.95, 9.08, 10.17, 11.31, 7.01, respectively, while the leachate pH of sample A was slightly acidic at 6.26. The addition of BA to cement content has a positive effect on arsenic immobilization. More specifically, the addition of SCBA results in the further widening of the pH range (7 to 12) after a curing period of 28 days. These significant results were due to the binder's effect and the hydration process (up to 28 days).

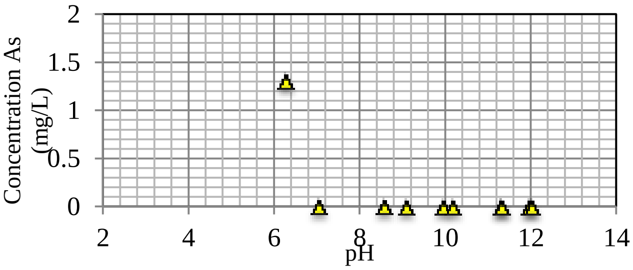

Fig 3: Relationship of pH and arsenic concentration in SPLP extraction

The intensity of pH affects the leachability of arsenic ranges between 9 to 10 [8]. In contrast to previous studies, this study found that arsenic concentration reduces when the pH exceeds 10 as shown by samples D, E, G, H, and I at 11.29, 11.95, 10.17, 11.31, and 12.02, respectively. However, at a lower pH, the concentration will be higher where sample A produces an arsenic concentration of 1.309 mg/L at pH 6.26. This suggests that leachability may be the main mechanism in this very pH-dependent study, as highlighted by [9].

c) Leachability of Cadmium (Cd): Cadmium is a naturally occurring toxic heavy metal that is commonly found in industrial workplaces, plant soils, and smoking activity. Due to its low permissible exposure to humans, a maximum concentration of 0.01 mg/L is allowed by the WHO in landfill leachate. Table 3 and Figure 4 show cadmium's leachability from S/S samples after a curing period of 28 days.

TABLE 3: REDUCTION OF CADMIUM AFTER S/S TREATMENT

Sample	**Initial Concentration (mg/L)**	**Average Concentration of SPLP at (18 Hour) (mg/L)**	**Reduction (%)**
A	79	74.44	5.77
B	79	9.957	87.40
C	79	0.882	98.88
D	79	0.712	99.10
E	79	0.663	99.16
F	79	7.230	90.85
G	79	0.790	99.00
H	79	0.649	99.18
I	79	0.539	99.32
J	79	0.350	99.56

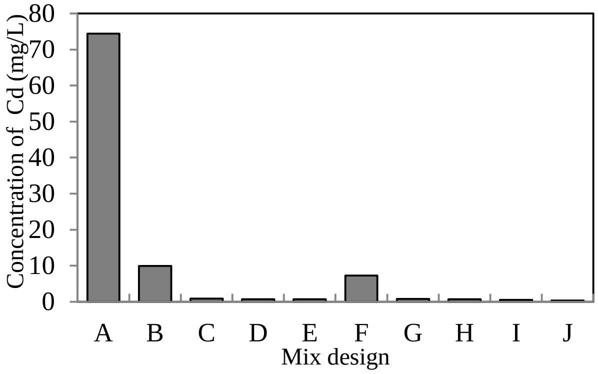

Fig 4: Concentration of cadmium after S/S treatment

Overall, all the samples were disturbed by cadmium exceeding the standard limit of 0.01 mg/L. Figure 5.17 clearly shows that sample A leached out cadmium with the highest concentration of 74.44 mg/L, followed by samples B and F with 9.957 mg/L and 7.23 mg/L, respectively. The significant result in these three types of samples indicates cadmium's high solubility rate during the treatment process. This is due to the presence of certain substances such as chloride ions or other substances in the soil that affect cadmium's leachability. Similarly, the low content of binders like cement and SCBA contributes to the increase of cadmium concentrations. For example, sample B containing only 5% of cement reduced the cadmium concentration to 9.957 mg/L compared to sample C containing 10% cement, which only successfully reduced the cadmium concentration to 0.882 mg/L. The difference between these two samples is 90%, even though the cement content difference was only 5%. A study conducted by [10] explained that cadmium would form a soluble and stable cadmium complex when contaminated soil contains a high quantity of chloride ions.

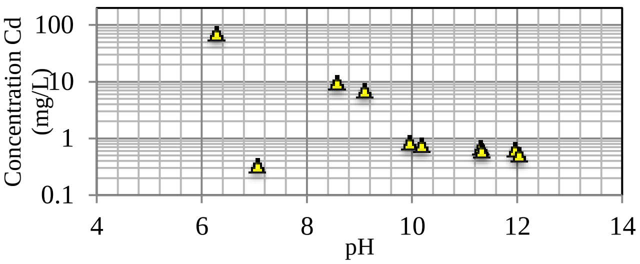

Fig 5: Relationship of pH and cadmium concentration in SPLP extraction

Figure 5 shows the effect of pH on the leachability of cadmium in the SPLP test. It can be concluded that the solubility of cadmium increases with decreasing pH. The concentration of cadmium was very low at a pH of between 10 and 13. In addition, some researchers have found that cadmium will be stable and bond within the cement matrix as long as its pH exceeds 6.8 [11].

d) Leachability of Chromium (Cr): Chromium compounds are used in dyes, paints, and leather tanning. These compounds are often found in soil and groundwater at abandoned industrial and landfill sites. In certain concentrations, these metals become toxic to humans, animals, and plants. Major factors governing the toxicity of chromium compounds are oxidation state and solubility [12]. Therefore, the WHO set the minimum concentration of chromium as 0.05 mg/L and below. Table 4 shows the average concentration of chromium in SPLP leachate after S/S treatment. Overall, there is a consistent result where all the samples stabilized and solidified by cement, and bagasse ash is below the limit set by WHO for drinking water. However, sample A shows a concentration that exceeded the permissible limit at 0.932 mg/L. Sample E showed outstanding results with a reduction of chromium concentration of up to 100%.

Samples mixed with cement and bagasse ash such as samples F, G, H, and I successfully reduced chromium absorption concentration by over 99%. These significant results show that bagasse ash has a positive impact on the treatment of chromium-contaminated soils due to the achievement of permissible limits and the reduction in the use of cement. This is expected to reduce the cost of treatment at the application level.

Additionally, better results were shown by sample J as shown in Figure 6, where this sample containing ash alone managed to reduce the leachability of chromium with a concentration of 0.048 mg/L in SPLP leachate or a reduction of 99.94%. This significant condition was due to ash's ability to bind the chromium ion in soil samples not incorporated with cement. In addition, according to [6], SCBA contains CaO, which helps in chromium precipitation.

TABLE 4: REDUCTION OF CHROMIUM AFTER S/S TREATMENT

Sample	**Initial Concentration (mg/L)**	**Average Concentration of SPLP at (18 Hour) (mg/L)**	**Reduction (%)**
A	82	0.932	98.86
B	82	0.034	99.96
C	82	0.011	99.99
D	82	0.008	99.99
E	82	0	100
F	82	0.044	99.95
G	82	0.039	99.95
H	82	0.024	99.97
I	82	0.019	99.98
J	82	0.048	99.94

Furthermore, Figure 6 shows that conducting the SPLP test for chromium is not as sensitive. The results indicated no difference between soil samples' leaching (sample A) and the samples mixed with the binder. Although the soil sample did not exceed the permissible limit, a 99% reduction in chromium concentration was achieved.

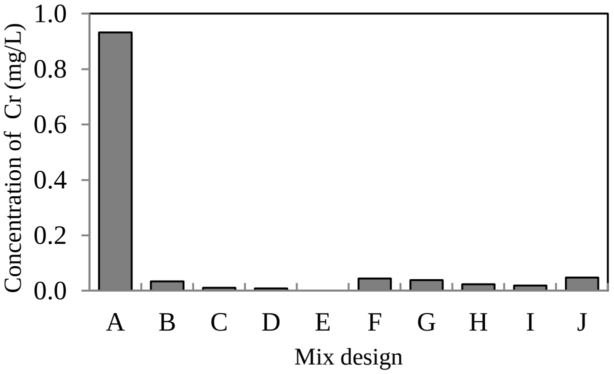

Fig 6: Concentration of chromium after S/S treatment

Figure 7 shows that pH does not affect chromium's leachability in the range between 6 to 13. This can be demonstrated through the pH difference between sample A (6.26) and sample E (11.95). Although there are significant differences between these two pH values, the difference in chromium concentration is small.

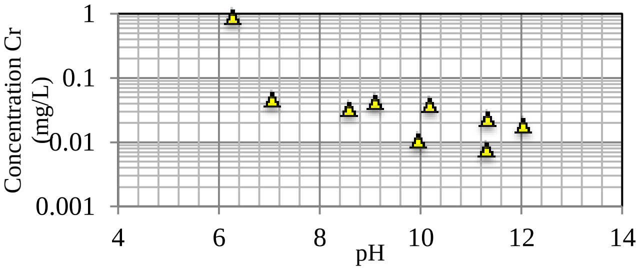

Fig 7: Relationship of pH and chromium concentration in SPLP extraction

e) Leachability of Lead (Pb): The WHO has identified lead as 1 of the 10 chemicals of major public health concern where action and regulations are needed to protect humans' health, especially children [13]. Even though there is no known level of lead exposure that is considered safe, the WHO limits the release of lead into landfill leachate with a concentration of less than 0.05 mg/L. Table 5 shows the average concentration of lead in SPLP leachate after a 28-day curing period.

Table 5 shows that sample A leached out lead at a high concentration of 87.172 mg/L or 94.75%. The presence of NaCl in soil influenced this significant condition because, according to [14], NaCl was able to convert insoluble lead compounds to soluble lead chloride (PbCl). However, with the increase in the samples' pH, this compound can be precipitated to Pb(OH)2, which can easily be turned into a soluble form. These conditions have been demonstrated by samples containing cement and bagasse ash where the concentrations of lead in the landfill leachate decrease after the remediation process. Samples E and I showed the highest reductions of 96.75% and 95.12% or leachate concentrations of 2.99 mg/L and 4.49 mg/L, respectively.

TABLE 5: REDUCTION OF LEAD AFTER S/S TREATMENT

Sample	**Initial Concentration (mg/L)**	**Average Concentration of SPLP at (18 Hour) (mg/L)**	**Reduction (%)**
A	92	87.172	5.25
B	92	10.788	88.27
C	92	6.232	93.23
D	92	6.112	93.36
E	92	2.990	96.75
F	92	10.322	88.78
G	92	7.345	92.02
H	92	6.644	92.78
I	92	4.490	95.12
J	92	12.113	86.83

Figure 8 shows the same reduction pattern produced by samples mixed with cement and ash. Although the samples' results were not as good as the samples containing cement alone, the results were satisfactory with a decrease of the lead of more than 90% as shown by samples G, H, and I with final concentrations of 7.345 mg/L, 6.644 mg/L, and 4.49 mg/L respectively. Likewise, sample J that contained bagasse ash alone managed to reduce the leachability of the lead up to 88.83% or a final concentration of 12.113 mg/L. This condition clearly shows that bagasse ash can successfully bind more lead ions if the percentage is increased to 10% - 20%.

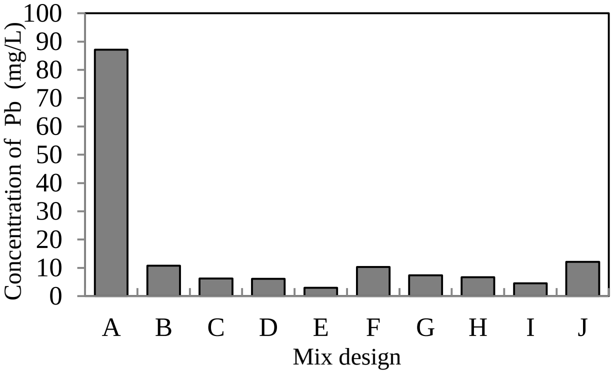

Fig 8: Concentration of chromium after S/S treatment

Figure 9 shows the relationship of pH with lead concentrations after S/S treatment. The figure indicated that all samples have leached out Pb in concentrations greater than 0.05 mg/L, which exceeds the WHO drinking water standard. In general, the diagram shows the sample pH between 6.26 and 11.95. Normally, the precipitation process's pH is between 9 and 10 [15] or 6 to 9 [16]. Therefore, this study concludes that pH value affects the leachability of lead. Still, the leaching fluid used in the SPLP test contributed to the increase in lead concentration in landfill leachate.

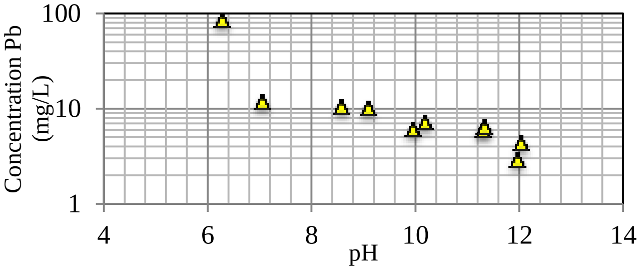

Fig 9: Relationship of pH and lead concentration in SPLP extraction

f) Leachability of Zinc (Zn): Zinc is an indispensable trace element found in almost all food and potable water in the form of salts or organic complexes. In water, zinc can be found in abundant quantities due to the leaching of zinc from piping and fittings. Increased zinc content can lead to health difficulties such as stomach cramps, skin irritations, vomiting, nausea, anemia, root trouble in the pancreas, protein metabolism, and arteriosclerosis [17]. The WHO stated that 5 mg/L is considered as the standard concentration for zinc in drinking water. Table 6 shows the average concentration of zinc in SPLP leachate after a curing period of 28 days.

Table 6 shows that the soil sample leached out the zinc at the highest rate of 42.221 mg/L or a reduction of 43.87%. Similarly, some samples did not exceed the minimum drinking water level, such as samples B, C, F, and J with concentrations of 6,734 mg/L, 6.221 mg/L, 5.11 mg/L, and 5.302 mg/L, respectively. Even though these samples did not exceed the permissible limit, the reduction of zinc concentration in the final landfill leachate exceeded 90%.

TABLE 6: REDUCTION OF ZINC AFTER S/S TREATMENT

Sample	**Initial Concentration (mg/L)**	**Average Concentration of SPLP at (18 Hour) (mg/L)**	**Reduction (%)**
A	77	43.221	43.87
B	77	6.743	91.24
C	77	6.221	91.92
D	77	4.231	94.51
E	77	3.443	95.53
F	77	5.11	93.36
G	77	4.219	94.52
H	77	3.91	94.92
I	77	2.11	97.26
J	77	5.302	93.11

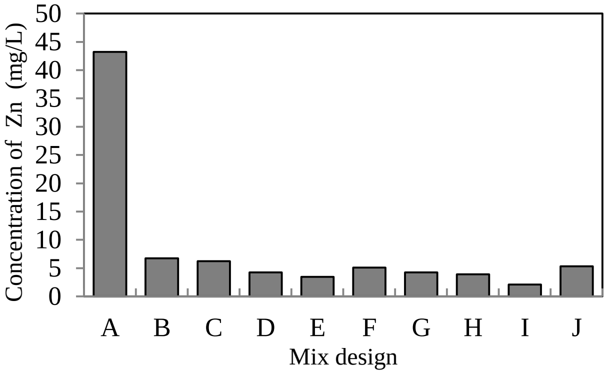

Fig 10: Concentration of zinc after S/S treatment

Figure 10 clearly shows that samples treated with cement (samples D and E) successfully reduced the zinc concentrations to 4.321 mg/L and 3.443 mg/L, respectively. Better results were obtained by samples mixed with cement incorporated with SCBA. The final concentrations of landfill leachate were 4.219 mg/L, 3.91 mg/L, and 2.11 mg/L for samples G, H, and I.

Figure 11 shows the leaching of zinc influenced by pH. According to [18], the common pH for zinc to achieve the desired precipitation is between 5.3 and 9. However, in contrast to this study, zinc has strongly bonded to the cement matrix, and this affects the leachability of zinc by the SPLP

Extraction fluid. This significant condition shows that a pH of more than 9 is ideal as the leachability of zinc decreased by 97% in the final landfill leachate, as shown by a sample I (pH = 12.02).

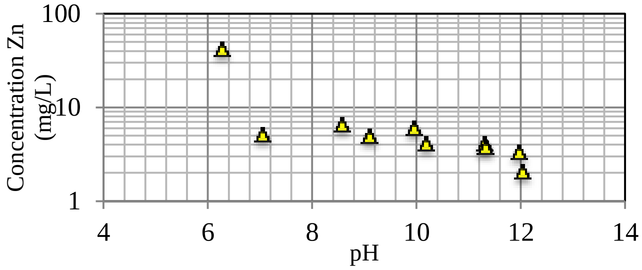

Fig 11: Relationship of pH and zinc concentration in SPLP extraction

D. Conclusion

In conclusion, all the S/S samples with OPC and SCBA has successfully reduced the arsenic concentrations exceeding 99% after 18 hours of extraction. This significant result takes place due to arsenic adsorption to organic elements, especially in samples containing SCBA. For the stabilization of cadmium, all the samples were disturbed by cadmium exceeding the drinking water standard limit of 0.01 mg/L. This is due to the presence of certain substances such as chloride ions or other substances in the soil that affect cadmium's leachability. Similarly, the low binder content of OPC and SCBA contributes to the increase in cadmium concentrations. For the treatment of soil containing chromium, consistent results below the WHO limit for drinking water were obtained by all S/S samples. However, sample A showed a concentration exceeding the permissible limit, while sample E showed outstanding results, reducing chromium concentrations of up to 100%.

Similarly, the lead treatment showed a satisfactory reduction pattern produced by samples mixed with OPC and SCBA. Although these samples' results were not as good compared to the samples containing OPC stand-alone, it is satisfactory as there was a decrease of more than 90% in lead content. Finally, for the treatment of zinc contaminated soil, samples B (5% OPC), C (10% OPC), F (15% OPC), and J (5% SCBA) did not exceed the minimum drinking water level. However, the results are still considering the reduction of zinc concentrations in the final landfill leachate exceeded 90%.

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