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EFFECT OF VOLTAGE VARIATION ON THE GEOTECHNICAL PROPERTIES AND REMOVAL EFFICIENCY OF HEAVY METAL CONTAMINANTS FROM CONTAMINATED SOIL USING ELECTROKINETIC REMEDIATION TECHNIQUE

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ABSTRACT: *A lot of research work has shown that despite the effectiveness of the electrokinetic remediation technology in decontaminating heavy metal contaminated soils, more work is still required to fully understand the role of voltage in the remediation process. There is need to establish the optimum voltage that would best remove heavy metals from such contaminated soil and its attendant effect on the geotechnical properties of the remediated soil. Effect of voltage variation on the removal efficiency of lead, copper and the geotechnical properties of remediated heavy metal contaminated soil using electrokinetic remediation technique was investigated in this research. The contaminated soil was remediated by applying direct current (DC) to the remediation setup at 0.5V/cm, 1.0V/cm, 1.5V/cm and 2.0V/cm. The concentration of the heavy metals after remediation were determined using the Oxford Instrument Analyzer to evaluate removal efficiency, geotechnical properties tests were also conducted on the soil specimens at each phase of remediation. The results showed that the lead removal efficiency was highest at 2.0V/cm (86%) with the shortest remediation time of 5days and lowest at 0.5V/cm (39%) at 9days. 52% of copper was removed at 2.0V/cm in 5days and 29% at 0.5V/cm after 9days of remediation. At 1.0V/cm, the lead and copper removal efficiency are 75% and 40% respectively. There was no significant change in the Specific Gravity of all the soil samples with the test results lying between 2.0 and 2.2. The soil is generally silty fine sand with not less than 40% passing the sieve no.200 (75micron). 45% passed through sieve 75micron for unremediated soil and slightly reduced to 40%, 40.4% and 40.2% for 30V, 45V and 60V respectively. The soil is non-plastic with the liquid limit of between 25.8% and 29.5% belonging to the A-4 group of soil. The maximum dry density improved across all the three compactive efforts, from 1.8390g/cm^3 to 1.8480g/cm^3 with WAS compactive effort and from 1.8000g/cm^3 to 1.8320g/cm^3 with BSL method with an average optimum moisture content of 10%. The CBR values increases with increase in voltage applied. The unsoaked CBR values averagely increased with 31%, 18% and 7% for BSH, WAS and BSL compactive efforts respectively. The durability index with resistances of 89% and 90% to loss in strength was recorded at 1.0V/cm and 1.5V/cm respectively, this, when compared to the resistance to loss in strength of 71% in unremediated soil has respectively 25.3% and 26.8% durability advantages. There was also a consistent increase in the UCS values, from 381kN/m^2 to 474kN/m^2 and from 351kN/m^2 to 447kN/m^2 when WAS and BSL methods of compaction were used. Generally, there was improvement in the geotechnical properties of the remediated soil. These improvements are maximum at 1.0V/cm and 1.5V/cm with little or no further improvement at 2.0V/cm. It is recommended that 1.0V and 1.5V are suitable for remediation purpose since it requires low energy consumption.*

KEYWORDS: Electrokinetic remediation, removal efficiency, voltage, geotechnical properties, heavy metal.

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INTRODUCTION

The aim of electrokinetic remediation technology is to remove heavy metal contaminants from low permeability contaminated soils under the influence of an applied direct current. In recent times, a number of promising laboratory and pilot-scale studies and experiments has been conducted by researchers (Haruna et al., 2023; Reddy *et al*., 2006).

Heavy metals refer to a group of toxic elements that are both biologically and industrially important. Particularly, the widespread contamination of soil with lead and copper causes one of the most complex environmental problems that can seriously affect soil properties and environmental quality. Heavy metals are released into the soils by both natural and anthropogenic sources. Although soil contamination by heavy metals started in ancient times, the problem was provoked after the industrial revolution owing to dramatic increase in the use of heavy metals in various modern technologies (Khalid *et al*.; 2017).

Although, electrokinetic remediation methods have proven to be more effective than most traditional techniques used in remediating low permeability soils contaminated with heavy metal, there is still the problem of "what is the optimal voltage of the electrokinetic remediation process" that is most likely to produce the best result in terms of heavy metal removal and its impact on the properties of the soil (Mohammed *et al*., 2013).

Heavy metals at higher concentration are known to influence the growth of microbes in soil and their activities may directly affect soil properties (Minnikova et al., 2017).

Electrokinetic process has been effectively applied in remediating both heavy metals and organic compounds contaminated soil. Electrolyte such as ethylenediaminetetra acetic acid (EDTA), citric acid, sodium nitrate ($NaNO_3$) can be used to enhance the efficiency of electrokinetic remediation. The results from many laboratory studies have shown excellent contaminant removal efficiencies by the use of the electrokinetic technique (Sani et al., 2023; Haruna *et al.,* 2023).

Saleem (2021) carried out a research on efficient removal of copper and cadmium from contaminated soil utilizing electrokinetic process. From his findings, metal removal is positively related to the current density. His results showed that current density of 9.3 mA/cm^2 is capable of bringing the copper concentration below 83µg/g from 4975.6µg/g within 100 hours of experimental run. Rate of copper removal increased as the inter-electrode separation decreased achieving 68.7% and 89.5% copper removal at 30cm and 15cm electrode separation respectively.

Malavika and Deepthy (2017) studied the effectiveness of electrokinetic remediation in decontaminating lead contaminated soil. They also evaluated the efficiencies of different electrolyte such as 0.1M EDTA, 0.1M citric acid and tap water. They used different electrode materials which included copper and graphite. The soil was partitioned into three regions (S1, S2 and S3) representing areas close to cathode, middle and anode regions in that order. From their results, it was clear that 0.1M EDTA showed better result than 0.1M citric acid and tap water in decontaminating the soil. Concentration of lead was higher in region S1 and less in other regions which indicates metal migration from the soil. Thus, it was found that the migration of Lead occurred from anode to cathode region. When tap water and citric acid were used as electrolyte, the lead mobility was poor which resulted in less removal efficiency of lead. The result showed that EDTA removed about 68% of Pb from the contaminated soil. In

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determining the efficiency of the different electrode materials (graphite and copper) used, test to determine concentration of lead at different sections after electrokinetic process was conducted. From the results, the removal efficiency of lead in anode region is same in both cases, but graphite showed better migration of lead in all the three regions. The removal efficiency of the test is 68%. The difference in removal efficiency with increase in time duration was also studied, they considered 24hrs and 48hrs from where they discovered that percentage removal of lead after electrokinetic process was found that more amount of lead was concentrated towards the cathode region as the time duration increased.

Lee *et al.,* (2016) used monopotassium phosphate (KH_2PO_4) as an anolyte and showed that removal efficiencies increased by greater than 50% for asenic (As) and greater than 20% for copper (Cu). However, removal of the lead and zinc was relatively inefficient (below 20%). Hussein et al., (2019) studied remediation of Lead contaminated soil using clean Energy (solar energy as power source) in Combination with Electrokinetic method for three soil types (sand, sandy loam and silty loam). They found out that sand was the most conducive to electrokinetic lead (Pb) clean-up with an overall efficiency of 90.7%, followed by sandy loam (63.3%), and silty loam (42.8%).

Dellisanti (2016) carried out an in-field Joule heating vitrification of tons of zinc and lead rich ceramic waste by heating up to about 1850°C. They reported that the vitrification method was greatly efficient to clean-up tons of heavy metals contaminated waste materials and can be applied for cleaning huge volumes of soil. Temperature during vitrification plays a key role in the immobilization of heavy metal in soil samples.

Rosestolato *et al.,* (2015) remediated approximately 400kg of soil using electrokinetic remediation and removed about 60% of total mercury (Hg) from the contaminated soil in three months. Electrokinetic remediation method is also used in combination with other techniques/processes such as electrokinetic microbe joint remediation (Yu *et al.,* 2009), electrokinetic-chemical joint remediation, electrokinetic-oxidation/reduction joint remediation (Yang *et al.,* 2015), and coupled electrokinetic phytoremediation (Mao *et al.*, 2016).

From the fore-going, electrokinetic methods have proven to be effective than most traditional techniques used in remediating low permeability soils contaminated with mixed heavy metal contaminants. Most works reviewed on electrokinetic remediation were silent about the optimal voltage for electrokinetic remediation process, let alone monitor the effect of its variation on soil properties. There is need to explore more on the usual voltage gradient of 1V/cm for bench-scale studies by considering lower and higher voltages.

Hence, the focus of this research work is to vary the voltage applied to an electrokinetic remediation process and evaluate the impact on the geotechnical properties of the remediated soil and the removal efficiency of the predominant lead and copper.

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MATERIALS AND METHODS

Elecrokinetic remediated soil

In this research, the electrokinetic remediated soil was collected from a mining site located at Rima village 12km from Birnin Gwari town in Birnin Gwari Local Government Area of Kaduna State, Nigeria. Preliminary analysis on the soil indicated that the soil was predominantly contaminated with lead and copper deposits. The electrokinetic remediation experiment was conducted at the Civil Engineering Laboratory, Baze University, Abuja. The remediation was in four (4) phases applying voltage at 15V, 30V, 45V and 60V on four (4) contaminated samples.

The experimental setup was similar to the one adopted by Bimastyaji *et al.*, (2018). The setup consists of a square Cell (300mm long, 300mm wide and 250mm high) containing the contaminated soil, direct Current (DC) source, a voltage regulator with capacity of 15V, 30V, 45V and 60V to achieve 0.5V/cm, 1.0V/cm, 1.5V/cm and 2.0V/cm respectively. Four (4) graphite electrodes (2 each at cathode and anode), the electrode is 300mm long with a thickness of 8mm placed in the electrolyte compartments (Height: 250mm, Length:300mm and width: 50mm).

The cell is designed in such a way that it allows fluid flow in the two compartments from one end to the other, to achieve this, a well perforated plexi glass was used as the divider between the three compartments which permits injection of the processing fluid and allow electroosmotic flow between the anode and cathode. Electrodes used for this study was graphite placed vertically with a face to face configuration at the anolyte and catholyte compartments for passing direct current from the DC Supply using connecting cables attached to clips through the set-up. Solar power was used to ensure continuous power supply to the electrokinetic set-up. 0.1mole of EDTA (Ethylenediaminetetraacetic Acid) and 0.01mole of citric acid were used as the electrolyte at the cathode and anode compartments respectively. By connecting the setup to a direct current source, remediation process was automatically enabled with applicable voltage.

Figure 1: Laboratory set-up for the Electrokinetic Remediation Process

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Table 1a: Summary of the Laboratory scale setup for the remediation

Item	Height	Length	Width	Thickness	Concentration
EKR Cell	25cm	30cm	30cm	-	-
Electrode	-	30cm		0.8cm	-
Citric Acid	-	-	-	-	0.01M
EDTA	-	-	-	-	0.1M
Electrolyte compartment	25cm	30cm	5cm	-	-

Geotechnical properties of the soil

The geotechnical properties which include the natural moisture content, particle size distribution, Atterberg limit, specific gravity and compaction characteristics of were determined both before and after electrokinetic remediation processes.

California Bearing Ratio (BS 1377 Part 4:1990)

The California Bearing Ratio(CBR) test is an indirect measurement of resistance of soil material to penetration of standard plunger under controlled density and moisture conditions.

For British standard heavy compaction (BSH), the sample was sieved through 20mm sieve from which 6kg of the sample of soil specimen was obtained. Water was added to the soil in the quantity such that optimum moisture content was reached. Then the soil and water were mixed thoroughly. Spacer disc was placed over the base plate at the bottom of mould and a coarse filter paper is placed over the disc. The prepared mix was divided into five parts. The mould was cleaned and lubricant was applied. Then one part of the mould was filled with one part of the prepared soil. That layer was compacted by giving 62 evenly distributed blows using the 4.5kg rammer layer by layer until the fifth. The top layer of the compacted soil was scratched each time to enhance proper binding. The collar was removed and excess soil was strucked off. The base plate was then removed and the mould inverted and clamped to the baseplate. Surcharge weights of 2.5kg was placed on top surface of the soil. Mould containing the compacted specimen was placed in position on the testing machine. The penetration plunger was brought in contact with the soil and a load of 4kg (seating load) was applied so that contact between soil and plunger was adequately established. Then dial reading was adjusted to zero. Load was applied such that penetration rate was 1.25mm per minute. Load at penetration of 0.5, 1, 1.5, 2, 2.5, 3, 4, 5, 7.5, 10 and 12.5mm were noted. Hence, California Bearing Ratio was calculated at 2.5mm and 5.0mm (top and bottom) from which the average was evaluated. After this, the sample was soaked in water for 24hours to determine soaked cbr values

As for British standard light compaction effort, the sample was prepared as before. The prepared soil water mix was divided into three parts. Then each part of the soil was filled and compacted into the mould with each layer receiving 62 blows of the 2.5kg rammer. Hence, the sample was tested in the CBR machine in the manner described above.

And for West Africa standard compaction effort, the sample was prepared as before. The prepared soil water mix was divided into five parts. Then each part of the soil was filled and compacted into the mould with each layer receiving 30 blows of the 4.5kg rammer. Hence, the sample was tested in the CBR machine.

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The CBR was calculated as:

CBR=(*Measured load/Standard load*)×100% equation (3.1)

where standard load = 13.24kN of 2.5mm penetration

= 19.96kN of 5.0mm penetration

Unconfined Compression Strength (UCS)

The compacted specimen for the test had a minimum diameter of 38mm with a height of 76mm. The disturbed sample was compacted using all the three compactive efforts (BSL, WAS and BSH) at the appropriate optimum moisture content in a 1000cm^3 compaction mould, the sampling tubes were then driven into the compacted soil to obtain a cylindrical specimen. The specimen after extrusion was trimmed appropriately to conform to the dimensions above. The specimens were then wrapped in a thin rubber membrane. Eight (8) specimen were prepared to allow curing and testing of two (2) each at 7, 14, 21 and 28days after curing in open air. On attaining the curing ages above, the specimen was unwrapped and positioned in between the bottom and upper plates of the UCS testing machine. The dial gauge was adjusted to 0 and the loading steering was properly seated. Then, compression load was applied to induce deformation on the specimen so that axial strain can be recorded at 0, 20, 40, 60, 80, 100, 120, 140, 160, 180, 200, 250, 300, 350, 400, 450, 500, 550, 600, 650, 700, 750, 800, 850, 900, 950, 1000, the maximum compressive load that caused failure of the specimen was obtained and used in the calculation for the Unconfined Compressive Strength (UCS).

However, the durability test was conducted by curing (soaking) the cylindrical specimen obtained from each of the compactive efforts (BSH, BSL and WASC) in water for 7days. After which the specimen were tested and the strength calculated using the formula below.

$$= \frac{Dial\ gauge\ reading\ X\ PRF\ X\ \left(1-\frac{\Delta L}{Lo}\right)}{Cross\ sectional\ Area\ of\ the\ Specimen}$$ equation (3.2)

Where, PRF = Proving Ring Factor, Lo = Original Length, ΔL = Change in Length

Resistance to loss in Strength (%)

$$= \frac{\boldsymbol{UCS}(\mathbf{7}\boldsymbol{days\ cured}+\mathbf{7}\boldsymbol{days\ soaked})}{\boldsymbol{UCS}(\mathbf{14}\boldsymbol{days\ cured})}\ x\ 100$$ equation (3.3)

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RESULTS AND DISCUSSION

Table 2a and 2b below show the summary of concentration and removal efficiency of lead and copper contaminants before and after the electrokinetic remediation using the Oxford Instrument Analyzer (XRS-FP analysis).

Table 1b Results of metal analysis showing background metal concentration present in soil sample before remediation

Metal	Concentration (µg/g)
Cobalt (Co)	28.0
Nikel (Ni)	0.0
Copper (Cu)	70.0
Zinc (Zn)	5.0
Silver (Ag)	27.0
Gold (Au)	7.0
Lead (Pb)	170.0
Mercury (Hg)	3.0

Table 2a: Removal efficiency of Lead (Pb) at different voltage

Lead (Pb)						
Applied Voltage	**Concentration (mg/kg)**				**Removal Efficiency (%)**	**Duration (days)**
Unremediated	184					
Sampling point	**S1**	**S2**	**S3**	**Average**		
15V	97.0	102.0	136.0	111.7	39	9
30V	24.0	33.0	81.0	46.0	75	7
45V	20.0	31.0	50.0	33.7	82	5
60V	16.0	29.0	31.0	25.3	86	5

Table 2b: Removal efficiency of Copper (Cu)) at different voltage

Copper (Cu)						
Applied Voltage	**Concentration (mg/kg)**				**Removal Efficiency (%)**	**Duration (days)**
Unremediated	80					
Sampling point	**S1**	**S2**	**S3**	**Average**		
15V	53.0	27.0	90.0	56.7	29	9
30V	80.0	10.0	55.0	48.3	40	7
45V	70.0	14.0	50.0	44.7	44	5
60V	53.0	11.0	51.0	38.3	52	5

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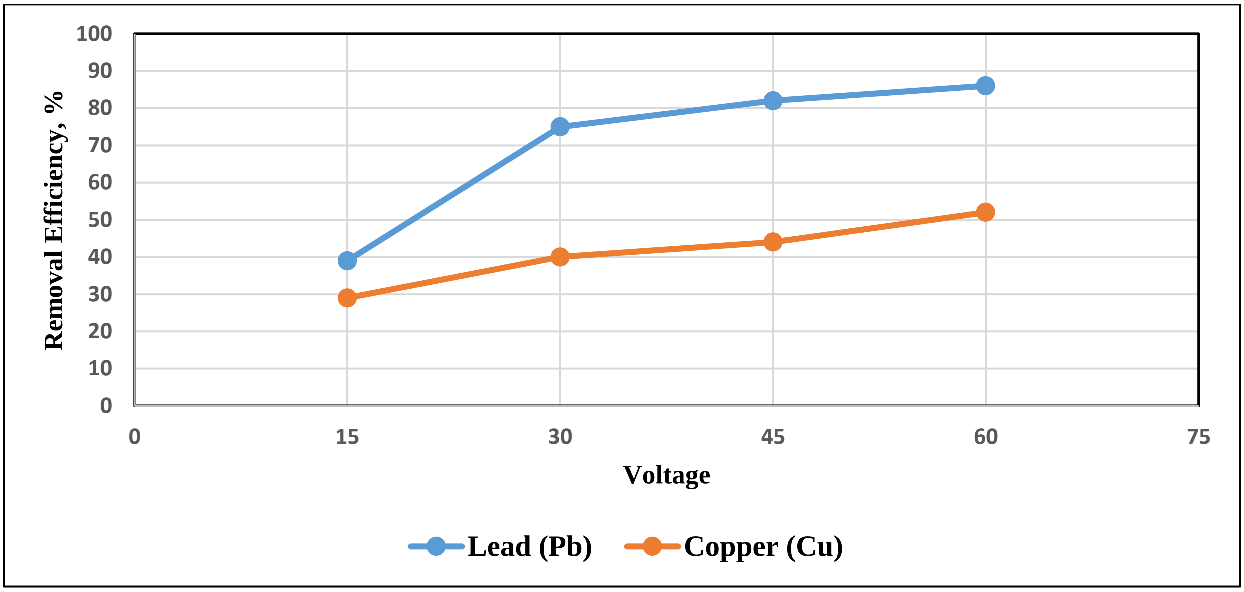

Figure 2: Concentration of lead and copper at different voltages

The geotechnical properties test results including the compaction characteristics of the electrokinetic remediated soil are presented below. The natural moisture content of the unremediated soil was 10.97%. Samples were collected after remediation was complete. This was to determine moisture content (processing fluid) of the remediation process. From the results, there was demand for more moisture as the applied voltage was increased. Available moisture at 15V was 24%, this increases steadily to 26% and 28% at 45V and 60V respectively. This is consistent with the findings of Saleem (2021). Figure 3 show demand of moisture induced by higher energy consumption at higher voltages.

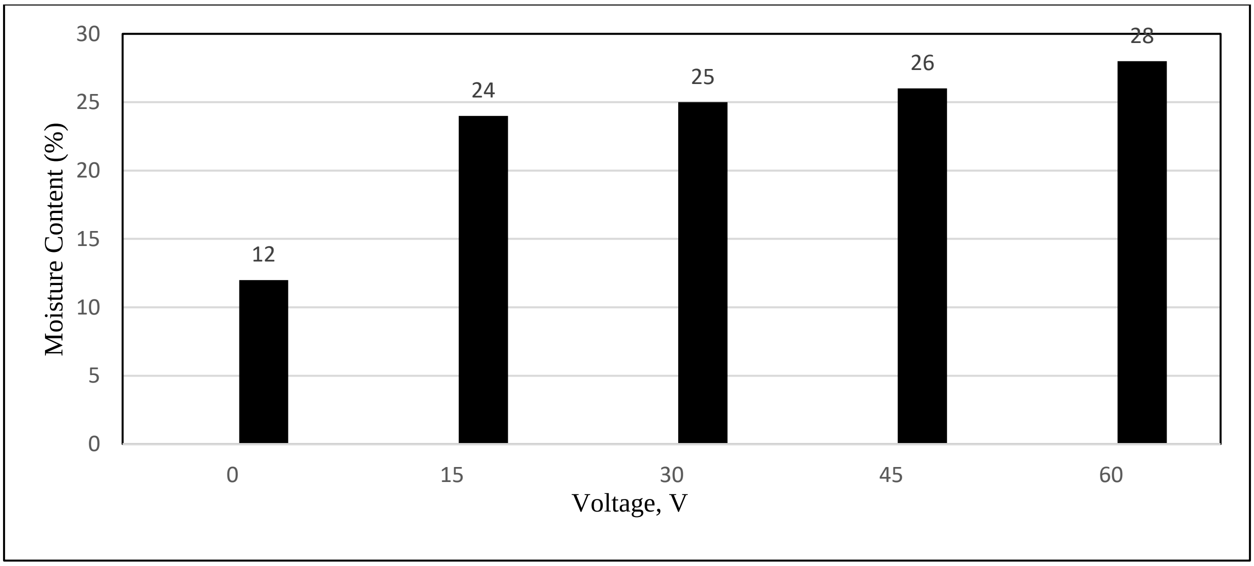

Figure 3 Variation of Moisture Content at different voltage

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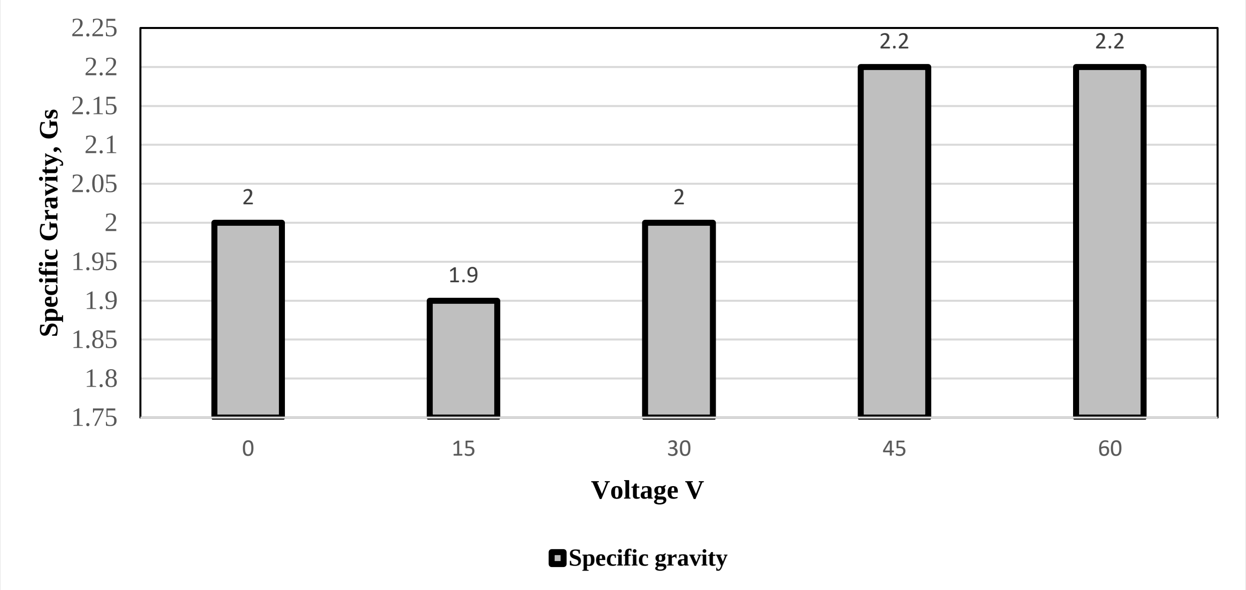

Figure 4 Variation of Specific Gravity at different voltage

The specific gravity in figure 4 shows an increase from 1.9 at 15V to 2.2 at 45V and 60V. This confirms the clogging and closure of pores preoccupied by the lead and copper particles, thereby making the soil denser compared to its status before remediation. This result, however, is similar to the reported data by Jayasekera, (2015).

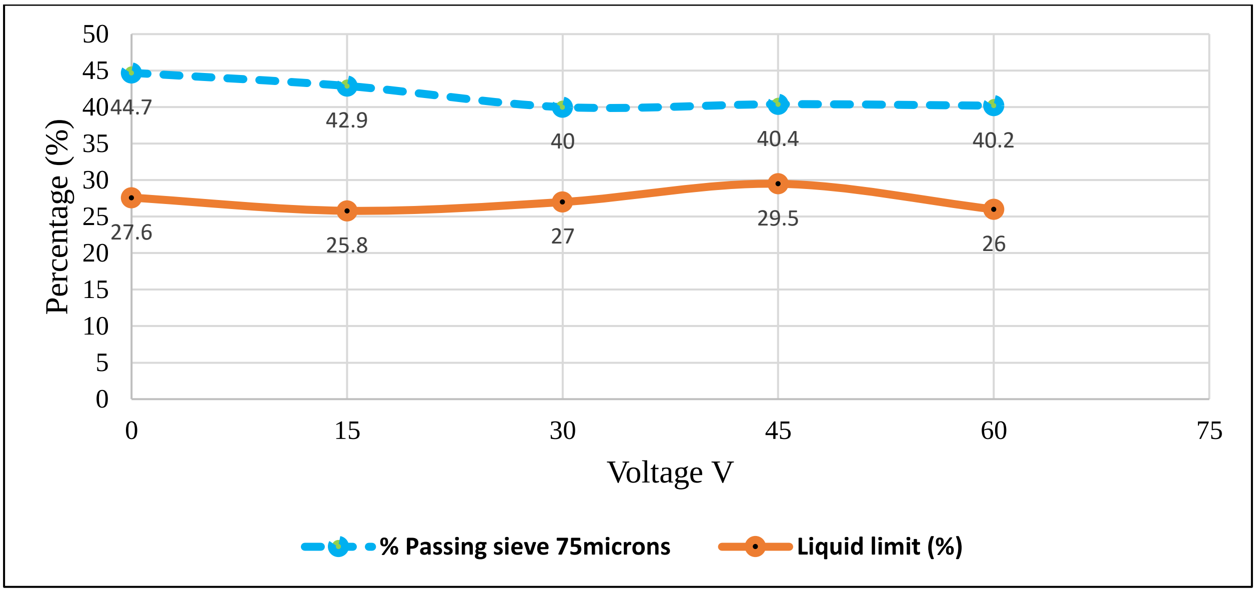

Figure 5: Summary of Sieve Analysis and Liquid Limit Test Results

The particle size distribution curves and liquid limit graph are shown in Figure 5 above. Percentage of soil fraction passing sieve 75micron for the unremediated soil was 45%, this slightly decreases to 40% when 30V, 45V and 60V were applied. The decrease is attributed to the removal of the combined particles of lead and copper which are desorbed from the soil during the remediation process thereby reducing the finer particles of the remediated soil. This is consistent with the findings of Sani et al. (2023). The atterberg limit test have liquid limit

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ranging between 26% and 29.5%. The soil is classified before and after remediation as an A-4 group of soil according to AASHTO classification system exhibiting clay of low plasticity CL or OL group according to the Unified Soil Classification System.

Table 3: Variation of Maximum Dry Density and Optimum Moisture content with Voltage at Different Compactive Efforts

	MDD g/cm^3			OMC (%)		
	BSH	**WAS**	**BSL**	**BSH**	**WAS**	**BSL**
0	1.9050	1.8390	1.8000	10.3	11.4	12.0
15	1.9010	1.7960	1.8450	10.0	10.5	11.1
30	1.9200	1.8600	1.8500	9.8	10.2	11.0
45	1.9140	1.8560	1.8400	9.9	10.6	11.8
60	1.9150	1.8480	1.8320	9.8	11.0	11.6

As shown in table 3, there was general improvement in the maximum dry density with the highest value recorded at 30V across all the three compactive efforts and at the lowest average optimum moisture content of 10.0%. The slight increase in the MDD is as a result of the physico-chemical changes and its consequent influence on the diffused double layer leading to the arrangement, grouping, orientation of the particles and pore spaces within the soil mass and soil fabrics agreeing with the work of Sani et al. (2023).

Table 4: Variation of Soaked and Unsoaked CBR with Voltage at different compactive efforts

	Unsoaked CBR (%)			Soaked CBR (%)		
Voltage	**BSH**	**WAS**	**BSL**	**BSH**	**WAS**	**BSL**
0	41	24	21	16	8	7
15	42	24	20	16	8	7
30	51	28	22	15	8	8
45	52	27	22	16	8	9
60	58	30	23	16	10	8

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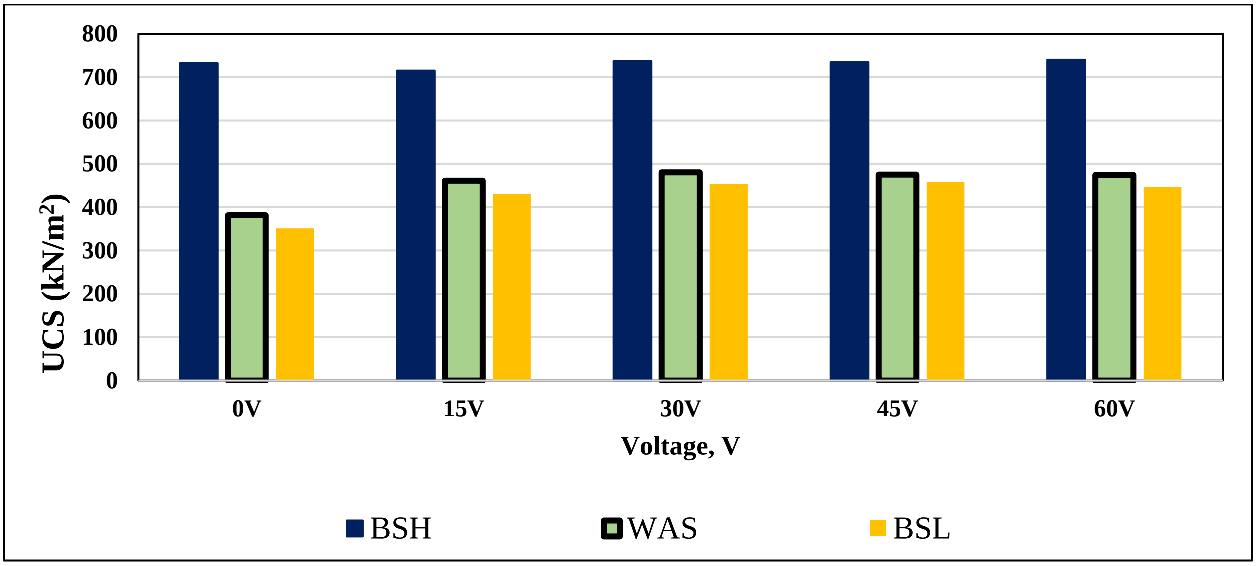

Figure 6 Variation of UCS with Voltage at different compactive efforts

As indicated above in figure 6, the UCS remain consistent when BSH compaction method was used across all the condition of voltage applied. In contrast, there was increase in the UCS when WAS and BSL compaction methods were used at 30V and 45V. This increase can be attributed to changes in the particle packing of the mixture due to the electrical charges induced. The new chemical properties exhibited by the remediated soil due to the double layer water presents in the clay mineral structure could also be responsible for the increase in the UCS. However, this improvement increased the soil inter-particle force that binds them together and the formation of aggregates, clods, and lumps or otherwise called crumbs, which agrees with literature.

Table 5: Resistance to loss in strength (%)

	BSH (kN/m^3)	WAS (kN/m^3)	BSL (kN/m^3)
Unremediated	71	77	62
15V	77	83	76
30V	89	82	75
45V	90	83	77
60V	76	85	77

Table 5 shows resistance to loss in strength in the soil before and after remediation. There was general increase in the resistance to loss in strength across all the three compactive efforts, at 30V and 45V, the durability index with resistances of 89% and 90% to loss in strength was recorded respectively when BSH was used. This, when compared to the resistance to loss in strength of 71% in unremediated soil has respectively 25.3% and 26.8% durability advantages.

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CONCLUSION

- Lead removal efficiency was highest at 2.0V/cm (86%) with the shortest remediation time of 5days and lowest at 0.5V/cm (39%) at 9 days. In contrast, 52% of copper was removed at 2.0V/cm in 5days and 29% at 0.5V/cm after 9days of remediation. At 1.0V/cm, the lead and copper removal efficiency are 75% and 40% respectively.
- Percentage of soil fraction passing sieve 75micron for the unremediated soil was 45%, this slightly decreased to 40% when 30V, 45V and 60V were applied.
- The CBR values increases with increase in voltage, these values peaked at 1.5V/cm and 2.0V/cm for both soaked and unsoaked samples. At 30V, the average unsoaked CBR value of 34% was achieved across the three compactive efforts compared to 28.7% for the unremediated soil.
- There was increase in the UCS when WAS and BSL compaction methods were used. The unremediated UCS of 381kN/m^3 increased to 474kN/m^3 at 60V for WAS compaction method and from 351kN/m^3 unremediated to 447kN/m^3 at 60V for BSL compaction method. This shows and improvement in the unconfined compression strength.
- When BSH compaction method was used, there was 25.3% and 26.8% durability advantages over the unremediated soil at 1.0V/cm and 1.5V/cm respectively.
- The soil is classified before and after remediation as an A-4 group of soil according to AASHTO classification system exhibiting clay of low plasticity CL or OL group according to the Unified Soil Classification System.
- There was an improvement in the geotechnical properties of the remediated soil at 1.0V/cm and 1.5V/cm beyond which there was no more significant changes.

RECOMMENDATION

For low permeability soil, Lead (Pb) and copper (Cu) are best removed from contaminated soil at high voltage with shorter remediation period. It is however recommended that applying 1.0V/cm and 1.5V/cm would yield better results in terms of improving the geotechnical properties of the remediated soil.

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CONFLICT OF INTEREST

No conflict of interest for both authors.

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