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Effect of Varying Voltage on Electro-Kinetic Remediation of Crude Oil Contaminated Soil

Sani J. E.[1], Moses G.[1], Ijimdiya T.S.[2], Adebayo K.[1], Imie E.[1] and Ige O. O.[3]

[1]Department of Civil Engineering, Nigerian Defence Academy, Kaduna, Nigeria.
[2]Department of Civil Engineering, Ahmadu Bello University, Zaria, Nigeria
[3]Department of Physics, Nigerian Defence Academy, Kaduna, Nigeria

Corresponding Author's Email:
ibramabdul@gmail.com

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Abstract

Electro-kinetic remediation (EKR) is a soil remediation technology that has been found to be effective in treating soils contaminated with various types and amounts of pollutants. However, research has shown that EKR can be improved by several strategies for more effective performance. This study investigated the effect of varying voltages on the electro-kinetic (EK) remediation of crude oil-contaminated soil. Crude oil-contaminated soil (COCS) was obtained from a polluted area near an oil field in Port Harcourt, Nigeria. During the remediation process, voltage was varied at 15V, 30V, 45V, and 60V, with volt gradients of 0.5, 1.0, 1.5, and 2.0V/cm, respectively. Throughout the experiment, 0.05 ml of sodium dodecyl sulphate (SDS) and 0.1 ml of sodium sulphate ($NaSO_4$) were used as electrolytes for both catholyte (base) and anolyte (acid), respectively. The total petroleum hydrocarbon (TPH) content of the COCS was 80,000 mg/kg. This value exceeds the permissible limits for soils and thus needs to be remediated before being used for any engineering purpose. The average removal efficiency after EK remediation improved progressively for EKR-15V, EKR-30V, EKR-45V, and EKR-60V at 27%, 49%, 56%, and 77%, respectively. The electrokinetic remediation enhanced the formation of more oxides and increased their concentrations. Although EKR-60V had the highest remediation efficiency, for all the EKR soils, the silica content increased while calcium oxide (CaO) increased for only EKR-30V and EKR-45V; hence, soil strength improved due to enrichment to become a silicate laterite. Hence, for more efficiency in EKR technology, higher voltage should be incorporated for remediating COCS.

Keywords: Voltage gradient, contaminated soil, Total Petroleum Hydrocarbon (TPH), Electrokinetic remediation and Removal efficiency.

1.0 Introduction

Contaminated soil is a tremendous global issue due to the rapid industrialization. Soil and groundwater pollution is escalating globally, resulting in significant environmental challenges. Various human activities, including agriculture, mining, and industrialization, contribute to soil contamination. Contaminants can take the form of organic compounds, inorganic substances, and heavy metals. Among the common soil pollutants are heavy metals and petroleum-based hydrocarbons. Additionally, many soils bear the burden of petroleum contamination due to oil spills during production and transportation (Cameselle *et al.*, 2013; Adebayo et al, 2023; Sani *et al.*, 2023; Haruna et al, 2023).

Petroleum exploration and production can cause soil pollution worldwide, which affects the environment and human health (Kvenvolden and Cooper, 2003; Adebayo *et al.*, 2023; Sani *et al.*, 2023). Inadequate waste disposal, accidental tank leakages, and spills can lead to the entry of petroleum and oil products into the natural environment (Huguenot *et al.*, 2015). Petroleum can disperse, evaporate, sink, and migrate into the soil and groundwater in significant amounts after an accident (Ochoa *et al.*, 2016; Adebayo et al, 2023).

In Nigeria, oil extraction and processing activities have resulted in massive land contamination (Sam, 2016; UNEP, 2011). The Niger Delta region of Nigeria has been severely impacted by oil extraction and processing activities, leading to significant land pollution (Sam, 2016). The increasing dependence of the Nigerian economy on hydrocarbon exploration and extraction has led to severe pressures on the environmental components and other receptive systems (Ite *et al.*, 2013), resulting from the accidental and incidental discharge of hydrocarbons and their products into the environment. There is increasing concern as large volumes of toxic organic substances continually enter the coastal environment of the Niger Delta (Eregha and Irughe, 2009; Linden and Palsson, 2013) through different routes, including leachate and seepage during operations, extraction, transportation, distribution storage, and refining (UNEP, 2011).

In recent times, there has been a growing interest in the remediation of soil and water contaminated with petroleum. The petrochemical activities of industries have led to significant environmental problems in water and soil ecosystems (Lopez-Vizcaino *et al.*, 2014; Sani et al, 2023; Haruna et al, 2023).

Electro-kinetics remediation (EKR) has emerged as one of the most versatile technologies for soil remediation over the past three decades because of its suitability to treat both inorganic and organic pollutants as well as radionuclides, saturated and unsaturated soil matrices, low-permeable soil, and heterogeneous soil layers (Oonnittan 2012; Adebayo et al, 2023; Sani et al, 2023; Haruna et al, 2023).

EKR is a newly developed technique, especially acknowledged for strengthening low-permeable soil. The electric potentials are applied to fluid transport in porous media and the physical and chemical transport of charged particles (Mosavat *et al.*, 2012). It refers to applying a low direct current (DC) or pulsed electric field connected through a pair of electrodes. In electro-kinetic remediation (EKR), the electrodes are inserted vertically into the soil. When an electric current is applied, three phenomena occur: electroosmosis, electromigration, and electrophoresis. These phenomena cause the ions to move towards the electrodes, as shown in Fig. 1 below (Cameselle *et al.*, 2013; Sani et al, 2023). Contaminants in the aqueous phase, or those desorbed from the soil surface, are transported towards the respective electrodes based on their charge. The contaminants can then be extracted into a recovery system or deposited at the electrode. Surfactants and complexing agents can be used to increase solubility and assist in the movement of the contaminant.

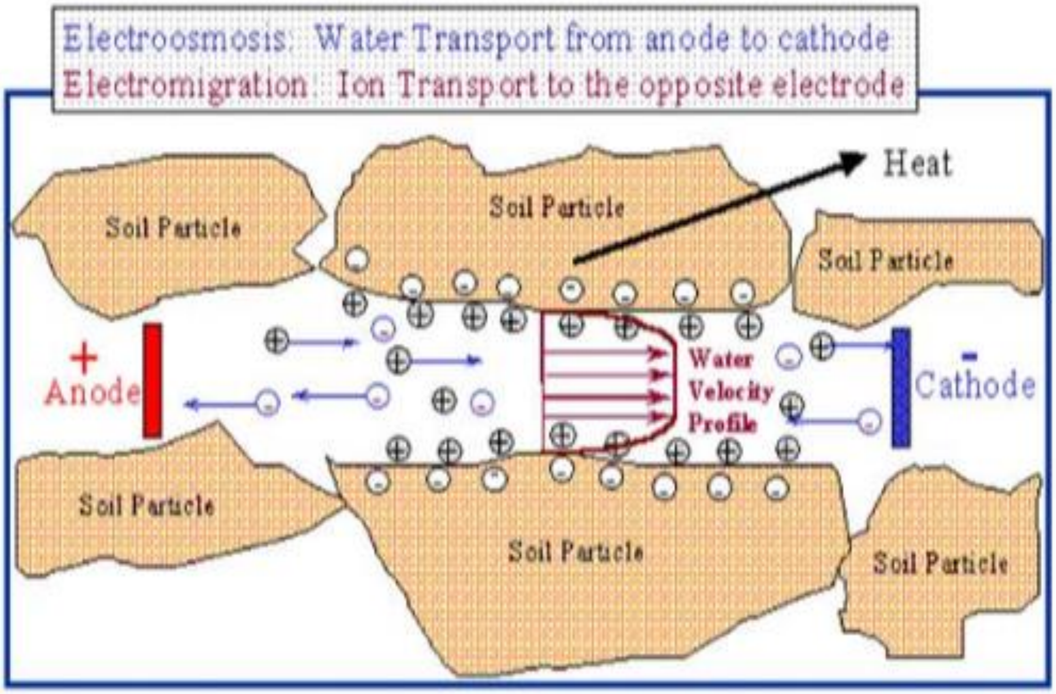


Fig. 1 Illustration of electro-kinetic phenomenon during electro-kinetic process (Cameselle *et al.*, 2013)

Electro-kinetics Remediation (EKR) has been tested in numerous field-scale tests and has been found to be commercially viable and technically effective when compared to other commercially available methods. The ability of EKR to enhance the removal process by various mechanisms will be shown to demonstrate its effectiveness in ground remediation technology for crude oil-contaminated soil.

Petroleum hydrocarbon compounds (PHC) are present in products such as fuels, oils, lubricants, waxes, and others and can be quantitatively expressed as total petroleum hydrocarbon (TPH) (Egedeuzu and Nnorom, 2013). The limit for organic contamination in soil is typically between 1000 and 2000 mg/kg, but diesel hydrocarbon contamination could be 1000 mg/kg (Egedeuzu and Nnorom, 2013). When the risk level of soil and/or groundwater contamination is unacceptable, remedial actions are recommended. Remediation technologies are measures adopted to control, reduce, mitigate, or eliminate the risk resulting from contamination of the soil and/or groundwater media. Selected approaches are needed to be efficient and cost-effective in achieving the remedial goals (Sharma and Reddy, 2004). Worldwide remediation technologies like soil washing (Dermont *et al.*, 2008), bioremediation (Virkutyte *et al.*, 2002), thermal desorption (Choi et al., 2020), soil vapor extraction (Virkutyte *et al.*, 2002), and soil flushing (Ng et al 2015) have been experimented with limited results. According to the United States Environmental Protection Agency (USEPA) in 2000 and 2006, remediation technologies have shown inadequate performance in remediating complex site conditions. These conditions include clayey soils due to their low permeability and complex composition; heterogeneous subsurface conditions (e.g., clay lenses within sand formation); organic contaminants that are hydrophobic; heavy metal contaminants; and mixed contaminants (e.g., organic compounds combined with heavy metals and/or radionuclides). The challenges of soil remediation include difficulty accessing the contaminants, high electrical conductivity, and exchangeable sodium percentage. This affirms an urgent need to develop new technologies that can overcome these challenges as well as be cost-effective (Reddy *et al.,* 2006). Muhsina and Chandrakaran (2015) used an unenhanced electrokinetic (EK) setup to remove 80% and 45% of contaminants at the cathode and anode, respectively, from an oil-contaminated clayey soil after 18 days by using a 0.6 V/cm-specific voltage. The overall efficiency of the treatment and restoration of properties was in the range of 65-75%.

The above information suggests that crude oil content in soils can exceed permissible limits, posing dangers to humans and the environment. Electrokinetic technology can be used to remediate contaminated soils. However, there has been little to no research on the effect of varying voltage on the electrokinetic remediation of oil-contaminated soil. This research therefore evaluates the effect of voltage variation on enhancing electrokinetic remediation of crude oil-contaminated soil.

2.0 Materials and Methods

2.1 Materials

The material used for the research is crude oil-contaminated soil (COC), gotten from a crude oil-polluted area near an oil field (Latitude $4^{o}\ 50^{'}\ 19^{''}$ and Longitude $7^{o}\ 4^{'}$ 18”) in Port Harcourt, Nigeria. The topsoil was removed with a locally made hoe, hand trowel, and shovel to a depth of 0.2m to clear off the vegetable soil. The contaminated soil was then dug to a depth of about 0.6–1 m, covering a total area of over 1.81 m^2.

2.2 Methods

2.2.1 Laboratory Experiment

Laboratory Experiment Laboratory tests were carried out on crude oil-contaminated soil (COC) and electro-kinetically remediated soil (EKR). These tests were done before and after treatment. During the remediation process, voltage was varied at 15V, 30V, 45V, and 60V; hence, their volt gradient is 0.5, 1.0, 1.5, and 2.0V/cm, which is represented in this study as EKR-15V, EKR-30V, EKR-45V, and EKR-60V, respectively. See Table 1 below. Table 1:

Table 1: Experiment work Plan

No.	Test's Code	Voltage	Voltage gradient	Purging solution at Anode compartment	Purging solution at Cathode	Mode of the Electric field	Remediation Duration (Days)
1	EKR 1	15 V	0.5 V/cm	0.1 M Na_2SO_4	0.05 M SDS	Continuously	31
2	EKR 2	30 V	1 V/cm	0.1 M Na_2SO_4	0.05 M SDS	Continuously	26
3	EKR 3	45 V	1.5 V/cm	0.1 M Na_2SO_4	0.05 M SDS	Continuously	9
4	EKR 4	60 V	2 V/cm	0.1 M Na_2SO_4	0.05 M SDS	Continuously	14

Voltage Gradient Ranges between 0.5 and 2 VDC/cm, as shown in the summary of the experiment work plan in Table 1. The operation was terminated only when the effluent rate at the cathode was significantly low (zero) and the fluctuating current remained constant for days. At the end of the process, the remediated soil was sliced into five equal parts and air-dried. The TPH and chemical oxide at each slice were determined to ascertain the level and variation in the remediation of the contamination carried out via varied voltage on the soil.

2.2.2 Electro-kinetic Cell

The electrokinetic remediation setup used in this research was based on the model adopted by (Yu *et al.*, 2019), and it’s in line with Cameselle et al (2013) recommendation of a simple electrokinetic cell setup. The electro-kinetic remediation cell was set up as shown in Fig. 2.

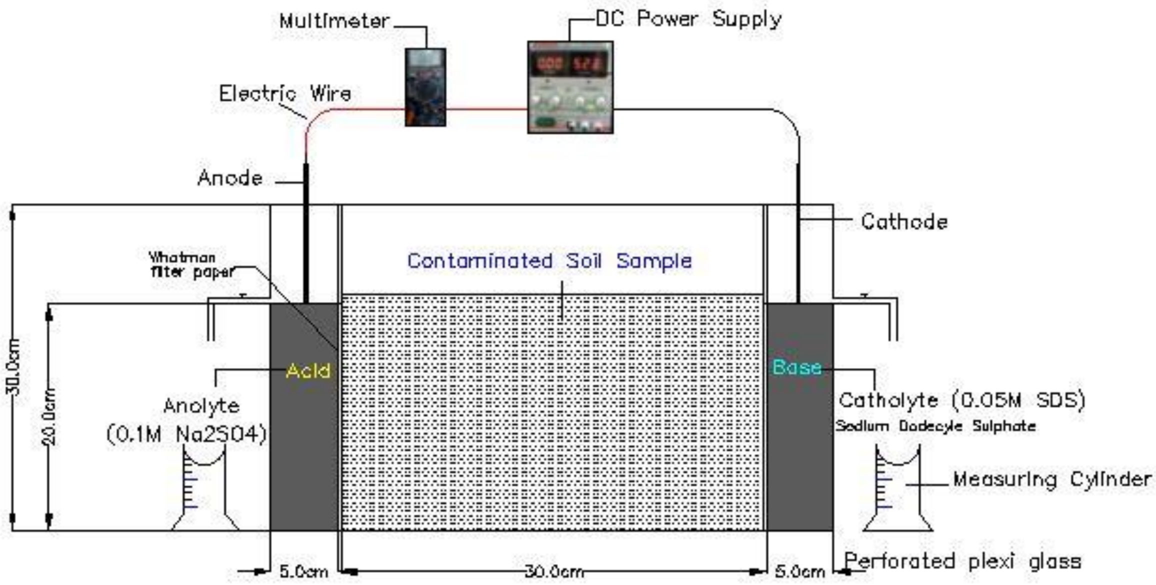


Fig. 2: Electro-kinetic cell set up

2.2.3 Electro-kinetic Test Procedure

The middle partition of the setup was filled with the contaminated soil up to 17kg at optimum moisture content. The height of the soil was 250mm, while 0.05 mL of sodium dodecyl sulphate (SDS) and 0.1 mL of sodium sulphate (NaSO4) were used as electrolytes as both catholyte (base) and anolyte (acid), respectively, throughout the experiment (Gholamreza and Milad 2018). Each electrolyte compartment has a volume of 1,065 mL in its reservoirs, as shown in Fig. 2 above. Two electrodes were each inserted into these chambers, connected with a wire having a contact point from a 100-volt DC power supply with varying voltages at 15V, 30V, 45V, and 60V, respectively. Four set-ups with the respective varying voltages were remediated differently, as shown below in Fig. 3. These set-ups were powered by an installed solar system for a constant supply of power.

Fig. 3: EK system with DC Supply and a solar-powered system

The voltage gradient ranges between 0.5 and 2 VDC/cm, as shown in the summary of the experiment work plan in Table 1 above. A multimeter was used to monitor both the voltage and current supplied through the system. Each end of the electrode chamber has a drilled hole of 10mm diameter to allow the passage of electro-osmotic flow (EOF) that occurs in the system, and this effluent is collected at the base of the cathode compartment and its volume recorded daily. A pH meter was used to measure the pH condition of the effluent daily during purging, while a multimeter was used daily to take records of current flow in the system. The electrokinetic cell was allowed to run for a minimum of nine to a maximum of thirty-one (9 to 31) days. This depend on the effluent or flow rate of EOF during the experiment. The operation was terminated only when the effluent rate at the cathode was significantly low (zero) and the fluctuating current remained constant for days. At the end of the process, the remediated soil was sliced into five equal parts and air-dried. The TPH and chemical oxide at each slice were determined to ascertain the level and variation in the remediation of the contamination carried out via varied voltage on the soil.

The removal efficiency (Re) of the charcoal and active carbon was calculated from Cho et al., 2020:

$$\text{Re}\ (\%) = \left(\frac{C0 - C}{C0}\right) \times 100$$

Where;

C_0 is the initial contaminant concentration in soil (mg/kg) and

C is the residual concentration of contaminant in soil after the treatment (mg/kg).

3.0 Results and Discussion

3.1 Effect of Varying Voltage on the Electrokinetic Remediation of COCS

The results obtained from all the laboratory experiments carried out on the crude oil-contaminated soil (COCS) and the EK-remediated soil by varying the voltage from 15V to 60V and checking its removal efficiency and the engineering properties of the soil about its suitability for waste containment facilities are summarized in the tables and figures below. The graph showing the accumulated EOF with time during EKR is shown in Fig. 4. For all voltage considered the EOF reduces with time in hours. Electrical current variation on the other hand is presented in Fig. 5 which shows an increase in current before it become steady indicating end of remediation. The graph generally shows that the increase in voltage leads to reduction in remediation time. Summary of Total Petroleum Hydrocarbon (TPH) of the Soil Sample is presented in Table 2.

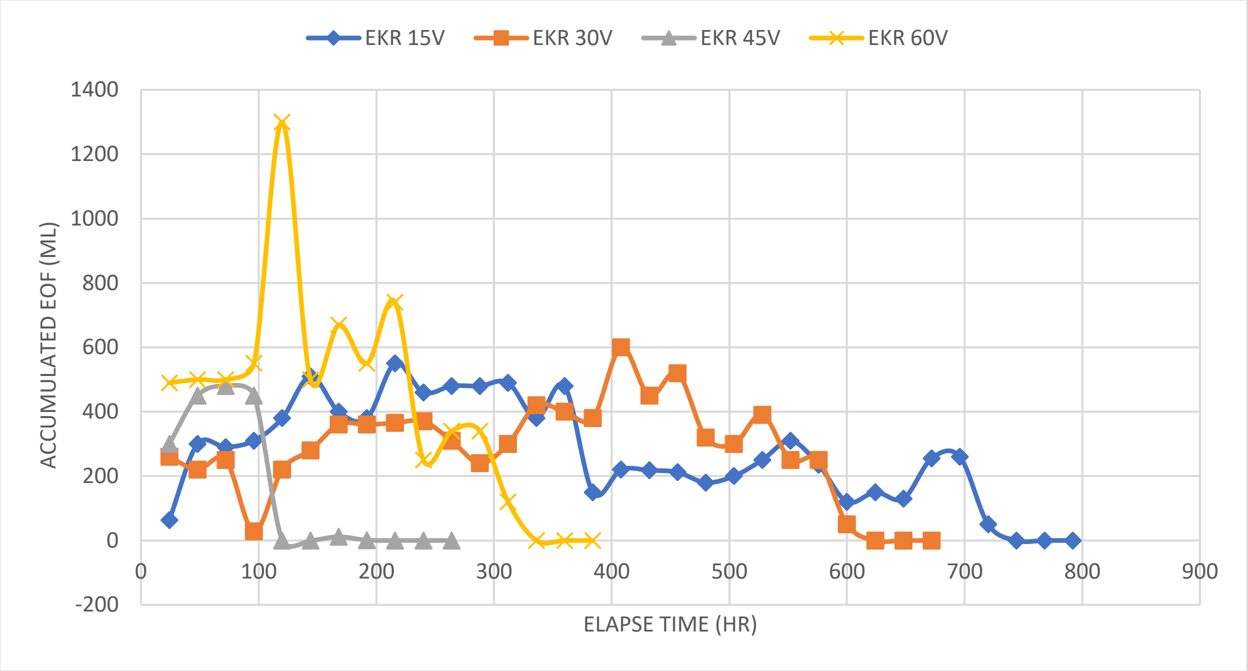


Fig.4: Accumulated Electro-osmotic flow (EOF) with Time during Electro-kinetic Remediation

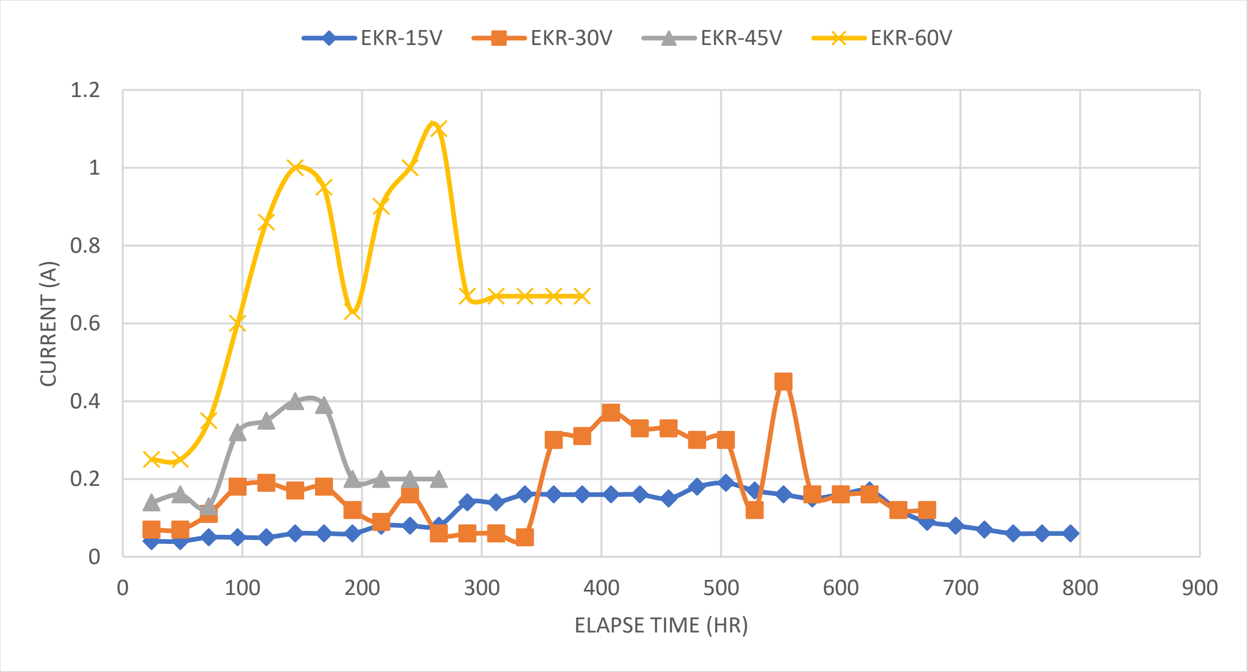
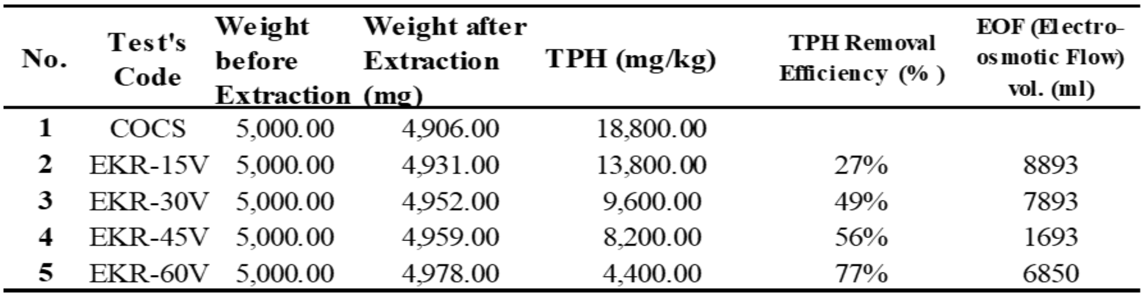


Fig. 5: Electrical Current variation with Time during Electro-kinetic Remediation

Table 2: Summary of Total Petroleum Hydrocarbon (TPH) of the Soil Sample



No.	Test's Code	Weight before Extraction	Weight after Extraction (mg)	TPH (mg/kg)	TPH Removal Efficiency (%)	EOF (Electro-osmotic Flow) vol. (ml)
1	COCS	5,000.00	4,906.00	18,800.00		
2	EKR-15V	5,000.00	4,931.00	13,800.00	27%	8893
3	EKR-30V	5,000.00	4,952.00	9,600.00	49%	7893
4	EKR-45V	5,000.00	4,959.00	8,200.00	56%	1693
5	EKR-60V	5,000.00	4,978.00	4,400.00	77%	6850

3.2 Electrokinetic Remediation Efficiency

In this research, to determine the remediation efficiency, Total Petroleum Hydrocarbon (TPH) was conducted by the gravimetric method (the Toluene cold extraction method) on the soil samples.

From the results obtained as shown in Table 2, the remediation efficiency showed a progressive increase as the voltage increased from TPH percentage removals for EKR-15, EKR-30, EKR-45, and EKR-60 of 27%, 49%, 56%, and 77%, respectively. See Fig. 6 below

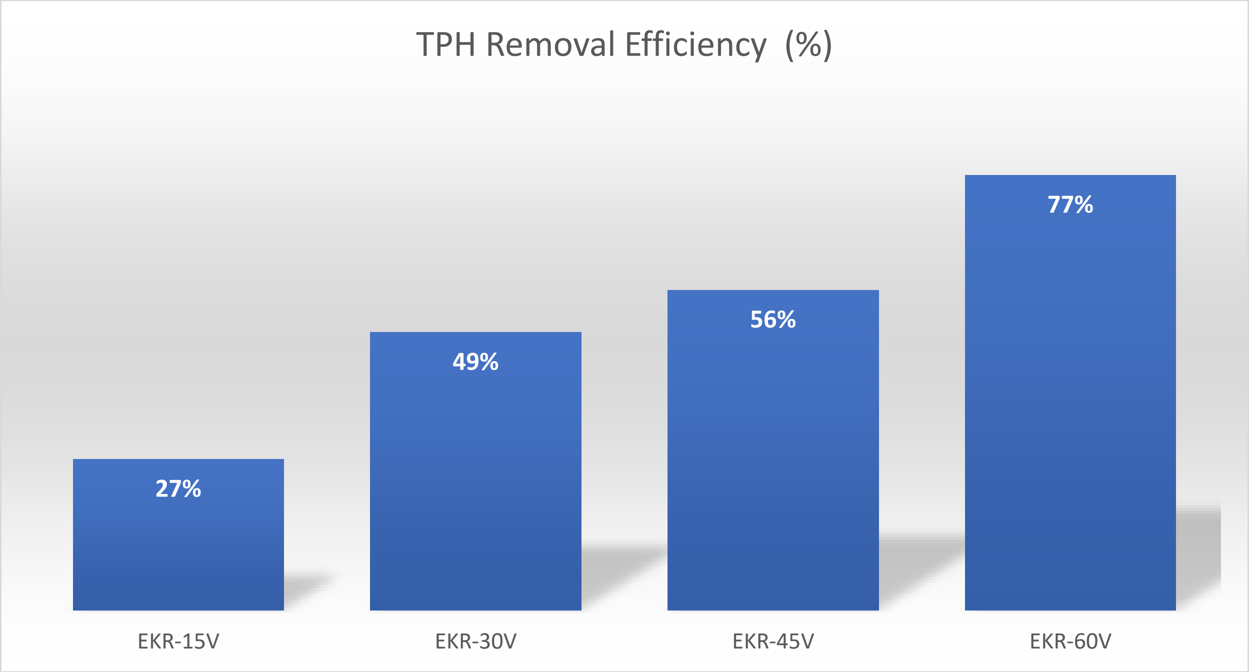


Fig. 6: Remediation Efficiency (%) of EKR soil.

3.3 pH of Electro-kinetic Remediated Soil

After the completion of the EKR operation, the system was disconnected, and the soil sample was carefully extruded from the EK cell and divided into five parts, from the anode to the cathode, and mapped as S_5, S_4, S_3, S_2 and S_1, respectively. A small portion was taken from each section (S_5-S_1) for a pH test, as plotted in Figure 7 against each section, respectively, from the anode to the cathode. The remaining soil samples were thoroughly mixed and taken for post-remediation physiochemical analysis and remediation efficiency testing.

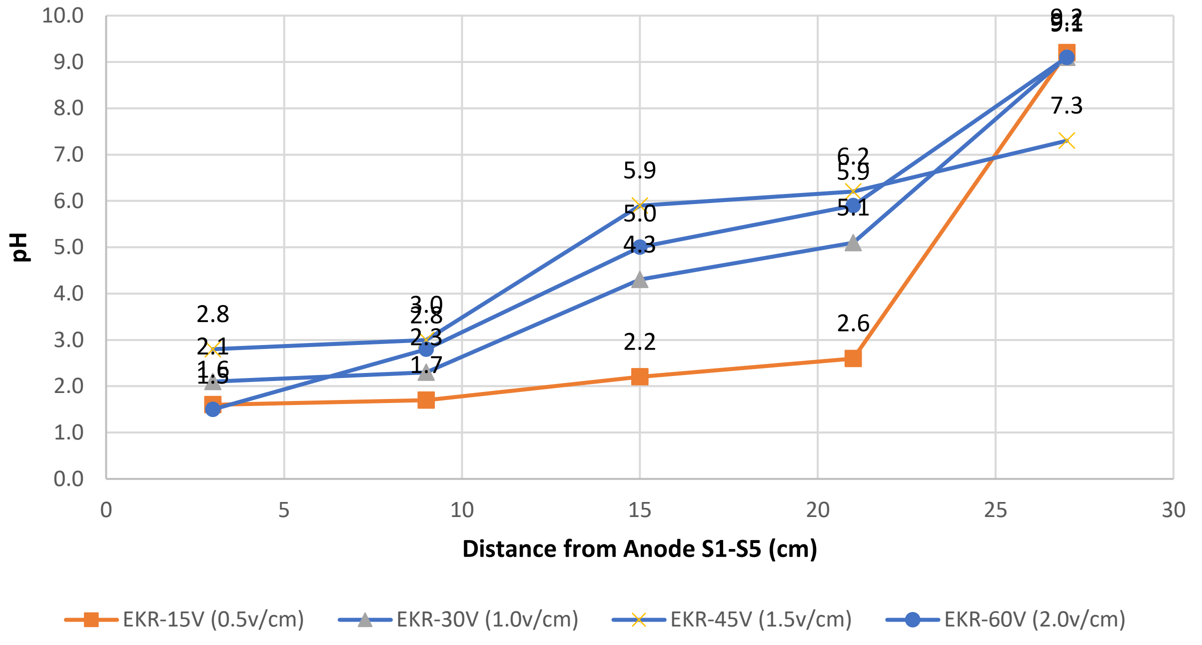


Figure 7: pH of soil section at the completion of EKR

The soil sections are identified as below:
S1: @60mm from the anode; S2 : @120mm from the anode; S3: @180mm from either the cathode or anode; S4 :@240mm from the anode; S5 :@300mm from the anode.

This change in pH from the cathode to the anode, or better explained as pH changes in the EKR soil, resulted from the chemical reactions (oxidation and reduction) called electrolysis that occurred in the two electrode compartments where water got oxidized to generate hydrogen ions H^+at the anode (acidic front) and hydroxide ions (OH^-) at the cathode (basic front). This led to ion migrations, which triggered pH changes in the soil (Fatemeh *et al.*, 2021; Jayasekera, 2015). This caused the pH of the COC soil to increase, showing that the soil properties changed from being moderately acidic to neutral.

3.4 Oxide composition of crude oil contaminated (COC soil) and electrokinetic remediated soil (EKR soil)

The oxide concentration of crude oil-contaminated soil (COCS) and the electrokinetic composition of both the COC soil and the EKR soil were determined using the X-ray florescence analyzer. The percentage oxide composition of both the COC soil and the EKR soil is shown in Table 3 below. From the oxide concentration, the ratio of silicon oxide to aluminum oxide (SiO_2/Al_2O_3) and the ratio of silicon oxide to iron oxide are more than 2.0 for both COC soils and EK-remediated soils, which implies non-lateritic soil. For all the EKR soil, the silica content increased while calcium oxide (CaO) increased for only EKR-30V and EKR-45V; hence, soil strength improved due to enrichment to become a silicate laterite. As shown in Figure 8 below. It is worthy of note that one of the basic constituents' elements that makes up the pozzolanic property that generates reactions into the formation of cementitious properties is responsible for the improved EKR soil strength.

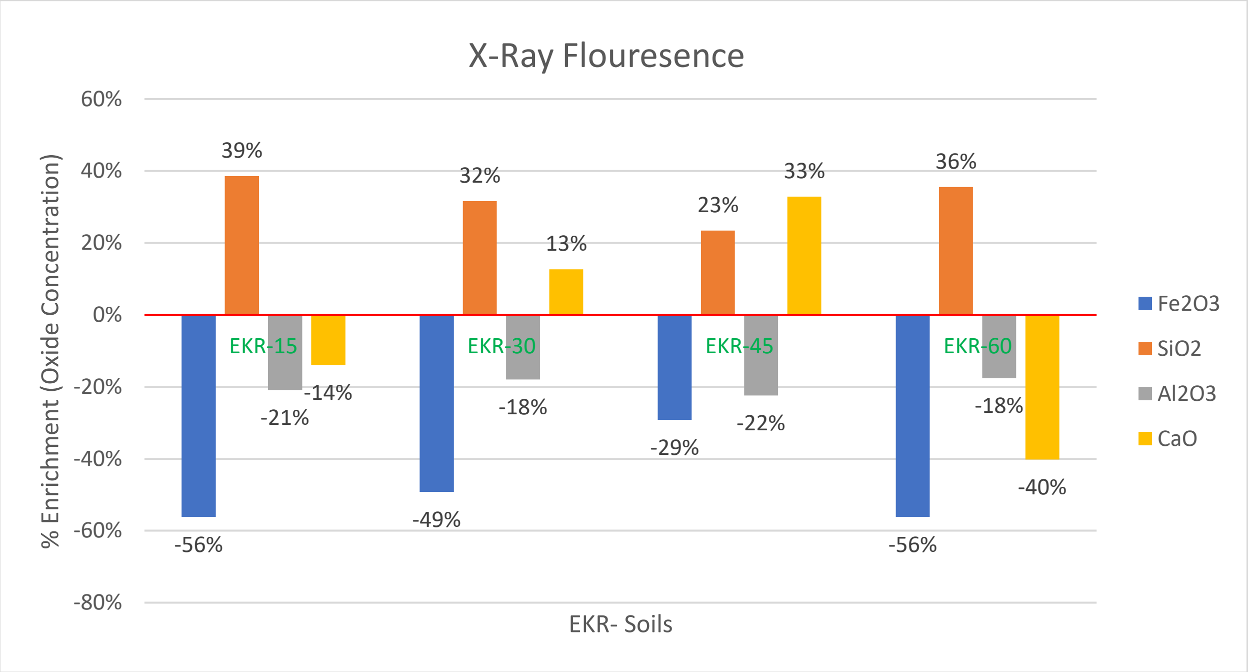


Figure 8: X-Ray Fluorescence for COCS and EK Remediated Soil

Table 3: Oxide concentration of crude oil contaminated soil (COCS) and the electrokinetic

S/N	OXIDEs	COCS (%)	EKR Soil (%)			
		0V	EKR-15V	EKR-30V	EKR- 45V	EKR-60V
1	Fe_2O_3	20.337	8.923	10.331	14.405	8.912
2	SiO_2	47.022	65.145	61.880	58.041	63.733
3	Al_2O_3	25.679	20.310	21.071	19.926	21.164
4	MgO	0.000	0.000	0.000	0.000	0.000
5	P2O5	0.489	0.294	0.226	0.170	0.267
6	SO3	0.415	1.365	2.187	1.552	2.379
7	TiO2	3.173	1.763	2.036	2.853	1.810
8	MnO	0.109	0.038	0.047	0.067	0.030
9	CaO	0.632	0.544	0.712	0.840	0.378
10	K2O	0.475	0.394	0.347	0.445	0.232
11	V2O5	0.187	0.105	0.139	0.173	0.085
12	Cr2O3	0.035	0.063	0.017	0.080	0.047
13	Co_3O_4	0.090	0.043	0.041	0.034	0.044
14	NiO	0.007	0.001	0.001	0.013	0.000
15	CuO	0.049	0.053	0.022	0.087	0.052
16	$Nb2O_3$	0.374	0.016	0.024	0.034	0.021
17	Ta2O5	0.017	0.043	0.018	0.031	0.061
18	ZnO	0.018	0.005	0.013	0.030	0.011
19	Cl	0.525	0.665	0.654	0.947	0.605
20	ZrO_2	0.368	0.113	0.161	0.196	0.119
21	SnO2	0.000	0.000	0.000	0.000	0.000
22	MoO3	0.000	0.006	0.009	0.005	0.004
23	Wo3	0.000	0.006	0.035	0.000	0.010
24	BaO	0.000	0.062	0.000	0.000	0.000

4.0 Conclusion

The effect of varying voltage on electro-kinetic remediation of crude oil-contaminated soil has been duly investigated, and the results have been presented, analyzed, and discussed. From the study, the following conclusions are drawn:

1. The total petroleum hydrocarbon (TPH) content of the COC soil before remediation was 80,000 mg/kg. After EK remediation with varying voltage, it was observed that the remediation efficiency showed a progressive increase as the voltage increased from TPH percentage removals for EKR-15, EKR-30, EKR-45, and EKR-60 of 27%, 49%, 56%, and 77%, respectively.
2. The electrokinetic remediation enhanced the formation of more oxides and increased their concentration. From the oxide concentration, the ratio of silicon oxide to aluminum oxide (SiO_2/Al_2O_3)

and the ratio of silicon oxide to iron oxide are more than 2.0 for both COC soils and EK-remediated soils, which implies non-lateritic soil. Although, for all the EKR soil, the silica content increased while calcium oxide (CaO) increased for only EKR-30V and EKR-45V, hence soil strength improved due to enrichment to become a silicate laterite.

4.1 Recommendation

From the research conducted on the effect of varying voltage on electro-kinetic remediation of crude oil-contaminated soil, the following recommendations can be made:

1. An electro-kinetically remediated soil with varying voltage can be recommended, and it's economical for large-scale remediation technology.
2. To enhance the remediation performance efficiency, more research should be conducted on different varieties of electrolytes, closer electrode spacing, processing timeframes, and higher grades of electrodes when applied to crude oil-contaminated soil.
3. crude oil-contaminated soil can be remediated to achieve a higher efficiency of over 70% with a voltage up to 60 volts, and this improved soil will enhance suitable environmental conditions.
4. Increasing the applied voltage will reduce the duration of the of the process. It took 31 days to complete remediation at 15V; these were reduced to 26, 9, and 14 days for 30V, 45V, and 60V, respectively

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Conflict of Interest

No potential conflict of interest was reported by the author(s).

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