Remediation of Crude Oil Contaminated Soil Through Thermolysis and Electrokinetic Methods

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Abstract- This study investigates the remediation of crude oil-contaminated soil using thermolysis and electrokinetic techniques, focusing on their efficiency in removing contaminants and improving soil properties. Various remediation methods were tested, including thermal (T), electrokinetics (E), thermal-electrokinetics (TE), and electrokineticsthermal (ET). Among these, the electrokineticsthermal (ET) method demonstrated the highest efficiency, achieving a 39.4% removal of total petroleum hydrocarbons (TPH), with the lowest TPH removal of 18.9% using the thermal (T) method. The elemental analysis, conducted via X-Rav Fluorescence (XRF), indicated that oxygen and silicon were the most abundant elements, suggesting the presence of oxides and silicate minerals in the samples. The Maximum Dry Density (MDD) and Optimum Moisture Content (OMC) of the soil samples reveal moderate variations in moisture content across the treatments. The California Bearing Ratio (CBR) results indicated that the remediated soil samples, particularly those treated with thermal and electrokinetic combinations, exhibited enhanced load-bearing capacities, making them more suitable for construction applications compared to the untreated contaminated soil. The findings highlight the potential of integrated thermolysis and electrokinetics as effective remediation methods for improving the strength and environmental safety of contaminated soils.

Indexed Terms- Contaminated Soil, Electrokinetics, Thermal, Maximum Dry Density (MDD) and Optimum Moisture Content (OMC), California Bearing Ratio (CBR).

I. INTRODUCTION

The oil industry contributes significantly to the economy of countries that have the oil underground, which is why the exploration, production, refining,

transportation and consumption of petroleum products are increased each day. The poor management practices of hydrocarbons, accidents during production, transportation fuels and other processed products, and the bunkering have brought environmental problems due to which it has become apparent contamination of large areas of surface soil and the allocation of water bodies (Adeola et al., 2022). Soil as a non-renewable resource and a very important system which aids ecosystem survival, soil contamination and remediation has been obtaining serious considerations around the world (Ite et al., 2018). The Niger Delta has been reported as one of the most heavily oil-impacted regions in the world due to over five decades of oil exploitation activities, coupled with poor management practices that have led to the contamination of soil and groundwater resources (Sam et al., 2017). Since the inception of the Nigerian oil sector, 13 million tonnes of hydrocarbons have been reported as spilled in the Niger Delta as a result of sabotage, pipeline vandalism (individuals that break pipeline during oil theft), well blowout, and engineering failure (e.g. pipeline rupture). Considerable oil contamination of the land has been reported and recent estimates suggest that over 2000 land-based oil-contaminated sites exist (Adeola et al., 2022; Ite et al., 2018; Sam et al., 2017).

The challenge associated with contaminated soil and water is how to reclaim or finally dispose of the materials that have been reclaimed. Many methods that use chemical compounds to repair the soil have been used in recent years to recover the soil so that it can be reused again (Malekzadeh & Nalbantoglu, 2013). There are numerous remedial solutions available to clean up a hazardous waste site, but the technical difficulty, effectiveness, and prices of these options might differ significantly. Although traditional ground burial and land disposal are frequently affordable, they do not offer a long-term solution and are not always the best options (Yeung, 2017). For removing contaminants such as organics and inorganics from solid porous media, the most common ex-situ methods employed include soil washing, and ligand extraction. Ex-situ methods may not be technologically challenged that much; however, they suffer from several problems. Apart from the generic problems of any ex-situ process, i.e., the need to excavate the media and place it in an external reactor, the above-mentioned processes suffer from several disadvantages. The nature and the magnitude of the problems are ever changing, bringing new challenges and creating a constant need for developing newer and more appropriate technologies (Demcak & Balintova, 2015). There are four main conventional soil remediation techniques for the removal of various pollutants and contamination from soils. These techniques include: bioremediation, thermal soil remediation, air sparging, and encapsulation (Azhar et al., 2017).

Electrokinetic remediation (EK) is a technology that uses an electric field to remove contaminants from soil. It is a relatively new technology, but it has shown promise for remediating soil contaminated with crude oil. EK works by applying a direct current (DC) electric field to the soil. This creates a potential difference between two electrodes, which drives the movement of ions and water through the soil. This process, called *electrokinesis*, can be used to remove contaminants from soil in a number of ways, such as electromigration, electroosmosis, and electrophoresis. EK has been shown to be effective in removing crude oil from soil in a number of laboratory and field studies. In one study, EK was able to remove 90% of the crude oil from a contaminated soil within 30 days. In another study, EK was able to remove 70% of the crude oil from a contaminated soil within 60 days (Prakash et al., 2021). EK has a number of advantages over other remediation technologies, such as excavation and landfilling. It is a relatively noninvasive technology, and it can be used to remediate soil in situ. This means that the soil does not need to be excavated and transported to another location for treatment. EK is also a relatively energy-efficient technology (Han et al., 2021).

Pyrolysis is the thermal technology for decomposition of organic matter (in the absence of oxygen or inert atmosphere) into liquid, gases and char. Pyrolysis is a

technology that could be applied to extract thermally intact organic molecules or to crack large molecules from complex matrices, while cracking of the large organic molecule may form other by-products (Adeniran et al., 2023). Higher contaminant removal efficiency can be achieved in a shorter treatment time with thermal treatment such as pyrolysis. Pyrolysis operates at lower temperatures and anoxic atmospheric conditions compared to conventional thermal treatment methods, during which large organic molecules can be broken down into smaller molecules that can be more easily removed (Kang et al., 2020). In addition, pyrolysis can be applied to remove a wide range of pollutants and leaves the soil intact for future use, and for these merits, pyrolysis is receiving much attention as an important soil remediation method. In comparison, when dieselcontaminated soil was treated by pyrolysis at 250 °C, the concentration of contaminants was noticed to decreased from 6272 mg/kg to 359 mg/kg (i.e., ~95% removal). This research is aimed at evaluating the effect of thermal and electrokinetic remediation (EKR) as an effective method to remediate crude oil contaminated soil for use in road construction.

II. METHODOLOGY

2.1 Preparation of crude oil-contaminated soil Crude oil-contaminated soil was artificially prepared by vigorously mixing crude oil and soil at a ratio of 1:5 at known weights. The simulated crude oilcontaminated soil was stored in a vessel at room temperature for two weeks. The soil was later divided into three (3) parts and labelled as T: Thermal, E :Electrokinetics, E-T: Electrokinetics -Thermal, T-E: Thermal-Electrokinetics and finally CS: Contaminated Soil.

2.2 Remediation Techniques and Methods

Site characterization is often the first step in a contaminated remediation strategy. It consists of the collection and assessment of data representing contaminant type and distribution at a site under investigation (Moses et al., 2019). The results of a site characterization form the basis for decisions concerning the requirements of remedial action. Additionally, the results serve as a guide for design, implementation and monitoring of the remedial system. Each site is specific; therefore, site

characterization must be corrected to specific site requirements. An inadequate site characterization may lead to the collection of unnecessary or misleading data, technical misjudgement affecting the cost and duration of possible remedial action, or extensive contamination problems resulting from inadequate or inappropriate remedial action. Site characterization is often an expensive and lengthy process, therefore it is advantageous to follow an effective characterization strategy to optimize efficiency and cost (Sani et al., 2023).

2.3 Design of EK Testing Model

The Electrokinetic (EK) experimental model will be constructed for this study. The model was originally designed by Liaki (2006) but has been modified to achieve the objectives of this study. The rectangular model tank will be made of nonconductive glass to prevent short circuiting during electrokinetic treatment. The tank will consist of main compartment where soil samples for treatment will be placed and two small compartments for the anode and cathode.

2.3.1 Sample Preparation

The slurry sample will be prepared by mixing the contaminated soils with deionised water to achieved 90% water content. The water content of slurry has been chosen based on 1.5 times liquid limit (LL). Many researchers have used water content of 2 times liquid limit for their slurry preparation to produce homogeneous sample. However, according to Malekzadeh & Nalbantoglu, (2013) found that water content of 1.5 times LL caused no apparent detrimental effect on the uniformity of the sample. The slurry sample will then be mixed using a mechanical mixer and blended thoroughly for 30 minutes.

2.4 Pyrolysis operation

The feed (2 kg of prepared contaminated soil) will be charged into the reactor through the feeder. The pyrolysis is then started by gradually heating the reactor to temperatures between 400°C to 800°C and time of 4 to 5 hours. This will ensure the proper breakdown of hydrocarbons and contaminants. The processed soil is then allowed to cool and was labelled Sample T for further analysis. The pyrolysis process will involve two (2) samples: fresh prepared contaminated (Sample T) and the treated soil from the EK cell which will be labelled Sample ET.

2.5 Electrokinetic Experimentation Process

During electrokinetic experimentation, two different samples of the contaminated soil will be processed. The first sample will be the fresh prepared contaminated soil that will be treated in the EK cell whereas the second sample will be from the thermal (pyrolysis) process. The soil sample was placed in an electrokinetic cell between the two electrodes (anode and cathode). Distilled water was added as a suitable electrolyte solution, after which a direct current (DC) electric field of 2 V/cm was applied across the contaminated soil. After 48 hrs of operation, the treated soil was collected, and the contaminant removal efficiency was assessed.

III. RESULTS AND DISCUSSION OF RESULTS

3.1 Removal efficiency of contaminants

To determine the efficiency of contaminants removal for thermal, electrokinetics, thermalelectrokinetics and electrokinetics-thermal, and that of the contaminated soil, the total petroleum hydrocarbon (TPH) of the samples was analyzed using the gravimetric method and the result presented in Figure 1.



UCS: Uncontaminated Soil, E-T: Electrokinetics, Thermal, T-E: Thermal-Electrokinetics, E: Electrokinetics, T: Thermal, CS: Contaminated Soil,

Figure 1: Removal Efficiency of the methodsSamples.

3.2 Elemental analysis of the soil samples

The elemental analysis of the contaminated soil sample and remediated samples was determined by the use of X-Ray Fluorescence (XRF) and the results presented in Table 1

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Concentration							
Element	CS	Е	Т	ET	TE		
0	43.024	44.612	44.73	44,775	43.377		
Mg	0	0	0.179	0	0		
Al	7.047	8.172	6.852	8.362	6.415		
Si	21.613	24.197	24.923	24.169	22.494		
Р	0.282	0.2	0.316	0.301	0.223		
S	0.973	0.762	0.753	0.796	0.825		
Cl	1.445	1.04	0.835	0.951	0.852		
К	2.351	2.898	2.334	2.38	2.418		
Са	9.755	6.359	6.923	6.193	6.162		
Ti	1.257	1.325	1.168	1.113	1.246		
V	0.06	0.077	0.047	0.062	0.085		
Cr	0.122	0.051	0.057	0.059	0.024		
Mn	0.13	0.112	0.119	0.127	0.109		
Fe	10.822	9.356	9.757	9.567	14.753		
Со	0.079	0.052	0.03	0.038	0.07		
Ni	0.015	0.011	0.004	0.007	0.011		
Cu	0.097	0.106	0.194	0.099	0.111		
Zn	0.538	0.398	0.46	0.463	0.521		
Zr	0.261	0.151	0.228	0.195	0.177		
Nb	0.017	0.031	0.032	0.029	0.05		
Мо	0	0	0	0.002	0.028		
Sn	0	0	0	0.084	0		
Ba	0	0.041	0	0.102	0		
Та	0.107	0.05	0.061	0.107	0.049		
W	0.005	0	0	0.018	0		

Table 1: Elemental analysis of the soil samples using XRF

The concentration of oxygen is relatively consistent across all samples, ranging from 43.024 wt.% in sample CS as the lowest to 44.775 wt.% in sample ET as the highest. Oxygen is the most abundant element in the samples, likely due to the presence of oxides found in soils. Silicon concentrations range from 21.613 wt.% in sample CS to 24.923 wt.% in sample T. Silicon is another major component, indicative of silicate minerals like quartz, which are common in soil. Thus, its low in CS because it contains high contamination of crude oil. Aluminum levels vary between 6.415 wt.% in sample TE and 8.362 wt.% in sample ET. This suggests the presence of aluminosilicates, such as

clay minerals. Calcium shows a reduction in concentrations from 9.755 wt.% in sample CS to 6.162 wt.% in sample TE. The presence of calcium indicates carbonates or other calcium-bearing minerals in the soil samples. Iron is present in significant amounts, particularly in sample TE (14.753 wt.%). Iron oxides or hydroxides could be prevalent, which are common in soils, especially in contaminated or oxidized environments. Magnesium is present only in trace amounts, detected in sample T (0.179 wt.%). Potassium shows small variations, ranging from 2.351 wt.% in sample CS to 2.418 wt.% in sample TE, likely from feldspar or mica minerals.

Other trace elements such as V, Cr, Mn, Ni, Cu, and Zn also appear in the analysis. Vanadium (V) was detected in very low concentrations, with the highest at 0.085 wt.% in sample TE. Vanadium is often present in crude oil as an undesired metal.

3.3 Atterberg Limits (LL, PL, PI, and SL)

Figure 2 shows the Atterberg Limits (LL, PL, PI, and SL) of the treated soil samples (E, T, E-T and T-E) and contaminated soil. The Liquid Limit (LL) indicates the moisture content at which soil changes from a plastic to a liquid state. Sample T is the most stable, with a high PL (37.05%), a moderate PI (4.95%), and the highest SL (2.86%), making it the least prone to deformation and shrinkage. Sample T-E has the lowest PI (3.12%), indicating minimal plasticity and excellent stability under load, with a good balance of workability and shrinkage potential with moderate SL (1.43%). Sample E shows the highest LL (42.5%) as well as highest plasticity with PI of 9.94%, making it potentially less stable under varying moisture conditions. Whereas Sample E-T has the lowest LL (33%) and PL (27.25%), indicating that it might be more brittle when dry but still offers some stability with a moderate SL (2.14 %). Contaminated Soil (CS), exhibits a moderate PI (5.79 %) and very low SL (0.71%), suggests it is prone to instability, particularly in terms of shrinkage, which could be problematic for construction applications.



Figure 2: Atterberg Limits (LL, PL, PI, and SL) for the treated soil samples (E, T, E-T and T-E) and contaminated soil.

3.4 Sieve analysis results

Table 2 presents the sieve analysis results for soil samples. The table present the percentage of soil passing through various sieves: No. 7 (2.36 mm),

No. 36 (0.425 mm), and No. 200 (0.07 mm). These results help in understanding the particle size distribution within the soil samples, which is crucial for determining the soil's textural class, permeability, compaction characteristics, and suitability for construction purposes. The grain size distribution curve is shown in Figure 3.

			Sieving %		
Soil	passing				
Sample	No. 200	No. 36	No. 7	3/4"	
	0.07mm	0.425mm	2.36mm	19mm	
Е	1.43	54.56	99.06	100	
Т	2.86	57.46	94.34	100	
E-T	2.14	55.26	94.48	100	
T-E	1.43	46.56	93.12	100	
CS	0.71	79.38	98.12	100	

Table 2: Sieve analysis results for soil samples



Figure 3: Grain Size Distribution Curve for soil samples.

All the soil samples analysed have a high percentage of material passing through the No. 7 sieve, ranging from 93.12% to 99.06%. The highest percentage passing was seen in the Sample E (99.06%), indicating that most of the soil sample is finer than 2.36 mm, suggesting a higher content of sand and finer particles. The lowest percentage passing was observed in the sample T-E (93.12%), which indicates a slightly coarser material but still dominated by fine particles. Since all samples have more than 93% of soil passing through this sieve, they are likely to have a good number of fine particles, which could contribute to higher compaction and stability in construction but may

also affect drainage.

The percentage passing through the No. 36 sieve varies more significantly, ranging from 46.56% to 79.38%. The highest percentage passing was observed in the Sample CS (79.38%), indicating a significant number of finer particles, closer to the size of silts and fine sands. The lowest percentage passing is in the sample T-E (46.56%), indicating a relatively coarser material compared to other samples. The differences in percentage passing through this sieve indicate variability in the finer fractions among the samples. A higher percentage suggests a more uniform and finer soil texture, which could be beneficial for certain construction applications but may reduce permeability.

The percentage passing through the No. 200 sieve is the smallest across all the soil samples, ranging from 0.71% to 2.86%. The highest percentage passing is in Sample T (2.86%), indicating a slightly higher clay or very fine silt content. The lowest percentage passing is in the Sample CS (0.71%), indicating a very low content of particles finer than 0.07 mm, which suggests minimal clay content. Thus, the low percentage passing through the No. 200 sieve is generally preferred for construction materials as it indicates less clay content, which can improve drainage and reduce issues related to soil plasticity and shrinkage.

The Contaminated Soil (CS) sample has a high percentage passing through both the No. 7 (98.12%) and No. 36 (79.38%) sieves, indicating a high content of fine particles. However, it has the lowest percentage passing through the No. 200 sieve (0.71%), indicating minimal clay content. This combination suggests that contamination may have altered the soil's texture, making it finer overall but with less clay content.

3.5 Direct shear box parameters of the soil samples

Direct shear box parameters such as cohesion, angle of internal friction and unit weight of the treated soil samples (E, T, E-T, and T-E) are presented in Figure 4a, 4b, and 4c, respectively.

Cohesion (C) is a measure of the intermolecular forces within the soil, contributing to its shear strength. As depicted in Figure 3a, the values range from 6.00 to 12.00 kN/m². Typically, higher cohesion values suggest that the soil has a good capacity to stick together, which can be beneficial for stability.



Figure 4a: Cohesion (C) for the treated soil samples (E, T, E-T and T-E) and contaminated soil.

Thus, as indicated in Figure 3, soil treated through electrokinetics (E) shows the highest cohesion value of 12.00 kN/m², indicating strong internal bonding. This is followed by the contaminated soil (CS) sample with a cohesion value of 9.00 KN/m² indicating moderate strength, slightly stronger than E-T and T-E samples. The soil treated through thermal treatment (T) shows the lowest cohesion value of 6.00 kN/m², suggesting weaker soil structure. The soil sample that undergoes electrokinetic followed by thermal followed by electrokinetic treatment (T-E) both exhibits moderate cohesion (8.00 kN/m²), indicating a balanced internal structure.



Figure 4b: Angle of Internal Friction (φ) for the treated soil samples (E, T, E-T and T-E) and contaminated soil.

Angle of internal friction indicates the soil's resistance to shearing under load. Higher values generally reflect better shear resistance and a stronger soil matrix. Thus, from the Figure 3b, Sample E has the highest angle of internal friction at 15.00° , indicating higher resistance to shear forces. Sample T has the lowest at 11.00° , suggesting low shear resistance.

Sample E-T has a moderate value at 13.00° , showing a balanced shear resistance. Whereas Sample T-E has a slightly lower at 12.00° , but still indicative of moderate shear resistance.



Figure 4c: Unit weight (γ) of the treated soil samples (E, T, E-T and T-E) and contaminated soil.

Figure 4c shows the unit weight of the treated soil samples (E, T, E-T and T-E) and contaminated soil. There is no much difference between all the samples analysed in terms of unit weight. Sample CS has the highest unit weight at 18.39 kN/m³, indicating the densest soil among the samples whereas Sample T-E is slightly lower at 18.34 kN/m³. Sample E, E-T and T shows minimal difference with unit weight of 18.20 kN/m³, 18.26 kN/m³ and 18.28 kN/m³ respectively.

3.6 Maximum Dry Density (MDD) and Optimum Moisture Content (OMC)

Maximum Dry Density (MDD) measured in Mg/m³ and Optimum Moisture Content (OMC) of the treated soil samples (E, T, E-T and T-E) and contaminated soil are depicted in Figure 5a and 5b. Maximum dry density reflects the density of the soil when it is compacted at its optimum moisture content. The values are consistent with little difference in all soil samples ranging from 1.79 to 1.80 Mg/m³, which is typical for many subgrade soils. High MDD values suggest that the soil can be compacted to a dense state, which is desirable in construction.



Figure 5a: Maximum Dry Density (MDD) content for the treated soil samples (E, T, E-T and T-E) and contaminated soil.



Figure 5b: Optimum Moisture Content for the treated soil samples (E, T, E-T and T-E) and contaminated soil.

OMC is the moisture content at which the soil can be compacted to achieve its maximum density. The OMC values of the soil samples analysed ranges from 9.48% to 16.46% in Figure 4b, with higher values indicating that more water is needed for optimal compaction. Higher OMC can be a concern in wet conditions as it might lead to difficulty in achieving desired compaction.

3.7 California Bearing Ratio (CBR)

The California Bearing Ratio (CBR) test measures the strength of the soil in relation to a standard crushed rock. The CBR test is a penetration test used to evaluate the subgrade strength of roads and pavements. Figure 6 provides the CBR values for the contaminated soil (CS) and the different remediated soil samples (E, T, E-T and T-E). The CBR value for the contaminated soil sample is 27.68%. This value is relatively low, indicating that the contaminated soil has a weaker load-bearing capacity. Contamination typically affects the soil structure and reduces its strength, leading to a lower CBR value. Sample E shows a slight improvement in the CBR value (29.07%) compared to the contaminated soil. The remediation process appears to have increased the soil's strength, though the improvement is modest. Sample T has an even higher CBR value (29.5%) than Sample E, suggesting that the treatment applied to this soil was more effective in enhancing its load-bearing capacity. The CBR value for Sample E-T is slightly lower than Samples E and T, indicating that this particular combination of remediation techniques did not enhance the soil strength as much as individual treatments. Finally, Sample T-E shows the highest CBR value (29.75%) among all the remediated soils, suggesting that the sequence of treatments applied here (thermal followed by electrokinetics) was the most effective in improving the soil's load-bearing capacity.



Figure 6: California Bearing Ratio (CBR) for the treated soil samples (E, T, E-T and T-E) and contaminated soil.

IV. CONCLUSIONS AND RECOMMENDATIONS

4.1 Conclusions

The following conclusions were drawn from the results obtained:

1. The contaminated soil (CS) had the highest level of total petroleum hydrocarbons (TPH). The results showed that the electrokinetics-thermal (ET) method was the most effective, achieving the highest removal efficiency (39.4%) with the lowest remaining TPH (18.9%). Thermal-electrokinetics (TE) was slightly less effective, while electrokinetics (E) and thermal (T) had moderate removal efficiencies, with thermal being the least effective among the remediation methods tested.

2. The analysis revealed that oxygen is the most abundant element across all samples, with concentrations ranging from 43.024% to 44.775%, suggesting the presence of oxides. Silicon is another major component, with its concentration indicating the presence of silicate minerals like quartz especially in less contaminated samples. Aluminum and calcium also showed variations, indicating the presence of aluminosilicates and carbonates, respectively.

3. The Maximum Dry Density (MDD) and Optimum Moisture Content (OMC) of the different soil samples subjected to various treatments with Sample E having MDD of 1.78 mg/cm³ and an OMC of 16.46%, indicating a lower density and higher moisture content. Sample T shows a slightly higher MDD of 1.80 mg/cm³ but a significantly lower OMC of 9.48%. Sample E-T and Sample T-E both have an MDD of 1.80 mg/cm³ and 1.79 mg/cm³, respectively, with OMC values of 13.46% and 13.03%, showing moderate moisture content. Whereas Sample CS has an MDD of 1.80 mg/cm³ and an OMC of 14.82%. Overall, the MDD values are relatively consistent, while the OMC varies more significantly across the samples.

4. It was found that the California Bearing Ratio (CBR) values for the different soil samples shows that; Sample E has a CBR of 29.07%, indicating good strength, Sample T has a slightly higher CBR of 29.50%, suggesting a marginally better load-bearing capacity, Sample E-T has a slightly lower CBR of 28.15%, indicating a slight decrease in strength, Sample T-E exhibits the highest CBR at 29.75%, showing the best load-bearing capacity among the samples whereas Sample CS has the lowest CBR at 27.68%.

4.2 Recommendations

Based on the findings summarized, the following recommendations can be made:

• The optimization of remediation methods should be considered since the electrokinetics-thermal (ET) method was the most effective in removing contaminants, it should be further optimized and applied in similar contaminated sites. This method could also be combined with other techniques to enhance overall efficiency.

- Additionally, further stabilization techniques should be considered to enhance the load-bearing capacity of remediated soils. This will prevent structural failures and ensure long-term stability and durability of the road construction.
- Further research in terms of combined remediation techniques should be conducted to explore other possible combinations of remediation techniques that could yield even better results for various soil types and contamination levels.

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