

RESEARCH ARTICLE



Effect of Charcoal and Active Carbon as Filter Media on Electrokinetic Remediated Crude Oil-Contaminated Soil

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Abstract: Research has shown that traditional electrokinetic remediation (EKR) technology has limited efficiency against different contaminants and soils. This has led to the use of several strategies to enhance the traditional EKR for higher contaminants removal efficiency. This paper separately uses charcoal and active carbon (AC) each 1 cm thick as filter media in EKR to enhance the removal of crude oil from crude oil-contaminated soil (COCS) containing 78,600 mg/kg of crude oil. The filters are placed very close to the anode compartment containing 0.01 M NaOH electrolytes. Graphite electrodes are used in a face-to-face configuration to pass 1 V DC/cm through the COCS. Crude oil removal efficiencies of 81.4% and 84.6%, respectively, are obtained from the EKR setups containing charcoal and AC filters after 18 and 14 days of remediation, respectively. This makes AC a better filter medium than charcoal for COCS. The results of the chemical oxides compositions show improvement from non-lateritic COCS to lateritic for the filter media-enhanced EKR soil, although AC has proven to be a better filter medium compared to charcoal for use in enhancing EKR technology for COCS. Nonetheless, with over 80% removal efficiency, charcoal or AC filters can be used to enhance EKR remediation of COCS.

Keywords: electrokinetic remediation, charcoal, active carbon, crude oil-contaminated soil

1. Introduction

The need for contaminants free soils for good health, environment, and structural uses cannot be overemphasized. About 10% of over two million tons of oil produced daily across the globe accounts for spillage [1]. During the transformation or weathering of released spilled oil from corroded pipelines and tankers, sabotage, oil production operations, and non-functional production equipment into the environment, various kinds of chemical, physical, geotechnical, and biological changes happen to the soil, water, and air [1, 2]. Various kidney, lungs, liver, reproductive, nervous and immune systems disorders, and even cancer are some harmful human and animal health issues that can be attributed to soil contaminated with petroleum [2].

Oil content of soils can be determined from its total petroleum hydrocarbons (TPHs). TPH limits are 1000–2000 mg/kg for organic soils [2]. It can also be used to show removal efficiency as how Sun et al. [3] after weathering at 50°C for 14 days using gravimetric method reported 99.6% and 65.3% of TPH removal for kerosene and diesel-contaminated soils, respectively. Alinnor and Nwachukwu [4] determined the TPH content in samples of groundwater and soil collected at depths up to 5.0 m at five communities in Rivers State, Nigeria by Gas Chromatograph – Flame Ionization Detector (GC-FID) method and obtained concentrations as high as 1651.00 mg/kg and 33076.00 µg/l in soil

and water, respectively. Similarly, Egedeuzu and Nnorom [5] obtained TPH mean concentrations of 24000 mg/kg by GC-FID method, far above the limit of 2000 mg/kg for soils. This shows that various methods are available to measure the oil content of samples to relative degrees of accuracy. Depending on the cost, technical know-how, and aim of testing, TPH of soils or groundwater measured by any method can represent their oil contents and these oil contents can exceed permissible limits.

Many technologies have been adopted in remediating soils contaminated with oil. Choi et al. [6] used thermal heating with temperature of about 400°C to desorb almost 100% of TPH, unresolved complex mixtures, polycyclic aromatic hydrocarbon (PAHs), and alkylated PAHs from soil collected at a landfill site within 15 min. Cho et al. [7] by microwave heating also achieved 91.1% TPH removal efficiency from coarse-grained soils, although 71.2% removal efficiency was achieved after a relatively longer time for fine-grained soil. These illustrate the inconsistent removal efficiencies for contaminants in soils of different gradations and at different depths. Moreover, pollutants, such as petroleum hydrocarbons, are difficult to remove due to their higher adsorption rate compared to soil particles [8].

Electrokinetic remediation (EKR) is an advanced in-situ and ex-situ technique used for the removal of toxic metals, organic pollutants, and radio-nuclide materials from contaminated soil [9]. This can be achieved in-situ by installing electrodes in drilled wells or trenches, which may incorporate pumping and conditioning systems and then applying a very low direct current (DC) electric

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potential. It can also be achieved ex-situ with the use of specially designed above-ground reactors [10]. The contaminants accumulate at the electrodes and are removed by either adsorption or withdrawal followed by treatment [11]. The large-scale viability and economics of EKR for site decontamination are remarkable. This is evident when Surya Ramadan et al. [12] removed 46.4% hydrocarbon from low permeable soil after integrating EKR into bioremediation for 7 days. Muhsina and Chandrakaran [13] with 0.6 V/cm specific voltage also removed 80% and 45% of oil, respectively, from the cathode and anode sides of a clayey soil using an unenhanced EKR setup after 18 days. Henceforth, 65–75% restoration of the soil properties was also achieved.

However, research has established that EK can only make the pollutants in the soil move to the designated region but cannot easily eliminate the pollutants. As such, there is a need to combine EKR with other technologies to achieve the effect of removing the moving pollutant in the soil [14]. Otherwise, electro-osmotic flow would reverse, which may prolong the entire remediation process [11]. The technique of coupling EKR and filter media (permeable reactive barrier (PRB)) is one of the suitable techniques that can enhance the removal efficiency of pollutants by EKR. This works by increasing the mobility and removal of pollutants, thereby reducing material and manpower inputs [15, 16]. Efficient application of this technology relies on the proper choice of the material properties for PRB employed, as they are contaminants specific [17]. Use of zero-valent iron, atomizing slag, humid acid, zeolite, and carbon active materials as filters in between the electrodes and contaminated soils in some EKR setups has shown remarkable performances in the removal of organic and inorganic contaminants [15]. The high surface area and porous structure of the reactive filters enable the contaminants to be removed by physical, biological, or chemical process as they pass through the filters [10].

The use of carbonaceous materials like active carbon (AC) obtained by either chemical treatment of charcoal or heating wood to a very high temperature between 600 and 900°C has been experimented. Even though it is pH dependent, the well-developed internal pores of AC together with the large amount of phenol, carboxyl, and hydroxyl groups distributed on its surface give it a good absorption ability and chemical stability [18]. The high cost of producing AC for use as filter medium in EKR technology to remove crude oil from crude oil-contaminated soil (COCS) is a limitation to this technique. This has made enquiry into the use of cheaper and readily available and effective materials like charcoal made by incinerating wood at moderate temperature and without the addition of chemicals like in AC but whose surface area can be increased by grinding it to finer particles as filter medium in EKR to achieve results similar to AC. Research has indicated a gap that a comparison of their performance in removing crude oil is yet to be fully reported [11, 12, 15]. This paper compares the effect of charcoal and AC as filter media on electrokinetic remediated COCS.

2. Materials and Methods

This research involves the usage of many materials in many laboratory experimental methods in comparing the effect of charcoal and AC as filter media on EKR of COCS.

2.1. Materials

The materials used for this research are as follows.

COCS collected at 1 m depth from the Nigerian Pipeline and Storage Company, Kaduna located around latitude 10°24'6" and longitude 7°29'32"; sodium hydroxide (NaOH); clean potable water; deionized water; and connecting wires, clips, and 8 mm diameter graphite electrode rods of length 300 mm all sourced within Kaduna State were used in this work.

EKR cell of overall dimensions, 400 mm by 200 mm by 300 mm made from clear plexiglass plate; charcoal (Figure 1), of sieve sizes between 300 and 150 µm, obtained by incinerating hardwood to a temperature of 300°C; and AC (Figure 1), heated to 750°C, of sieve sizes between 300 and 150 µm, obtained within Kaduna State were used in this work.

Figure 1
Charcoal and active carbon filter media materials



2.2. Methods

The methods adopted for this research work are discussed as follows.

2.2.1. TPH test

The gravimetric method (the toluene cold extraction method) as described in Sun et al. [3] and Adebayo et al. [15] was used to determine the TPH contents of the collected COCS and the filter media-enhanced electrokinetic remediated soils. The removal efficiencies are determined from Equation (1).

2.2.2. Electrokinetic remediation

The EKR setup used in this research is based on the model adopted by Yu et al. [10], Haruna et al. [11], and Adebayo et al. [15]. This consists of COCS, graphite electrodes, connecting cables and clips, plexiglass electrokinetic cell, NaOH, filter media, DC supply, solar power supply means, and reservoir tanks. The setup (described in Table 1) was connected as shown in Figure 2 with 0.01 mol NaOH electrolyte used at both the anode and cathode compartments of the setups. AC and charcoal (Figure 1) were used separately as filter media placed between two filter papers attached to the inside perforated wall of the middle partition adjacent to the anode in the EKR cell before loading the soil (Figure 2). One V DC/cm (30 V) was allowed to flow through soil in the setups from the anode to the cathode electrodes/compartments until no current drop was observed on the DC supply for three consecutive days.

At the end of the process, the remediated soil was extracted from the cell, sliced into five equal parts, and allowed to dry under normal room condition. The TPH and chemical oxide at each slice were determined to ascertain the level and variation in the remediation of the contamination carried out with the two-filter media.

Table 1
Summary of electrokinetic remediation setup

Details	Setup 1	Setup 2
Contaminant	Crude oil	Crude oil
Length of soil	30 cm	30 cm
Depth of soil	30 cm	30 cm
Width of soil	20 cm	20 cm
Electrical potential	30 V	30 V
Anolyte conc.	NaOH (0.01 M)	NaOH (0.01 M)
Catholyte conc.	NaOH (0.01 M)	NaOH (0.01 M)
Filter material	Charcoal	Active carbon
Thickness of filter	1 cm	1 cm
Depth of filter	30 cm	30 cm
Width of filter	20 cm	20 cm

Table 2
Details of filter media-enhanced EKR COCS

Details	Charcoal	Active carbon
Initial TPH	78,600 mg/kg	78,600 mg/kg
Duration	18 days	14 days
Average remediation efficiency	81.4%	84.6%

3.2. Remediation process

As seen in Table 2, similar to 18 days observed in Muhsina and Chandrakaran [13] with unenhanced EKR of clayey soil whose oil content was not up to 78,600 mg/kg with 0.6 V/cm setup, the charcoal filter medium-enhanced EKR of the COCS process lasted 18 days using 1 V/cm specific voltage. This relatively longer duration considering the enhancement could be due to the higher concentration of the crude oil present in the contaminated soil in this research. The current variation was from 0.13 to 0.06 A with a total effluent of 5,820 ml collected at the cathode outlet. The pH was observed after 24 h as 10.1 at the slice closest to the anode and 10.8 at the slice closest to the cathode. These values were observed to increase to 10.3 and 11.1, respectively, before decreasing to 9.1 and 10.0, respectively, at the end of the remediation. This basic pH at the cathode agrees with Maturi and Reddy [19]. It also suggests that the system fairly established a buffering state during the remediation process as the pH change with remediation was minimal. The electrical conductivity (EC) after 24 h was 201 $\mu\text{S}/\text{cm}$ at the slice closest to the anode and 231 $\mu\text{S}/\text{cm}$ at the slice closest to the cathode. These were observed to increase to 290 and 296 $\mu\text{S}/\text{cm}$ before decreasing to 206 and 104 $\mu\text{S}/\text{cm}$, respectively, at the end of the remediation. This shows that desorbed metals are moved from the anode to the cathode before removal in the effluents collected at the cathode end during the remediation process involved the mobility of desorbed metals from the anode to the cathode before removal in the effluents collected at the cathode end. This also agrees with Virkutyte et al. [20] as a measure of the progress of the remediation process as the conductivity was observed to decrease with days of remediation.

When the AC was used as the filter medium in the EK remediation of the COCS, the remediation process as seen in Table 2, though lasted 14 days using the same 1 V/cm specific voltage, was also similar to Muhsina and Chandrakaran [13] with unenhanced EKR of clayey soil. This makes it about 29% faster than the charcoal filter medium EKR process. The current variation was from 0.12 to 0.05 A with a total effluent of 2,570 ml collected at the cathode outlet. The pH was observed after 24 h as 8.8 at the slice closest to the anode and 8.7 at the slice closest to the cathode. These values were observed to increase to 10.0 and 10.7, respectively, before decreasing to 8.9 and 10.6, respectively, at the end of the remediation. This initial lower pH compared to those with the charcoal filter medium enhancement could be due to the influence of the activation of the carbon by acid, which favored the faster mobility of the contaminants as emphasized by Maturi and Reddy [19]. The EC after 24 h was 292 $\mu\text{S}/\text{cm}$ at the slice closest to the anode and 234 $\mu\text{S}/\text{cm}$ at the slice closest to the cathode. These were observed to follow Virkutyte et al. [20] with decrease continuously to 95 and 127 $\mu\text{S}/\text{cm}$, respectively, at the end of the remediation. This suggests that the removal rate of the mobilized metal ions is faster for AC than charcoal filter medium-enhanced EKR process. It can also be concluded that with AC filter medium, a better remediation process for COCS is achievable than with charcoal filter medium.

Figure 2

Filter media-enhanced EKR of COCS



The removal efficiency (Re) of the charcoal and AC was calculated from Cho et al. [7]:

$$\text{Re}(\%) = \left(\frac{C_0 - C}{C_0} \right) \times 100 \quad (1)$$

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).

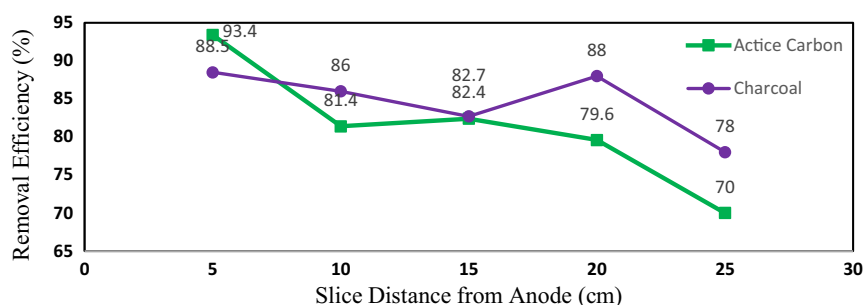
3. Results and Discussion

The results of all the laboratory experimental investigations carried out on the COCS and EKR-remediated soils incorporating charcoal and AC filters are summarized in tables and figures.

3.1. Contaminant level

The TPH of the COCS and those remediated by EKR technology enhanced with charcoal and AC filters determined by gravimetric method are presented in Table 2. The TPH value of 78,600 mg/kg for the COCS is above the safe limits for soils and should be remediated for human and environments concerns [2]. The technology incorporating charcoal and AC filters in enhancing the traditional EKR technology showed noticeable improvements in contaminants removal.

Figure 3
Removal efficiency of filter media-enhanced EKR soils



3.3. Removal efficiency

The removal efficiency of the charcoal and AC filters enhanced EKR of COCS is plotted against the slice distance from anode as shown in Figure 3. For the charcoal filter EKR process, the highest removal efficiency was achieved as 93.4% at the slice closest to the charcoal (anode) and the lowest removal efficiency was achieved as 70.0% at the slice farthest from the charcoal (cathode). This suggests that the location of the charcoal plays a vital role in the remediation process. Agreeing with Jamshidi-Zanjani and Khodadadi Darban [16], the charcoal filter, due to its amorphous structure, can be seen to enhance the removal of the mobilized crude oil from the soil by adsorbing to its pores. Charcoal filter medium-enhanced EKR of COCS has an average removal efficiency of 81.4%. This result exceeds the 71.2% and 65.3% reported by Sun et al. [3] and Cho et al. [7] with diesel-contaminated soil, respectively.

Similarly, for the AC filter medium-enhanced EKR process, the highest removal efficiency was achieved as 88.5%, a value lower than that of the charcoal filter but at the same slice (closest to the anode). This lower removal efficiency at the slice closest to the anode with AC could be due to the fewer visible pores on the surface of the AC when compared to those on the charcoal. The lowest removal efficiency was achieved as 78%, a value higher than that of the charcoal filter but at the same slice (closest to the cathode). This agrees with Yu et al. [10] where the chemical activation of the AC is emphasized to play a role in increasing the surface activity of the filter, thereby increasing the rate of contaminants removal by adsorption. This also implies that the location of the AC plays a vital role in the remediation process and the crystalline nature of the AC may be responsible for its lower crude oil adsorption ability. AC filter medium-enhanced EKR of COCS has an average removal efficiency of 84.6%. This result represents a 3.9% increment to that of the charcoal filter medium-enhanced EKR and implies that AC has higher average crude oil removal efficiency than charcoal as filter medium in an EKR setup. This could be due to the higher ability of the AC to remove the crude oil and dissipate it into the anolyte than the charcoal.

The removal efficiency of the charcoal and AC filters enhanced EKR of COCS can also be ascertained from the color variations in the resulting soil sample. Relative to the reddish-brown laterite color in Okeke et al. [21], the mud brown color of the COCS is seen in Figure 4 to change to plaster brown when charcoal was used as the filter medium in the enhanced EKR process. A khaki brown color is observed when the COCS was remediated with AC as filter medium in the EKR setup. Although the color improvement with AC filter is better than that with charcoal filter, the reddish-brown of laterite observed by Okeke et al. [21] is seen to be absent in the COCS and the filter media-enhanced EK-remediated

Figure 4
Samples of COCS, charcoal, and active carbon EKR soils



soils. This could be because of the relative presence of dark colored crude oil in all the samples.

From the results of the chemical oxides composition of the COCS and EKR soils enhanced with filter media shown in Table 3, it can be generally observed that there are relative enrichments in the lateritic constituents of the filter media-enhanced EKR soil with decrease in their bases and silicate contents. COCS with lateritic constituents of 45.17% classifies as a lateritic lithomarge. Charcoal and AC filter media-enhanced EKR soils have lateritic constituents of 44.13% and 46.56%, respectively, indicating that they are also lateritic lithomarge.

According to Okeke et al. [21], the $\text{SiO}_2/\text{Al}_2\text{O}_3$ and $\text{SiO}_2/\text{Fe}_2\text{O}_3$ for the COCS are 2.00 and 2.02 indicating that the COCS can be categorized as non-lateritic soil, for the filters enhanced EKR

Table 3
Chemical composition of the COCS and filter media-enhanced EKR soils

Components (oxides)	Weight (%)		
	COCS	Charcoal EKR	Active carbon EKR
Al_2O_3	21.785	22.405	22.822
SiO_2	43.537	42.015	42.158
SO_3	1.062	4.712	1.187
K_2O	1.180	0.412	1.062
PbO	0.020	0.052	0.002
CaO	7.830	2.917	6.219
TiO_2	1.677	1.721	1.735
MnO	0.104	0.113	0.221
Fe_2O_3	21.604	21.392	21.875
NiO	0.002	0.014	0.001
LOI	1.199	4.247	2.718

soils, the $\text{SiO}_2/\text{Al}_2\text{O}_3$ and $\text{SiO}_2/\text{Fe}_2\text{O}_3$ are 1.88 and 1.92, and 1.85 and 1.93 for charcoal and AC filters enhanced EKR soils respectively. These indicate that the filter media-enhanced EKR soils are lateritic soils. This improvement could be due to the slight increase in their lateritic constituents with a proportionate relative decrease in the metals content with remediation. This shows that the use of charcoal and AC as filter media in enhancing the removal of crude oil from COCS by EKR technology can affect the degree of laterization of the COCS from a non-lateritic soil to a lateritic soil. However, AC when compared to charcoal filter medium has better laterization effects on the EK-remediated COCS.

4. Conclusions

Effect of charcoal and AC filters on COCSs remediated by electrokinetic technology has been determined. Conclusions from the results presented and discussed are as follows.

The TPH content of a soil can be as high as 78,600 mg/kg. The use of filter media as enhancement in EKR of COCS is a viable technology. Charcoal and AC are good filter media for enhancing the removal of crude oil from COCSs when remediated by electrokinetic technology. It can be concluded that AC, with an average removal efficiency of 84.6%, is a better material than charcoal with an average removal efficiency of 81.4% when used as filter medium in enhancing EKR technology for non-lateritic COCSs.

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Ethical Statement

This study does not contain any studies with human or animal subjects performed by any of the authors.

Conflicts of Interest

The authors declare that they have no conflicts of interest to this work.

Data Availability Statement

The data that support the findings of this study are openly available at <https://doi.org/10.4314/njt.v42i2.3>, reference number [15].

Author Contribution Statement

Kabiru Adebayo: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization, Project administration, Funding acquisition. **John Sani Engbonye:** Conceptualization, Methodology, Investigation, Writing – review & editing, Supervision. **Abdulrahman Musa:** Resources. **Moses George:** Conceptualization, Methodology, Investigation, Writing – review & editing, Visualization,

Supervision. **Ige Olumide Oluwasanmi:** Investigation, Supervision.

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