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Effect of Ethanol and Acetone Cosolvents in Enhancing Electrokinetic Remediation of Crude Oil Contaminated Soil Obtained from a Pipeline and Storage Company, Kaduna Nigeria

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ABSTRACT: A lot of work has shown that despite the effectiveness of the traditional electrokinetic remediation (EKR) technology in decontaminating soils with different types and amounts of contaminants, it can be enhanced by a number of strategies for extra effective performance. This work presents the effect of ethanol and acetone cosolvents in enhancing EKR of crude oil contaminated soil (COCS), collected at a depth of 1 m from a petroleum pipeline and storage company, Kaduna Nigeria using graphite electrodes to pass 1 V DC/cm across EKR setups enhanced by incorporating 20% ethanol and 20% acetone separately as cosolvents in the anode compartments of the setups. The total petroleum hydrocarbon results showed that the crude oil content of 78,600 mg/kg present in the COCS exceeds permissible limits for soils. Average removal efficiencies of 74.61% and 67.79% obtained from 20% ethanol and 20% acetone cosolvents enhancements respectively showed that 20% ethanol, with higher removal efficiency, is a better cosolvents for COCS than 20% acetone. Although 20% ethanol has been shown to be a better cosolvents compared to 20% acetone, either of them can be incorporated into EKR technology for enhancing the remediation of COCS.

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Soil and sediments are natural sinks of organic matter, carbonates, iron oxides, sulphides as well as toxic metals. Crude oil along with aliphatic and aromatic compound contains some hazardous materials such as benzene, toluene, ethyl benzene, and xylene (BTEX) that can pollute the soil and groundwater environment, accumulate in human bodies and living things, and destroy the ecosystems (Li *et al.*, 2016). Cancer and various disorders on lungs, kidney, liver, reproductive and immune systems as well as the nervous system are harmful animal and health issues attributed to soil contaminated with petroleum (Egedeuzu and Nnorom, 2013; Meshari, 2021). The impurity of the environment created by petroleum hydrocarbons compounds (PHC) products like fuels, oils, lubricants, waxes, and others can be quantitatively expressed as total petroleum hydrocarbon (TPH) (Egedeuzu & Nnorom, 2013; Meshari, 2021). Limits for organics may be at 1000 – 2000 mg/kg, but diesel hydrocarbon contamination could be 1000 mg/kg (Egedeuzu & Nnorom, 2013; Meshari, 2021). Remedial actions are recommended when the risk level is unacceptable. Measures adopted to control, reduce, mitigate or eliminate the risk resulting from contamination of the soil and/or groundwater media known as remediation technologies are required (DOE, 2009). Selected approaches are needed to be efficient and cost-effective in achieving the remedial goals (Sharma & Reddy, 2004). Worldwide remediation technologies like soil washing (Dermont *et al.*, 2008), bioremediation (Virkutyte *et al.*, 2002), thermal desorption (Choi *et al.*, 2020) soil vapour extraction

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(Virkutyte *et al.,* 2002) and soil flushing (Virkutyte *et al*., 2002) have been experimented with limited results. USEPA (2000 and 2006) posited that these remediation technologies have shown inadequate performance to remediate complex site conditions with: 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 metals contaminants; and mixed contaminants (e.g., organic compounds combined with heavy metals and/or radionuclides). The difficulty in accessing the contaminants and high electrical conductivity and exchangeable sodium percentage are significant challenges encountered (Mohammed *et al.,* 2013). This affirms an urgent need to develop new technologies that can overcome these challenges as well as be cost-effective (Reddy, 2010). For oil contaminated soils, many remediation technologies have been experimented with relative results. Choi *et al*. (2020) recommended elevated temperature of about 400^0C for achieving about 100% remediation of oil contaminated soils from landfills and gas sites by thermal desorption in about 15 minutes. Cho *et al.* (2020) also obtained 91.1% removal of TPH from coarse grained soils by microwave heating, although 71.2% removal efficiency was recorded for fine grained soil after a relatively longer time. These show that even if there are different remediation methods for contaminated sites, they fail to deliver consistent results when the contaminated area is located at a considerable depth and the soil type is clay due to its low permeability. Moreover, pollutants, such as petroleum hydrocarbons, have a high adsorption rate compared to soil particles, making their removal or destruction more difficult (Streche *et al*., 2018). The use of electrokinetic remediation (EKR) in large scale has been demonstrated to be a viable and economic approach to decontamination. This is emphasised when Bimastyaji *et al*. (2018) integrated EKR into bioremediation of low permeable hydrocarbon contaminated soil and was able to achieve 46.4% removal efficiency in 7 days’ operation. Muhsina and Chandrakaran (2015) also used an unenhanced electrokinetic (EK) setup to remove 80% and 45% of contaminants at cathode and anode respectively from an oil contaminated clayey soil after 18 days by using 0.6 V/cm specific voltage. The overall efficiency of the treatment and restoration of properties was in the range of 65-75%. Cosolvents which involves careful mixing of two or more solvents, enhances aqueous solubility and transport of organic pollutants like phenols, polychlorinated biphenyl, benzenes, alkanes, polycyclic aromatic hydrocarbons (PAH) and halogenated hydrocarbons. Common cosolvents include methanol, ethanol, propanol, acetone, tetrahydrofuran, and butylamine (Yeung et al., 2011). Reddy et al. (2006) investigated the use of four-flushing agents (3% Tween 80, 5% Igepal CA-720, 20% n-Butylamine and 10% hydroxypropyl-□-cyclodextrin or HPCD) in the removal of PAHs and heavy metals. It was discovered that the use of cosolvents (20% n-Butylamine) led to increased soil pH, current density and electroosmotic flow were also best with the Cosolvents. This shows that cosolvents may enhance removal of organic contaminants more than surfactants. The concentration of the cosolvents is also a deciding factor as shown by Mutari and Reddy (2008) where the removal efficiency of PAHs was higher with 20% n-Butylamine than that with 10% n-Butylamine.

The fore-going has shown that crude oil content in soils can exceed permissible limits thereby posing dangers to human and environments, these contaminated soils can be remediated by electrokinetic technology. However, little to no research has been done to show the effect of cosolvents on electrokinetic remediation of oil contaminated soil. This research therefore evaluates the effect of ethanol and acetone cosolvents in enhancing electrokinetic remediation of crude oil contaminated soil obtained from a pipeline and storage company, Kaduna Nigeria.

MATERIALS AND METHODS

Many materials and equipment were used in different laboratory experiments to determine the effect of cosolvents on electrokinetic remediated crude oil contaminated soil (COCS).

Materials: The materials for this research work include:

i. Contaminated Soil: The crude oil contaminated soil (COCS) used for this work was obtained at a depth of 1 m from the Nigerian Pipeline and Storage Company, Kaduna located around latitude $10^0$24’6” and longitude $7^0$29’32”.

ii. Distilled Water: The distilled water obtained from the Department of Fashion Design and Clothing Technology; Kaduna Polytechnic, Kaduna State, Nigeria was used.

iii. Water: Tap water from the borehole provided near the Civil Engineering laboratory of the Nigerian Defence Academy, Kaduna, Kaduna State, Nigeria was used.

iv. Electrodes: 8 mm graphite electrode, 300 mm long obtained from a local Laboratory equipment store at Lagos Street in Kaduna State, Nigeria was used.

v. EKR Cell: EKR cell made from clear Plexiglas plate of overall dimension, 400 mm by 200 mm by 300 mm, with middle internal partition, 300 mm by 200 mm by 300 mm and two outer partitions, 50 mm by 200 mm by 300 mm adjoining the middle was used.

vi. Connecting Wires and Clips: Flexible connecting wires and battery clips obtained from a local electrical store at Lagos Street in Kaduna State, Nigeria were used.

vii. Sodium Hydroxide (NaOH): NaOH obtained from a laboratory chemical store at Constitution Road in Kaduna State, Nigeria was used.

viii. Acetone: Acetone obtained from a laboratory chemical store at Constitution Road in Kaduna State, Nigeria was used.

ix. Ethanol: Ethanol obtained from a laboratory chemical store at Constitution Road in Kaduna State, Nigeria was used.

x. Toluene: Toluene obtained from a laboratory chemical store at Constitution Road in Kaduna State, Nigeria was used.

xi. DC Supply: 30 V, 5 A DC supply was used.

Determination of Total Petroleum Hydrocarbon (TPH): The TPH contents of the contaminated soil and those of the cosolvents enhanced electrokinetic remediated soils in this research were conducted by gravimetric method (the Toluene cold extraction method) as described in Yue *et al*. (2021). The removal efficiencies of the filter media were evaluated from Cho *et al.* (2020).

Electrokinetic Remediation: The electrokinetic remediation setup used in this research is based on the model adopted by Bimastyaji *et al.* (2018). As shown in Fig. 1, it consists of direct current (DC) supply, electrokinetic unit, electrodes, connecting cables and clips, electrolytes (purging solutions), solar power supply means and reservoir tanks. The DC supply is used to supply constant 30 V and maximum of 5 A through graphite electrodes (two at both ends) to the contaminated soil in the setup to achieve 1 VDC/cm across the setup. The electrokinetic unit, as described in Table 1 is made from a Plexiglas material was constructed into a cuboid, 40 cm long, 20 cm wide and 30 cm high. This consists of three sections; 30 cm long, 20 cm wide and 30 cm high middle partition which collects the contaminated soil to be remediated and two adjacent partitions, 5 cm long, 20 cm wide and 30 cm high attached to the middle partition by a well perforated Plexiglas divider. This is to ensure electroosmotic flow between the anode and cathode purging solutions through the soil. Two graphite electrodes each 8 mm diameter, and 30 cm long were placed vertically with a face-to-face configuration at the anolyte and catholyte compartment for passing direct current from the DC supply using connecting cables attached to clips through the setup. Solar power setup as used to ensure continuous power supply to the setup. Twenty percent (20%) acetone (20% acetone with 80% distilled water content) and 20% ethanol (20% ethanol with 80% distilled water content) both buffered with 0.01 M NaOH were used separately as cosolvents for anode purging solution and distilled water buffered with 0.01 M NaOH was used as purging solution for the cathode where graphite electrodes were placed.

Fig. 1: Cosolvents Enhanced Electrokinetic Remediation Set-up

Table 1: Summary of Electrokinetic Remediation Setup

Details	**Experiment 2**	**Experiment 3**
Contamination	Crude Oil	Crude Oil
Length of Soil (cm)	30.0	30.0
Width of Soil (cm)	20.0	20.0
Depth of Soil (cm)	30.0	30.0
Electrical potential (V)	30.0	30.0
Anode Purging Solution	20% acetone + 0.01 M NaOH	20% ethanol + 0.01 M NaOH
Cathode Purging Solution	Distilled Water + 0.01 M NaOH	Distilled Water + 0.01 M NaOH

The middle partition of the setup was filled with the contaminated soil, and purging solutions were introduced into the two outer partitions as shown in Table 1. The setup was connected as shown in Fig. 1 above, with provisions for continuous supply and recycling of purging solution using reservoirs at anode and cathode ends. Power supply was switched on and the remediation was carried out while pH and current variations were determined across the soil length through-out the duration of the process. Effluents from the cathode valve was collected and measured. The process was left on until no more effluent was produced at the cathode. At the end of the process, the remediated soil was sliced into five equal parts and the TPH at each slice was determined to ascertain the level and variation in the remediation of the contamination carried out.

RESULTS AND DISCUSSION

The results of the electrokinetic remediation of COCS enhanced by the use of 20% acetone and 20% ethanol cosolvents are shown in Table 2 and Fig. 2.

Table 2: Summary of Cosolvents Enhanced EKR of COCS

Details	20% Acetone	20% Ethanol
Initial TPH (mg/kg)	78,600	78,600
Duration (days)	12	6
Average Remediation Efficiency (%)	67.79	74.61

From Table 2 above, the total petroleum content (TPH) of the COCS collected at a depth of 1 m from the Nigerian Pipeline and Storage Company, Kaduna was observed to be 78,600 mg/kg. This content far exceeds the 1000 – 2000 mg/kg allowable limits reported by Egedeuzu and Nnorom (2013): and Meshari (2021). This necessitated the remediation of the COCS to improve its properties. The use of 20% acetone and 20% ethanol as cosolvents in the enhancement of EKR for the COCS gave remarkable results.

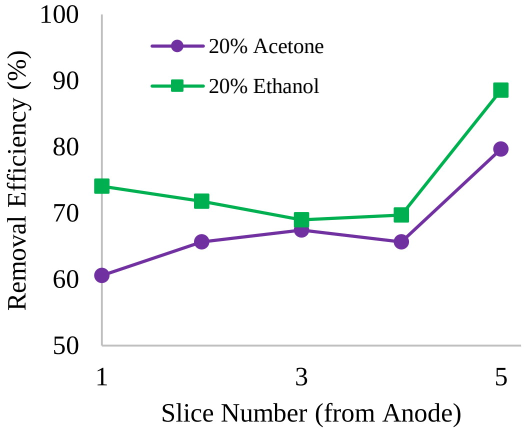

Fig. 2: Removal Efficiency of Cosolvents Enhanced EKR Soils

Similar to Cho *et al.* (2020) where 71.2% TPH removal efficiency was recorded for fine grained soil after a long period by microwave heating, it can be seen from Fig. 2 and that with 20% acetone cosolvents, a maximum remediation efficiency of 79.64% was achieved at the slice closest to the cathode compartment of the EKR setup. The lowest remediation efficiency was 60.56% which was obtained at the slice closest to the anode compartment. For the 20% ethanol cosolvent, as enhancement in the EKR setup, the maximum remediation efficiency was 88.55% also at the slice closest to the cathode compartment of the setup. The lowest efficiency was 68.96% obtained at the middle slice of the setup. Cosolvents enhanced EKR has also resulted in faster remediation when compared to Muhsina and Chandrakaran (2015) where oil contaminant was removed from a clayey soil in eighteen (18) days. Comparing the performance of both cosolvent; from Table 2, 20% acetone cosolvents took twelve (12) days to achieve an average remediation efficiency of 67.79% while 20% ethanol cosolvents took six (6) days to achieve an average remediation efficiency of 74.61%. This indicates 100% improvement in the use of 20% ethanol over 20% acetone when the duration of the remediation is considered. This is considered an advantage as it reduces the quantity of cosolvents, power supply and manpower required to achieve the remediation. An average of 74.61% remediation efficiency provided by the using 20% ethanol when compared to 67.79% obtained from the use of 20% acetone is also another reason to stress the advantage of using 20% ethanol over 20% acetone as cosolvents in EKR of COCS. The use of 20% ethanol as enhancement in remediating COCS does not only provide faster result but better efficiency, the 74.61% average efficiency provided by using 20% ethanol gives a 10.06% advantage over the 67.79% average efficiency obtained from the use of 20% acetone.

CONCLUSION

Effect of ethanol and acetone cosolvents in enhancing electrokinetic remediation of crude oil contaminated soil has been investigated and data obtained revealed that the TPH content of a soil can exceed permissible limits and become as high as 78,600 mg/kg. and ethanol cosolvents is a better cosolvents compared to acetone cosolvents for enhancing the electrokinetic remediation of crude oil contaminated soil, while the removal efficiencies of the cosolvents increase with distances from the compartment containing the cosolvents.

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REFERENCES

Bimastyaji, S. R.; Agus, J. E.; Qomarudin, H. (2018). Integrating electrokinetic and bioremediation process for treating oil contaminated low permeability soil. *E3S Web of Conferences*, *31*, 03005

Cho, K.; Myung, E.; Kim, H.; Purev, O.; Park, C.; Choi, N. (2020). Removal of total petroleum hydrocarbons from contaminated soil through microwave irradiation. *Int. J. Environ. Res. Public Health, 17*, 5952.

Choi, B.; Lee, S.; Jho, E. H. (2020). Removal of TPH, UCM, PAHs, and Alk-PAHs in oil-contaminated soil by thermal desorption. *App. Biol. Chem., 63*, 83-89.

Dermont, G.; Bergeron, M.; Mercier, G.; RicherLaflèche, M. (2008). Soil washing for metal removal: a review of physical/chemical technologies and field applications. *J. Hazard. Mater.*, *152*(1), 1–31.

DOE 2009 Contaminated Land Management and Control Guideline No. 3: Remediation of Contaminated Sites. *Department of Environment Guideline Standard*. Retrieved from doe.gov.my (3-5).

Egedeuzu, C. S.; Nnorom, I. C. (2013). Total petroleum hydrocarbon and metal contents of soil, plant and borehole water samples from crude oil spill sites in Owaza, Abia State. *ABSU J. Environ. Sci. Technol.*, *3*, 405-416.

Li, T. T.; Wang, Y.; Guo, S. H.; Li, X. Y.; Xu, Y. N.; Wang, Y. (2016). Effect of polarity-reversal on electrokinetic enhanced bioremediation of pyrene contaminated soil. *Electrochim. Acta*, *187*, 567–575.

Meshari, S. A. (2021). Determination of total petroleum hydrocarbons (TPHs) in weathered oil contaminated soil. *Environ. Eng. Res.*, 27(5), DOI: https://doi.org/10.4491/eer.2021.324

Mohammed, H. E.; Nuhu, D. M.; Salihu, L.; Alaadin, B. (2013). Integrated electrokinetics-adsorption remediation of saline-sodic soils: effects of voltage gradient and contaminant concentration on soil electrical conductivity. *Sci. World J.*, http://dx.doi.org/10.1155/2013/618495

Muhsina, M.; Chandrakaran, S. (2015). electrochemical remediation of oil contaminated fine-grained soil. *Asian J. eng. Technol.* (ISSN: 2321 – 2462) 3(4), Special issue for ICETTAS’15 301-309.

Reddy, K. R. (2010). Technical challenges to in-situ remediation of polluted sites. *Geotech. Geol. Eng. 28*(3), 211–221.

Reddy, K. R.; Ala, P. R.; Sharma, S.; Kumar, S. N. (2006). Enhanced electrokinetic remediation of contaminated manufactured gas plant soil. *Eng. Geol.*, *85*(1-2), 132-146.

Sharma, H. D.; Reddy, K. R. (2004). *Geoenvironmental Engineering: Site Remediation, Waste Containment, and Emerging Waste Management Technologies*, John Wiley & Sons, Inc., Hoboken, NJ.

Streche, C.; Cocârţă, D. M.; Istrate, I.; Badea, A. A. (2018). Decontamination of petroleum contaminated soils using the electrochemical technique: Remediation degree and energy consumption. *Sci. Rep.*, *8*(3), 272-285

USEPA. (2000). Innovative remediation technologies: fieldscale demonstration projects in North America (2nd ed.). EPA 542-B-00-004, Washington, D.C.

USEPA. (2006). Abstracts of remediation case studies, Vol. 10, EPA 542-R-06-002, Washington, D.C.

Virkutyte, J.; Sillanpaa, M.; Latostenmaa, P. (2002). Electrokinetic soil remediation-critical overview. *Sci. Total Environ.*, *289*(1), 97-121.

Yue, S.; Jie, M.; Ganngsen, Y.; Shuyu, L.; Hanyu, L.; Quanwei, S.; Baichun, W. (2021). Comparisons of four methods for measuring total petroleum hydrocarbons and short-term weathering effects in soils contaminated by crude oil and fuel oils. *Water Air Soil Pollut.*, *232*(381), https://doi.org/10.1007/s11270-021-05341-7

Yeung, A. T.; Hsu, C. N.; Menon, R. M. (2011). Electrokinetic extraction of lead from kaolinites: I. Numerical modelling. *Environmentalist.* 31, 26–32.