Effect of Cosolvent Concentration on the Hydraulic Conductivity of Electro-kinetic Remediated Crude Oil Contaminated

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Abstract
This study investigates the effect of cosolvent concentrations on the hydraulic Conductivity of electrokinetic remediation of crude oil-contaminated soil (COCS). The remediation process involved the use of acetone, distilled water, and graphite electrodes in electrokinetic remediation (EKR) setups, with varying cosolvent concentrations of 0.05M, 0.1M, 0.15M, and 0.2M. Initially, the total petroleum hydrocarbon (TPH) content in the contaminated soil was 48,000 mg/kg. The percentage removal efficiencies were 52.50%, 78.33%, 85.00%, and 64.58% for cosolvent concentrations of 0.05M, 0.1M, 0.15M, and 0.2M, respectively. The highest crude oil removal efficiency was achieved with the 0.15M concentration, demonstrating that this concentration was the most effective for EKR. Additionally, the index properties of the COC soil and EKR-treated soil classified the soil as A-7-6 under the AASHTO classification system (AASHTO, 1986) and as CL (clay of low plasticity) under the Unified Soil Classification System (ASTM, 1992). The hydraulic conductivity test was carried out by the falling head permeameter. The results of the hydraulic conductivity test for the three compactive efforts for COCS and EKR soil with 0.05M, 0.1M, 0.15M, & 0.2M cosolvent concentration, revealed that all values, except for COCS soil at 0% optimum moisture content (OMC), were below $1x10^{-9}$ m/s, meeting the regulatory threshold for landfill liners and covers in waste containment facilities.

Keywords: *Cosolvent, Cosolvent Concentration, Remediation, Hydraulic Conductivity,*

1. INTRODUCTION

Crude oil contamination of soil presents an unavoidable environmental challenge with serious consequences for ecosystems and human health (Adeloye et al., 2020). Traditionally, conventional remediation methods, such as excavation and landfill disposal, have been employed to tackle this issue; however, these approaches are often prohibitively expensive, environmentally disruptive, and may not address the root cause of the contamination (Adebajo & Adelaja, 2021). Due to these challenges, innovative approaches like electro-kinetic remediation have emerged as promising solutions for effectively cleaning hydrocarbon-contaminated soils (Ishaq et al., 2022). This presentation delves into the critical role of cosolvent concentrations in electro-kinetic remediation processes and their significant impact on transforming contaminated soil into a manageable waste material. This method offers a sustainable and economically viable solution to the problem of crude oil-contaminated soil. Crude oil spills, leaks, and improper disposal practices have led to widespread soil contamination worldwide. These incidents threaten terrestrial and aquatic ecosystems, pose risks to human health due to exposure to toxic substances, and compromise the structural integrity of the affected areas (Ezeokoli et al., 2019). Addressing the remediation of petroleum-contaminated soil has become a critical environmental concern. Traditional remediation

methods, such as excavation and off-site disposal, have significant limitations. They are often prohibitively expensive due to high transportation and landfill fees, cause substantial ecological disruption, and may merely shift the pollution problem to another location (Adebajo & Adelaja, 2021). Electro-kinetic remediation is indeed a promising technique for soil remediation, especially when dealing with contaminated sites where traditional methods might be too disruptive or ineffective. By utilizing electrochemistry, electro-osmosis, and electro-migration, this method can effectively mobilize and concentrate contaminants for removal or neutralization. The process involves strategically placing electrodes in the contaminated soil, which creates an electric field. This field causes the movement of charged particles (ions) and water within the soil,
Facilitating the migration of contaminants towards the electrodes. These contaminants can then be collected or treated at the electrodes. This method is advantageous because it can be implemented *in situ*, meaning it can be conducted directly at the contaminated site without the need for extensive excavation or transportation of the soil (Ishaq et al., 2022). Additionally, it reduces environmental disturbance and can transform contaminated soil into a containment area for safe waste disposal (Adebajo & Adelaja, 2021).
However, it's important to note that the effectiveness of electro-kinetic remediation can depend on several factors, including the type of contaminants, soil properties, and the specific design of the remediation system (Zhou et al., 2020). As the field develops, further research and advancements could enhance its applicability and efficiency in various environmental contexts (Ahmed et al., 2023).

Cosolvents, which are typically natural mixtures such as alcohols or surfactants, are commonly added to electro-kinetic remediation processes to enhance the removal efficiency of hydrophobic contaminants like crude oil (Zhu et al., 2020). The choice of cosolvent and its concentration significantly affects the overall success of the remediation process. Higher concentrations of cosolvents can improve the solubility of hydrophobic contaminants, making them more amenable to electro-migration and extraction (Oni et al., 2021). However, excessive concentrations of cosolvents may lead to negative effects, such as the release of other hazardous substances from the soil or increased energy consumption (Ishaq et al., 2022). Therefore, optimizing cosolvent concentrations in electro-kinetic remediation is crucial for achieving effective soil cleanup (Zhou et al., 2020).
Absolutely, crude oil has had a significant impact on modern society, providing essential energy and contributing to economic development. However, the environmental consequences, especially in areas like the Niger Delta, can be severe. Contamination from oil spills and leaks can drastically affect soil quality, reducing its geotechnical properties and leading to problems like building failures. These issues highlight the need for effective management and remediation strategies to mitigate the environmental and structural impacts of crude oil contamination. Balancing the benefits of crude oil with its environmental risks is crucial for sustainable development. This issue highlights the critical knowledge gaps and research needs regarding the effect of Cosolvent concentration on the hydraulic conductivity of Electro-kinetic Remediated Crude oil Contaminated soil.

2.0 MATERIALS AND METHODS

2.1 Materials

The materials for this research work include:

i. Soil sample: Soil sample was collected from Obio-Akpor Local Government area of River State, South-South geopolitical Zone of Nigeria, within the coordinates of longitude 4º50’19’’N and latitude 7º4’8’’E of the equator.

ii. Crude Oil: The crude oil sample was collected from Kaduna Refinery Petrochemical Company Limited, Kaduna state Nigeria.

iii. Water: Tap water from the borehole provided near the Civil Engineering laboratory of the

iv. Distilled water: The distilled water was obtained from the Department of Fashion Design and Clothing Technology, Kaduna Polytechnic, Kaduna State.

v. Electrodes: 8mm diameter by 300 mm long graphite electrode rods, was obtained from a laboratory equipment store a Lagos Street in Kaduna State was used

vi. EKR Cell: With little modifications to the set-up adopted by Yu et al. (2019), electro-kinetic remediation cell made from clear Plexiglas paste 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, 530 mm by 200 mm by 300 mm adjoining the middle was used.

vii. Connecting Wires and Clips Flexible connecting wires and battery clips obtained from a local electrical store at Lagos Street in Kaduna Stare were used.

viii. Acetone obtained from a local laboratory store at Kano Road in Kaduna State was used

2.2 Equipment

The equipment used for this research work include

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

ii. H Multimeter

iii. pH meter

iv. Conductivity meter

v. Thermometer

vi. Sets of sieves

vii. Oven

viii. CBR machine

ix. Compaction mould and rammers

2.3 Methods

The contaminated soil was subjected to the following:

2.3.1 Total Petroleum Hydrocarbon (TPH) Test

The total petroleum hydrocarbon (TPH) is used for any mixture of hydrocarbons that are found in crude oil. There are several hundred of these compounds, but not all occur in any one sample. Crude oil, which is used to make petroleum products, can contaminate soil with many of its different chemicals. It is not practical to measure each one of these chemicals, which comprise hexane, benzene, toluene, xylenes, naphthalene and fluorine in the soil. As such, it is useful to measure the total amount of TPH in a soil. TPH is the sum of volatile petroleum hydrocarbon (VPH) also known as petrol (or gasoline) range organics (PRO or GRO), which includes

hydrocarbons from C6-C10 and extractable petroleum hydrocarbon (EPH), which range from C10-C28.
Several methods can he used in the determination of the TPH of a crude oil contaminated soil, which include: gravimetric method, infrared spectrometry (R), gas chromatography-flame ionization detection (GC-FID), and ultraviolet spectrophotometer (UV). The TPH contents of the crude oil contaminated soils and those of the mole concentration of electro-kinetic remediated soils in this research were determined using the gravimetric method (the Toluene cold extraction method). Exactly 5000 g of each contaminated and treated soil sample was dried at room temperature for 72 hours. Exactly 2500 g of exam sample was placed in a 50 ml beaker, into which was added 10 ml of Toluene. The mixture was manually stirred continuously for 30 minutes, left to stand in a fume cupboard for 2 hours and afterwards filtered using Whatman No. 42 filter paper. The residue was allowed to dry in an oven at 50 ^{0}C. The TPH was computed as:

$$TP\left(\frac{mg}{kg}\right) = \frac{(Wo - W_1)(mg)}{Wo\,(kg)} \tag{1}$$

Where;
W0 = initial weight of sample

W1 = weight of sample after solvent extraction.

2.3.2 Hydraulic Conductivity Test

The hydraulic conductivity test was carried out, as recommended by Head (1992), by the falling head permeameter. The specimen was compacted using three (3) compactive efforts and then soaked in a water tank for 24 hours to allow for maximum saturation. During the test, the specimen was restrained from swelling vertically during saturation. After saturation, the samples were then assembled with a falling head permeameter to carry out a hydraulic conductivity test, and permeation was done with a permeating liquid (tap water). During permeation, specimens were allowed to undergo free vertical swelling, and the test lasted for 6 hours. The changes in the hydraulic head were taken at (3) three-hour intervals (at zero, three and the sixth hour).
The hydraulic gradient that ranged from 5 to 15 was adopted, though variation of the measured coefficient of hydraulic conductivity with hydraulic gradients more than 100 was found insignificant in clay as reported by (Shackelford et al., 2000). Hydraulic gradients in the range of 25 to 100 have also been used by many researchers, like Jo et al. (2001), and Oluremi (2015) for clay and sand mix samples. The coefficient of permeability (k) was calculated from equation 2.

$$k = \frac{2.303aL}{At} \log_{10} h1/h2 \tag{2}$$

Where: a = the cross - sectional area of standpipe.
L = the Length of the specimen.
A = the cross – sectional area of the soil sample
h_1 and h_2 = Heights of standpipe's initial and final water levels.
t = Time in minutes.

Plate 1: Hydraulic conductivity apparatus used for the test.

3. RESULTS AND DISCUSSION

3.1 Characteristics of contaminated soil and Electro-kinetic remediated soil

The soil is classified as A-7-6 soil based on the American Association of State Highway and Transportation Officials (AASHTO) classification system (AASHTO 1986) and CL or OL soil based on Unified Soil Classification System (USCS) (ASTM, 1992). A summary of the properties of the crude oil contaminated soil and Electro-kinetic remediation soil is provided in Table 1 and that of the treated soil is provided in Table 2.

Table 1: Characteristics of COC soil and EKR soil with 0.05M, 0.1M, 0.15M & 0.2M) cosolvent concentrations.

Properties	COCS	EKR- 0.05M	EKR- 0.1M	EKR- 0.15M	EKR- 0.2M
% Passing BS sieve No. 200	47.00	51.00	44.00	44.00	44.00
Liquid limit, %	27.60	32.40	28.70	27.21	29.79
Plastic limit, %	19.40	18.60	15.30	16.79	14.21
Plasticity index, %	38.40	41.80	44.55	42.15	42.00
AASHTO classification	A -7 -6	A - 7 -6	A - 7 - 6	A - 7 - 6	A – 7 - 6
USCS classification	CL	CL	CL	CL	CL

Table 2: Total Petroleum Hydrocarbons of the soil Sample

S/no	Soil Sample	Weight Before Extraction (mg)	Weight After Extraction (mg)	TPH (mg/kg)	TPH Removal Efficiency (%)	EOF (ml)
1.	COCS	5,000.00	4,960.00	48,000.00	-	-
2.	0.05M	5,000.00	4,886.00	10,800.00	52.50%	1,970.0
3.	0.1M	5,000.00	4,948.00	9,600.00	78.33%	3,070.0
4.	0.15M	5,000.00	4,964.00	7,800.00	85.00%	4,219.0
5.	0.2M	5,000.00	4,915.00	6,200.00	64.58%	1,740.0

From the results obtained in the remediation efficiency, the percentage removals were 52.50% for 0.05M, 78.33% for 0.1M, 85.00% for 0.15M and 64.58% for 0.2M, respectively as shown in Figure 1. The best removal efficiency of crude oil occurred in EKR soil with 0.15M, (85.00%) when Acetone was employed as the reagent. The other removal efficiencies were (78.33%, 64.58% and 52.50%) for EKR soil with 0.1, 0.2, and 0.05 cosolvent concentration, respectively.

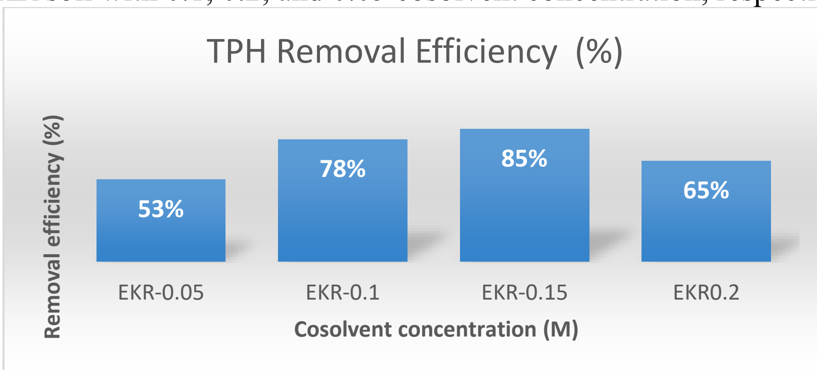

Figure 1: Total petroleum removal efficiency for EKR cosolvent concentration.

The remediation efficiency recorded could be because of the reagents that were employed as electrolytes, in the enhancement of crude oil contaminant desorption and ion migration. The remediation seems to be better for EKR soil with 0.15M which recorded 85.00%. However, the remediation efficiency was not as effective and efficient as the one reported by Sani et al. (2023), Cameselle et al. (2020), and Asadollahfardi and Razaee (2019). In light of this, using an effective cosolvent as an enhancement for the EK technique is very important.

3.2 Hydraulic Conductivity

Hydraulic conductivity is a key engineering parameter used to evaluate the performance of potential material for application as liner and cover system for engineered waste containment facility (Mitchell and Jaber, 1990). The results of the hydraulic conductivity test for the three compactive efforts for COC soil and EKR soil with 0.05M, 0.1M, 0.15M, & 0.2M cosolvent concentration, the graphical representation shown in figure 2 below. The hydraulic conductivity value of obtained for the COC soil were $4.65x10^{-12}$m/s, $1.13x10^{-8}$m/s, $1.06x10^{-11}$m/s, and $1.25x10^{-11}$m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively for BSL compactive effort, for WAS compactive effort the value obtained were $3.67x10^{-12}$m/s, $7.39x10^{-12}$m/s, $8.56x10^{-12}$m/s, and $1.10x10^{-11}$m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively. For BSH compactive effort the values obtained were $3.74x10^{-12}$m/s, $7.63x10^{-12}$m/s, $6.90x10^{-12}$m/s, and $7.83x10^{-12}$ m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively, the graphical representation shown in figure 2 below. The result indicates that there is an increase in hydraulic conductivity as moulding water content (MWC) increased. These results agree with the findings reported by Shackelford et al., 2000; Osinubi and Amadi, 2009). Generally, a low hydraulic conductivity value, less than $1x10^{-7}$ m/s is considered adequate by most regulatory agencies (Daniel and Benson, 1990: Daniel and Wu, 1993).

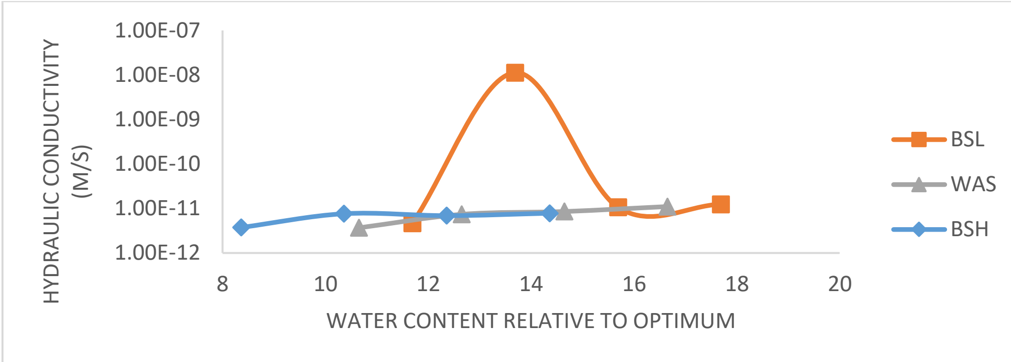

Figure 2: Variation of hydraulic conductivity with water content relative to optimum for COC soil.

The hydraulic conductivity value of obtained for EKR soil with 0.05M cosolvent concentration were $4.42x10^{-12}$m/s,$8.99x10^{-12}$m/s, $9.61x10^{-12}$m/s, and $1.14x10^{-11}$m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively for BSL compactive effort, for WAS compactive effort the value obtained were $3.92x10^{-12}$m/s, $7.26x10^{-12}$m/s, $6.85x10^{-12}$m/s, and $1.81x10^{-12}$m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively. And $3.09x10^{-12}$m/s, $1.07x10^{-11}$m/s, $2.50x10^{-11}$m/s, and $2.72x10^{-11}$ m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively were obtained for BSH compactive effort. The highest and the lowest values obtained from BSH compactive effort the values were $2.72x10^{-11}$ m/s and $3.09x10^{-12}$m/s, at 4%OMC and -2%OMC respectively, the graphical representation shown in figure 3 below. The result also indicates that there is an increase in hydraulic conductivity as moulding water content (MWC) increased. These results agree with the findings reported by Shackelford et al., 2000; Osinubi and Amadi, 2009). Generally, a low hydraulic conductivity value, less than $1x10^{-7}$ m/s is considered adequate by most regulatory agencies (Daniel and Benson, 1990: Daniel and Wu, 1993).

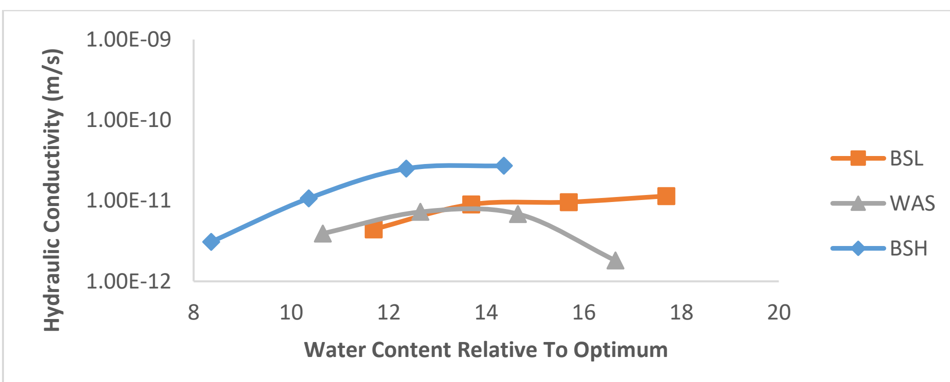

Figure 3: Variation of hydraulic conductivity with water content relative to optimum for EKR soil with 0.05M cosolvent concentration.

values obtained The hydraulic conductivity values of obtained for EKR soil with 0.1M cosolvent concentration were $4.17x10^{-12}$m/s,$8.18x10^{-12}$m/s, $8.72x10^{-12}$m/s, and $1.07x10^{-11}$m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively for BSL compactive effort, for WAS compactive effort the values obtained were $3.10x10^{-12}$m/s, $6.40x10^{-12}$m/s, $5.96x10^{-12}$m/s, and $6.37x10^{-12}$m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively. And $1.11x10^{-11}$m/s, $1.48x10^{-11}$m/s,

$1.34x10^{-11}$m/s, and $7.45x10^{-12}$ m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively were obtained for BSH compactive effort. The highest and the lowest were $1.42x10^{-11}$ m/s and $3.10x10^{-12}$m/s, at 0%OMC and -2%OMC from BSH and WAS respectively, the graphical representation shown in figure 4 below. The result also indicates that there is an increase in hydraulic conductivity as moulding water content (MWC) increased. These results agree with the findings reported by Shackelford et al., 2000; Osinubi and Amadi, 2009). Generally, a low hydraulic conductivity value, less than $1x10^{-7}$ m/s is considered adequate by most regulatory agencies (Daniel and Benson, 1990: Daniel and Wu, 1993).

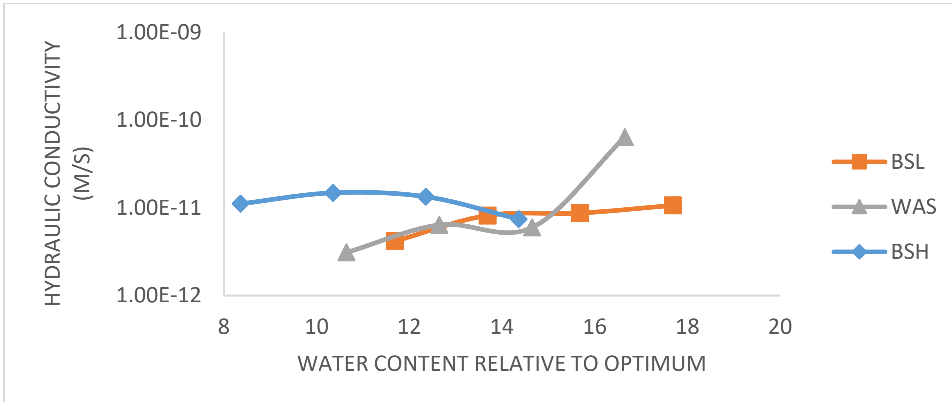

Figure 4: Variation of hydraulic conductivity with water content relative to optimum for EKR soil with 0.1M cosolvent concentration

The hydraulic conductivity values of obtained for EKR soil with 0.15M cosolvent concentration were $2.57x10^{-12}$m/s, $6.80x10^{-12}$m/s,$7.68x10^{-12}$m/s, and $7.70x10^{-12}$m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively for BSL compactive effort. For WAS compactive effort, the values obtained were $2.38x10^{-12}$m/s, $5.54x10^{-12}$m/s, $6.85x10^{-12}$m/s, and $6.48x10^{-12}$m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively. For BSH, compactive effort, the values obtained were $1.52x10^{-11}$m/s, $2.16x10^{-11}$m/s, $1.23x10^{-11}$m/s, and $5.64x10^{-11}$ m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively. The highest and the lowest values obtained were $2.16x10^{-11}$ m/s and $2.38x10^{-12}$m/s, at 0%OMC and -2%OMC from BSH and WAS respectively, the graphical representation shown in figure 5 below. The result also indicates that there is an increase in hydraulic conductivity as moulding water content (MWC) increased. These results agree with the findings reported by Shackelford et al., 2000; Osinubi and Amadi, 2009). Generally, a low hydraulic conductivity value, less than $1x10^{-7}$ m/s is considered adequate by most regulatory agencies (Daniel and Benson, 1990: Daniel and Wu, 1993).

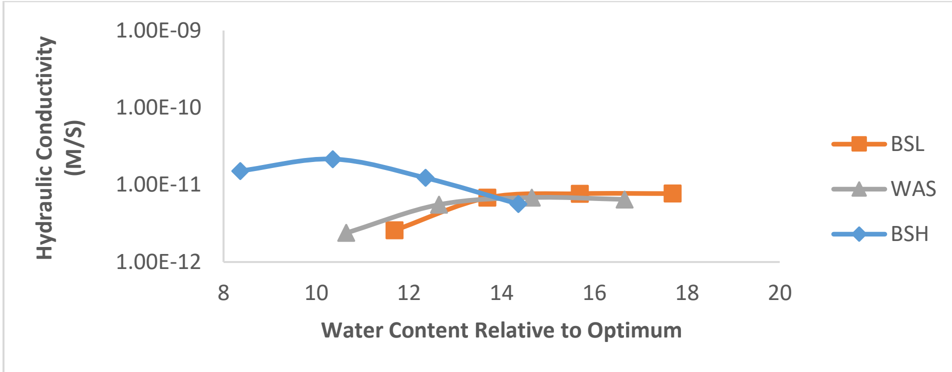

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Figure 5: Variation of hydraulic conductivity with water content relative to optimum for EKR soil with 0.15M cosolvent concentration

The hydraulic conductivity values of obtained for EKR soil with 0.2M cosolvent concentration were 3.20x10^{-12}m/s, 7.65x10^{-12}m/s,9.31x10^{-12}m/s, and 1.16x10^{-11}m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively for BSL compactive effort. For WAS compactive effort, the values obtained were1.96x10^{-12}m/s, 5.43x10^{-12}m/s, 7.68x10^{-12}m/s, and 1.26x10^{-11}m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively. For BSH compactive effort, the values obtained were 1.46x10^{-11}m/s, 1.96x10^{-11}m/s, 1.49x10^{-11}m/s, and 5.92x10^{-11} m/s at -2% OMC, 0%OMC, 2%OMC and 4%OMC respectively. The highest and the lowest values obtained were 1.96x10^{-11} m/s and 1.96x10^{-12}m/s, at 0%OMC and -2%OMC from BSH and WAS, respectively, the graphical representation shown in Figure 6 below. The result also indicates that there is an increase in hydraulic conductivity as moulding water content (MWC) increases. These results agree with the findings reported by Shackelford et al. (2000) and Osinubi and Amadi (2009). Generally, a low hydraulic conductivity value, less than 1x10^{-7} m/s is considered adequate by most regulatory agencies (Daniel and Benson, 1990; Daniel and Wu, 1993).

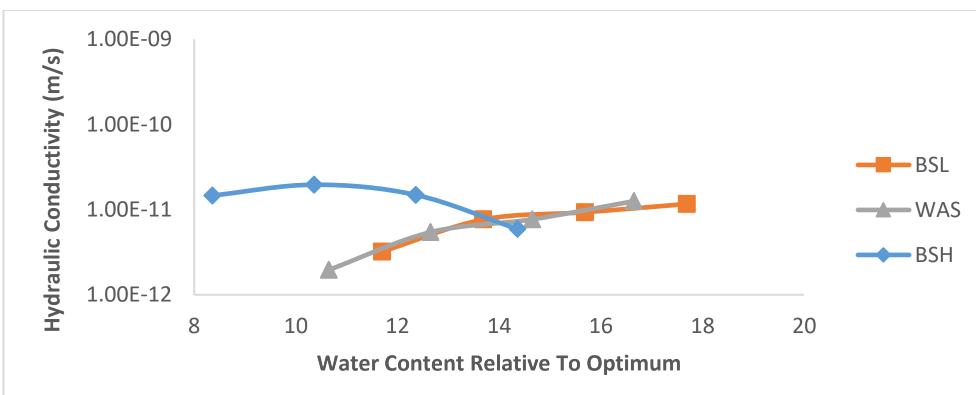

Figure 6: Variation of hydraulic conductivity with water content relative to optimum for EKR soil with 0.2M cosolvent concentration.

CONCLUSION

From the results obtained, the following conclusion can be drawn:

1. The highest recorded Total Petroleum Hydrocarbon (TPH) content in crude oil-contaminated soil was 85.00%, found in EKR soil at a cosolvent concentration of 0.15M when Acetone was used as the reagent.
2. The 0.15M cosolvent concentration proved to be both effective and efficient, and the values of hydraulic conductivity obtained at -2% OMC, 0% OMC, 2% OMC and 2% OMC for BSL, WAS and BSH satisfy the requirement of being less than $1x10^{-9}$ m/s and hence can be used for liner and cover for waste containment facility.

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