

Efficacy of Silica Encapsulation Technique in the Remediation of Hydrocarbon and Heavy Metal Contaminated Soils

> Asira Enim Enim; A. A. Uwakwe & B. A. Amadi Department of Biochemistry University of Port Harcourt, Rivers State E-mail: enimas1965@gmail.com

#### ABSTRACT

The study was carried out to determine the efficiency of silica encapsulation technology in remediation of soils contaminated with hydrocarbon and heavy metals to provide informative guidelines on the success of the technology. Soil samples at depths of 0-10cm, 10cm-20cm, 20-30cm, food crops, banana stem and eggshell were randomly collected from Bodo, Bomu communities (oil spill areas) and Apiapum (control) and taken to the laboratory for preparation, digestion, extraction and clean up. Methods were developed and validated for GC-FID and ICP-OES analysis of hydrocarbon and heavy metals respectively. Statistical tools for data analysis were mean, standard deviation, percentages, ANOVA and coefficient of correlation. Results revealed higher concentrations of hydrocarbon (8534.92 ±528.03 mg/kg/ in contaminated soil and all the heavy metal except Cd (1.05  $\pm$  0.03mg/kg) and Fe (260.13+3.83mg/kg in contaminated soil than control. In relation to soil texture, depth, and pH, the increasing order of efficiency of reduction of hydrocarbon by silica encapsulation was observed to be 295.83%>82%>76.81% for sandy, silt and clayey soils respectively; 92%>84%>78% for depths of 20-30cm, 10-20cm and 0-10cm respectively, and 92%>88%>77% for pH of 4.1, 5.6 and 8.3 respectively. Accordingly, the efficiencies of reduction of all heavy metals revealed increase in sandy except Ni (15%) followed by silt soil except  $Cd_{35}$ % and the least in clayey soil except Cd (50%). The highest efficiency of reduction of heavy metals was recorded by Cr and Fe/80% at depths of 0-10cm and the least was Pb/30% at depths of 0-10, 10-20cm, and 20-30cm, and in respect of pH, results showed that the efficiency of reduction of all heavy metals increased in the order, pH 4.1>pH 5.6>pH 8.3 except Fe at pH 8.3>5.6. The study has also shown that significant differences in efficiency of reduction of silica encapsulation exist between the soil texture, depth and pH. The research has therefore proven that silica encapsulation can be effectively used to remediate soil contaminated with hydrocarbon and heavy metals in an oil spilled area. Keywords: Heavy metals, hydrocarbons, efficiency, remediation soil, silica encapsulation

#### INTRODUCTION/LITERATURE

Never in the history of Nigeria had any socio-economic activities attracted huge attention of the world as the issues of petroleum oil spillage and its attendant effects on plants, humans, and the environment in Niger Delta of Southern Nigeria. Since the discovery and subsequent processing, production and transportation of petroleum oil products in commercial volume was made at Bomo by Dutch shell in 1959 and later at Bodo West, the people of Gokana LGA have had to contain with several oil spillages. From 1975-1997, more than 2, 976 cases of oil spills resulting in the discharge of more than 2.1 billion barrels of oil into the land (UNEP, 2011) had been recorded These spills were alleged to have arose from leakages of pipelines, underground and surfaces storage tanks and reckless disposal and management of waste and other by-products. Thus constituting the major sources of these hazardous and toxic substances into the soils has been known to constitute widespread danger (Chukwu *et al*, 2008; lwegbue *et al*, 2007) Ekundayo and Obiakwe, (2014). Accidental spills and improper disposal practices at contaminating sites have been known for contamination problems (USEPA, 2000).



Gokana LGA with well over of a population of 228.828 people (NPC, 2006), engaged in both commercial and subsistence agriculture can least be described as the ready sink for all the spills. The contaminated soil at Borny and Bodo sites require remediation in order to protect public health and the environment. The major disturbing factor stampeding clean – up of the contaminated site is the coexistence of hydrocarbons and heavy metals, as polycyclic aromatic hydrocarbon are difficult to treat due to their low volatility solubility and bio degradability (Hatheway, 2002). Though many existing remediation technologies are available for the treatment of PAH and heavy metals, the introduction of silica encapsulation technology in the remediation of mixed contaminants would open a significant environmental window in the quest for soil clean up. Silica encapsulation, based on its ability of converting soluble hazard organic and inorganic compounds with an acid resistant matrix to less toxic substances by adding an alkaline aqueous silica solution with emulsifying capacity to a hydrocarbon and chemically contaminated soil can permanently encapsulated the contaminants (Christen, 2005). The technology capacity is based on the ability of silica solution to penetrate porous surface, pulling any contaminate to the surface, were they are bonded with silica solution to form amorphous material, which later dries up to a find inert powder (Phelenani, 2007). It advantages range from cost and time effectiveness to permanently removal of the contaminants. Factors that affect silica encapsulation of heavy metals and hydrocarbons. The main factors that affect silica encapsulation are soil depth, metal charge and metal size; pH, and type of soil.

# Effect of Depth

Effect of soil depth on silica encapsulation and heavy metals is premised on the fact that changes in chemical composition of the soil decreases as the depth increases. For instance, percentage of sand decreases with increase in depth, while % of silt increases with increase in depth. This is because on average, weather becomes harsher as altitude increases, and weathering wears down soil to small particles (Charles, 1972). This heterogeneity could cause some sections treatment zone to receive more solution than others, hence increase soil solution required for remediation. Contaminant concentrations decreased significantly with increase in depth, since the surface is rich in contaminants than the underlying layer, greater accumulation in the top soil is probably due to the soil texture, ions, and mobility of contaminants in the soil (Yada *et al*,2002) Both sand and silt change uniformly as elevation gains. The % of sand decreases with altitude, while silt soil increases with altitude.

#### Effect of Metal charge and size on Silica Encapsulation

The metal charge and ionic size affect the encapsulation of soil contaminated with trace elements. The order of efficiency of encapsulation of metal ions can be represented as  $\mathcal{M}^{3+} > \mathcal{M}^{2+} > \mathcal{M}^{+} > \mathcal{M}^{\circ}$  this is attributed to the difference in sizes of metal ions. As the charge increases or the cation, the size of the particular metal ion gets smaller and it is easily encapsulated. Metal cations with high charge are therefore small in size than those with low charge and their encapsulation is enhanced by their small size. The effect of metal charge is more pronounced in magnesium, aluminum, calcium and potassium.



The order of encapsulation is  $Al^{3+} > Mg^{2+} > Ca^{2+} > K^+$ . Metal size does not affect the silica encapsulation of transition metals since the metals have almost the same sizes. Silica encapsulation of transition metals is mostly affected by the pH and soil type.

#### Effect of pH

Silica encapsulation of metals (trace) is more effective in acidic environment than in the basic environment. In acidic environment, silicate is protonated and can easily exchange with metals (trace), thus encapsulation of metals in acidic environment is enhanced. Low pH accelerates cation leaching from the soil and cation storage capacity decreases with decreasing pH. At low pH, silicon dioxide is protonated to form silanol. The protonation of silica leads to positively charged sites and poor adsorption of metals in soil surfaces. This causes metals to be available for encapsulation at low pH. Sinko (2010) observed that low pH favours hydrolysis, and that at pH >7 both hydrolysis and particle nucleation processes are dominated by condensation reaction, thus precursor molecules tend to aggregate into larger and denser particles. The larger particles in turn result in smaller surface area and larger pores. Studies done by Ortega reveal that silicate polymerization occurs where the samples are in acidic media. The degree of Cross-linking is directly proportional to the acidity of the soil (Ortega et al, 1991). In basic soil, reduction of metal concentration is limited. This can be traced to the polymerization of metal ions in high pH region. Metals tend to form polymeric ion or metal hydroxides, for instance  $Cu(OH)_2$  cannot fit into a regular crystal lattice. These metal hydroxides are known to block pores of silicate structure and decrease the retention of capacity (Algera *et al*, 2004). This means that at high pH, there would be no polymerization of silicate and the degree of cross-linking will be low, thus no significant encapsulation of metals will be observed. PH dependent negative charge increases with increasing pH causing soil surfaces to be negatively charged at high pH and adsorbed more metal ions. For hydrous silica, the pH dependent negative charge arises from ionization of weakly acidic surface silanol group. Si-OH  $S_{2}O^{-}+H^{+}$ Silica accepts a hydrogen ion to become a S.OH,<sup>+</sup> site having a positive charge or they release a hydrogen ion to become SiO<sup> $\cdot$ </sup> site having a negative charge. SiOH + H<sup>+</sup>  $SiO^{-} + H^{+}$  The concentration of  $S_{2}OH_{2}^{+}$  and  $SiO^{-}$  species depends SiOH,<sup>+</sup> SiOH on the pH of the aqueous phase. The  $S_1OH_2^+$  species increase at pH <7, while SO<sup>-</sup> species increases at pH>7. The presence of hydrocarbon in soil results in higher values of pH due to the slightly alkaline nature of the soil, and decreases the phosphorus of the soil (Obire and Nwaubeta, 2002).

#### Effect of Soil Type on Silica Encapsulation:

Silica encapsulation of transition metals has been found to be more effective in sandy soil than in clay soil. This has been traced to the following reasons: The relative simple composition of sandy soil. Clay soil particles have much larger surface area per unit volume than sandy soil. Also clay soils have few pores that are readily permeated by silica solution, so the useable surface area is quite small, thus silica encapsulation is less efficient in clay soil. Soil surfaces have to be positively charged for good encapsulation of metals, since oxygen in the silicate structure is the major anion that coordinates the



other cation, silicate surfaces have some permanent negative charge. This charge is due to imperfection in the crystal structure, due to their negative charge, silicate can attract bound cations to the surfaces (adsorption). Soils have high cationic exchange capacity (sum of exchangeable cation that soil can absorb) than sandy soil. lons with higher valency tend to exchange those with lower valency  $(Al^{3+}>Ca^{2+}>Mg^{2+}>K^+)$ . For ions of the same charge, the cation with the smallest hydrated radius is strongly absorbed because it moves close to the site of charge. The rate of ion exchange in the soil is affected by the type and quantity of organic and inorganic colloids. Metals adsorbed on clay soil are unavailable for encapsulation; they are then released when soil is subjected to harsh acidic or basic conditions. According to Marinescu *et al, (*2019), organic matter absorbs petroleum contaminants and through this process reduces its mobility, hinders biocidial activity and decreases bioavailability of the components for encapsulation. Also, plants exude soluble organic acid matter into the soil and in conjunction with organic matter potentially increase the adsorption of the petroleum (Cook and Hestenberg, (2013). Huessemann *et al*/(2004) observed that soil with low % of fine silt and clay demonstrated high degradation rate of hydrocarbon due to the aeration and porosity, which in turn depends on the soil texture. Xioa et al, (2004) reported that PAH were strongly bound to the organic matter since organic matter is an important binding agent for aggregation, the larger and more stable aggregates contain more PAH.

# Aim and Objectives of the study

#### Aim

The main aim of the study is to determine the efficiency of silica encapsulation technique in the remediation of soil contaminated with hydrocarbon and heavy metals.

#### Objectives

The present study is intended to achieve the following specific objectives. To determine the concentrations of hydrocarbon and heavy metals before and after silica encapsulation in the contaminated soils and compare their concentrations to uncontaminated soil. To determine the efficiency of reduction of hydrocarbon and heavy metals in the contaminated soil using silica encapsulation technology in relation to soil texture. To determine the efficiency of reduction of hydrocarbon and heavy metals in the contaminated soil using silica encapsulation technology in relation to soil depth. To determine the efficiency of reduction of hydrocarbon and heavy metals in the contaminated soil using silica encapsulation technology in relation to soil depth. To determine the efficiency of reduction of hydrocarbon and heavy metals in the contaminated soil using silica encapsulation technology in relation to soil depth. To

# MATERIALS AND METHODS

#### Materials

Beckman couller centrifuge machine (Canada), Calrius 690 gas chromatographed. (Perkin Elmer, UK), 200 induction couple plasmas – optical emission spectroscope (London, UK), magnetic stirrer (India), microwave digester (USA), ADIIO pH meter (Hangary), agate pestle, aluminium foil, 100ml and 200ml beakers, Bushi extraction machine, digestive tube, garden shovel, manual hammering hallow metal pipe, polythene bags, soil samples, CaCl<sub>2</sub> diphenylamine indicator, H<sub>3</sub>PO<sub>4</sub>, 0.5N Ferrous



ammonia sulphate solution, Conc.  $H_2SO_4$  1.0N  $K_2Cr_2O_7$ , amyl alcohol, ascorbic acid, 40% NaOH, NaF, 5% SDS, KHPO<sub>4</sub>, 0.5M NaHCO<sub>3</sub>, sodium hexa meta phosphate, sodium silicate, and water.

#### Methods

Soil sample of 100g each from two locations, each of the control (Apiapum in Obubra local government area of Cross River State and the research area (Bodo and Borny in Gokana local government area of River State) were collected from the depths of 0-10cm, 10cm and 20cm) with area measured 1km by 1km. The samples were put in polythene bags and taken the laboratory. In the laboratory, each soil sample was soaked in 200ml deionized water in a beaker overnight. Each soaked soil sample was mixed by using constant speed mixture (500 rpm). The complete mixed soil was sieved with micro sieve for 20minutes and kept for drying. Dried soil sample was pounded in an agate mortar and filtered with plastic vial. Soil sample was then digested with mixture of  $HNO_3$  and HF solution for 45 minutes at 175°C in a microwave digester. The digested soil was there after filtered and put in 100ml volumetric flask and made to 10ml with deionized water. Hydrocarbon extraction was carried out by soxhlet method (ASTM, 2005).. The metals extraction was done in line with procedure found in the literature (Mielke *et al*, 2004). Estimation of total hydrocarbon was done in line with USEPA method 418 (Schwarts et al, 2012) using GC/FID. Determination of heavy metals in the soil was conducted in line with EPA method using ICP-OES.

# RESULTS

Table1.0: Concentration in (mg/kg) of hydrocarbon in uncontaminated (control) and contaminated soils.

Constituent	Uncontaminated (control)	soil	Contaminated soil
TPH (mg/kg)	0.00± 0.00		*8,534.92± 528.03

\* Significant difference value. Mean  $\pm$  SD of triplicate of the samples Table2.0: Concentrations (mg/kg) of heavy metals in uncontaminated and contaminated soils.

Type of soil	Heavy metal/concentrations (mg/kg)							
	Cd	Cr	Cu	Fe	Mn	Ni	РЬ	Zn
Uncontaminate	*1.49	0.21	66.62	* 269.13	137.20	2.01	38.60	39.72
d soil	± 0.03	±0.03	± 2.69	$\pm 3.83$	±5.23	0.09	±3.93	±1.88
	1.05	<sup>*</sup> 15.41	* 153.0	226.72	* 401.30	*4.80	* 68.72	*193.50
Contaminated	± 0.11	1.43	22.41	24.56	± 71.46	± 0.51	±0.58	±7.47
soil								

\*Significant differences in values. Mean  $\pm$ SD of triplicates of the sample



# Table 3.1. Concentrations of hydrocarbon in the soil before and after Silica encapsulation in relation to soil depths

	Concentration (mg/kg)			
Soil depth (cm)	Before encapsulation	After encapsulation		
0 – 10	2616.00 ± 104.11	575.58 ± 64.84		
10 - 20	1262.00 ± 206.88	201.92 ± 16.47		
20 - 30	733.60 ± 22.00	$51.52 \pm 13.21$		

Values are meant  $\pm$  SD of triplicate determination of the sample.

Table 3.2 Concentration (mg/kg)	of hydrocarbon in relation to soil pH before and After
silica encapsulation	

	Concentration (mg/kg)			
Soil pH	Before Silica encapsulation	After Silica encapsulation		
4.I	255.76 ± 24.17	20.46 ± 1.42		
5.6	894.01 ± 60.31	107.28 ± 13.51		
8.3	$2300.83 \pm 289.39$	529,19 ± 20.70		

# Table: 3.3 Concentration (mg/kg) of hydrocarbon in the contaminated soil before and after silica encapsulation in relation to soil texture.

	Concentration (mg/kg)	Concentration (mg/kg)			
Soil texture	Before silica encapsulation	After silica encapsulation			
Clayey soil	$4531.68 \pm 90.83$	1045.00 ± 406.540			
Sandy soil	1,145.50 ± 608.49	*47.72 ± 27.22			
Silt soil	2852.44 ± 250.38	*513.44 ± 18.19			

\*Significant differences. Values are meant  $\pm$  SD of triplicate determination of the sample.

Table 4.1	Concentration of heavy metals in the contaminated soil before and after
silica enca	psulation in relation to soil depth (cm).

Heavy	depth/ concentration (mg/kg)					
metal	IOCM		10-20cm		20-30cm	
	Before	After	Before	After	Before	After
Cd	0.60±0.03	0.31±0.14	0.29±0.00	0.15±0.03	0.10±0.20	0.06±0.05
Cr Cu	7.75±0.55	*1.94±0.37	5.0±0.46	*1.54±0.08	3.69±0.40	*1.12±0.41
Fe	85.60±5.48	*34.26±3.37	50.0±2.58 136.70±2.6	*21.5±2.33 *25.59±3.50	17.50±0.67 19.22±1.05	*8.58±1.64 *7.69±0.58
Mn	136.70±8.18	*43.74±8.3 8	98.77±6.16 4.24±0.04	*34.57±12.08 1.65±0.46	65.93±2.49 1.24±0.22	*27.22±0.82 0.30±0.07
	236.60±17.74	*68.61±1.75		*8.91±2.13	,	4.20±1.49
РЬ	12.90±0.55	*4.52±0.31	14.60±1.10 42.18 <u>+</u> 3.21	*29.35±0.74	6.00±1.29 18.23±1.45	*9.48±1.0
Zn	48.12±1.29	* 27.43+2.0				
	112.93±0.82	*48.56±2.0				



Table 4 .2 Concentration (mg/kg) of heavy metals in the contaminated soil before and after silica encapsulation in relation to soil pH.

pH/ concentration					
pH	ł 4.1	pH 5.6		pH 8.3	
Before	After	Before	After	Before	After
0.09±0.03	0.06±0.02	0.11±0.02	0.08±0.02	0.17±0.04	0.13±0.04
	0.95±0.10				4.12±0.69
3.92±0.15	4.65±0.22	4.44±0.21	2.71±0.25	$6.58 \pm 0.16$	
			7.05±0.27		11.35±0.50
8.95±0.56		12.16±0.57			15.22±0.05
	5.46±0.34		8.43±0.19	18.02±2.79	_
13.99±1.26	64.4±2.05	<i>.</i>	96.59±1.55		158.50±2.22
		19.16±1.55			
$128 \pm 2.28$	0.25±0.01	0.1	0.38±0.02	28.14±3.39	0.58±0.05
	0	$172.48 \pm 3.43$	19.21±0.63		
0.43±0.02	$12.28 \pm 0.63$	. (	$64.44 \pm 6.1$	$255.64 \pm 5.06$	$30.59 \pm 3.70$
	$41.10\pm5.02$	0.00±0.03	4		98.45±1.33
21.17 + 1.50				$a^{9}a \pm a^{7}a$	
$21.1/\pm 1.50$		$30 cc \pm 1 37$		0.89 ± 0.01	
$60.52 \pm 4.0$		29.55 - 1.37		42.08 + 5.20	
00.33-4.0				43.00 - 3.39	
		88.03±1.01			
		94		123.06±3.37	
	$pH/concentration pH/Before 0.09 \pm 0.033.92 \pm 0.158.95 \pm 0.5613.99 \pm 1.26128 \pm 2.280.43 \pm 0.0221.17 \pm 1.5060.53 \pm 4.0$	pH/ concentration (mg/kg)   pH 4.1 After   Before After   0.09 $\pm$ 0.03 0.06 $\pm$ 0.02   0.95 $\pm$ 0.10 0.95 $\pm$ 0.10   3.92 $\pm$ 0.15 4.65 $\pm$ 0.22   8.95 $\pm$ 0.56 5.46 $\pm$ 0.34   13.99 $\pm$ 1.26 5.46 $\pm$ 0.34   0.43 $\pm$ 2.28 0.25 $\pm$ 0.01   0.43 $\pm$ 0.02 12.28 $\pm$ 0.63   21.17 $\pm$ 1.50 60.53 $\pm$ 4.0	$\begin{array}{c c c c c c c } pH/\ concentration\ (mg/kg) \\ \hline pH 4.1 & pH 5.6 \\ \hline Before & After & Before \\ \hline 0.09 \pm 0.03 & 0.06 \pm 0.02 & 0.11 \pm 0.02 \\ 0.95 \pm 0.10 & 0.95 \pm 0.10 \\ 3.92 \pm 0.15 & 4.65 \pm 0.22 & 4.44 \pm 0.21 \\ \hline 8.95 \pm 0.56 & 12.16 \pm 0.57 \\ 5.46 \pm 0.34 & 12.16 \pm 0.57 \\ 13.99 \pm 1.26 & 64.4 \pm 2.05 & 19.16 \pm 1.55 \\ 128 \pm 2.28 & 0.25 \pm 0.01 & 172.48 \pm 3.43 \\ 0.43 \pm 0.02 & 12.28 \pm 0.63 & \\ 41.16 \pm 5.62 & 0.60 \pm 0.03 \\ 21.17 \pm 1.50 & 29.55 \pm 1.37 \\ 60.53 \pm 4.0 & 88.03 \pm 1.91 \end{array}$	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$\begin{array}{ c c c c c c } \hline pH/concentration (mg/kg) \\ \hline pH/concentration (mg/kg) \\ \hline pH 4.1 & pH 5.6 & pH 8.3 \\ \hline Before & After & Before & After & Before \\ \hline 0.09\pm0.03 & 0.06\pm0.02 & 0.11\pm0.02 & 0.08\pm0.02 & 0.17\pm0.04 \\ 0.95\pm0.10 & & & & & & & & & & & & & & & & & & &$

Values are meant  $\pm$  SD of triplicate determination of the sample

Table 4.3 Concentration (mg/kg) of heavy m	netals in the contaminated soil before and
after silica encapsulation in relation to soil te	exture

Heavy	Concentration in mg/kg						
metal	Clay soil		Sandy soil		Silt Soil		
	Before	After	Before	After	Before	After	
Cd	0.20±0.02	0.10±0.07	$6.68 \pm 0.05$	0.06±0.01	$0.42 \pm 0.03$	0.42±0.03	
Cr	7.88±0.50	*4.96±0.50	$4.95 \pm 0.44$	2.38±0.35	$5.5 \pm 0.48$	*3.12±0.68	
Cu	93.13±3.82	*48.03±2.15	20.0 ±1.96	*1.65±0.5	$33.44 \pm 2.34$	*13.72±1.1	
Fe	146.70±6.16	*107.91±3.61	29.54±1.43	*14.77±0.3	49.40 ±3.61	9	
Mn	257.07±21.27	*141.39±1.65	52.23±6.0	2	187.46±12.6	*25.81±0.0	
Ni	$3.05 \pm 0.30$	*1.98±0.05	$1.10 \pm 0.53$	*19.34±1.6	$2.50 \pm 0.20$	I	
РЬ	$44.38 \pm 3.31$	*31.07±2.02	18.94±0.54	5	14.19 ±0.26	*97.48±3.5	
Zn	124.96±18.63	*84.94±2.10	25.21±3.21	*0.5±0.05	42.18 ±3.21	6	
				*4.74±0.18		1.55±0.04	
				*15.13±0.52		*9.59±0.25	
						*29.53±7.1	
						0	

Values are meant  $\pm$  SD of triplicate determination of the sample





Figure 1.0: Efficiency of reduction of petroleum hydrocarbon in the contaminated soil in relation to soil texture by silica encapsulation



Soil depth Figure :2.0 Efficiency of reduction of petroleum hydrocarbon in the contaminated soil in relation to soil depth by silica encapsulation





Soil pH

Figure 3.0 Efficiency of reduction of petroleum hydrocarbon in the contaminated soil in relation to soil pH by silica encapsulation



Figure 4.0.: Efficiency of reduction of heavy metals in the contaminated soil in relation to soil texture by silica encapsulation





Figure 5.0.0: Efficiency of reduction of heavy metals in the contaminated soil in relation to soil depth by silica encapsulation



Figure 6.0.: Efficiency of reduction of heavy metals in the contaminated soil in relation to soil pH by silica encapsulation

# DISCUSSION OF RESULTS

Table 1.0 showed very significantly (p < 0.05) higher concentration hydrocarbon in the contaminated soil (8534.92 ± 523.03mg/kg). Than uncontaminated soil.  $(0.00\pm0.00)$ . Table 2.0 revealed general increase in all heavy metal concentrations except Cd (1.05 ± 0.03mg/kg) and Fe (269.13+3.83mg/kg). Akamgbo and Jidere (2001) reported that oil spill leads to availability of macronutrient. In table 3.1, results revealed that the



concentration of hydrocarbon in the contaminated soil decreases with increase in depth before and after treatment with silica encapsulation. The depth of 20-30cm recorded the lowest concentration of hydrocarbon than other depths. Table 3.2 revealed higher reduction of hydrocarbon at pH 4.1 after silica encapsulation than at pH 5.6 and 8.3. Significantly lower concentration of hydrocarbon was left after silica encapsulation in sandy soil than clayey and silt soil (table 3.3) In table 4.1, it was observed that all the heavy metals were significantly  $(p \le 0.05)$  reduced at depth of 0-10cm Efficiency of reduction of hydrocarbon in the contaminated soil by silica encapsulation in relation to soil texture, depth, and pH: Figure 1.0 showed that the efficiency of reduction of hydrocarbon in the contaminated soil by silica encapsulation was higher in sandy soil (95.83%) than in silt soil (82%) and clayey soil (76.84%). This could be because sandy soil has larger particle size and pure sizes than clayey and silt soils. The larger the particle and pore sizes the less interfacial tension. The hydrocarbon concentration in the contaminated soil was observed to have decreased with increase in depth (Figure 2.0). This result was in tandem with the result obtained by Okop, 2010. This could be traced to decrease in pH as the depth increases in sandy. Different factors like soil texture and mechanical composition may also be responsible for the decrease in pH. Also, the efficiency of reduction of hydrocarbon in the contaminated soil was observed to decrease as the pH of the soil increases 92% > 85% > 77% for pH 41, 5.6 and 8.3 respectively (Figure4.0). The low efficiency of reduction recorded as pH increase may be due to the fact that silica deprotonates and has increased volume of porosity of encapsulation at lower pH. Secondly, in silica encapsulation, lower pH favours hydrolysis and particle nucleation process dominated by condensation reaction (Sinko, 2010). Efficiency of reduction of heavy metal in the contaminated soil by silica encapsulation in relation to soil texture, depth, and pH: Figure 4.0.0 revealed that the highest efficiency reduction of heavy metals was recorded by sandy soil in all the heavy metals except in Ni. This was followed by silt and clayey Soils.

The higher efficiency of reduction of heavy metals in sandy soil than silt and clayey soils could be attributed to the low affinity of heavy metals to sandy soil (Kabata-Pendias and Mukherjere, 2007), and large particle size and pore size of the sandy soil. Husamann et al (2007) reported that soil low in % of sill and clay soil demonstrate high degradation rate of hydrocarbon due to aeration and porosity. Also Falciglia et al 2011, while investigating soil textural behavior concluded that desorption efficiency is influenced by soil texture such as sandy soil showed highest desorption of contaminant. In figure 5.00, it was noticed that the highest efficiency of reduction of heavy metals was recorded in Fe (81%). at the depth of 10-20cm, while the least was recorded in Pb (30%) at depth, 20-30cm. however, efficiency of silica encapsulation was shown to be higher at 0-10cm depth in all the metals except Fe (10-20cm) and Ni (20-30cm). This probably could be as a result increase in organic acids produced by bacterial decomposition of organic matter at o-10cm depth, which promotes high mobilization of heavy metals. Figure 6.o. Revealed higher efficiency of reduction of heavy metals at pH of 4.1 than pH 5.6 and 8.3. The highest efficiency of reduction was recorded in Fe (95%) at pH 4.1 and the least was Zn (20%) at pH 8.3. generally, there was decrease in efficiency of reduction by silica



encapsulation as the pH increases. Ozombak and Cynthia, (1987) stated that at low pH, metal cations are mostly mobile under acidic condition and can easily be released, while anions tend to sorb to oxide of metals. This study also confirmed that metals with small atomic radii were encapsulated more than metals with larger atomic radii. The order of encapsulation of metals in the present work was observed to be Fe > Mn > Cu > Cr > Ni >Pb> Cd> Zn.

# CONCLUSION

There is no gain-saying that oil spillage in the research area has contaminated the soil with hydrocarbon and heavy metals. This spillage has corresponded affected the area, socially economically, environmentally and health wise as confirmed by the high concentrations hydrocarbon and heavy metals in the soil and their high pollution load indices in soil and food crops used in the study. It could confidently stated that silica encapsulation technologies can be used to effectively remedy soil contaminated with hydrocarbon and heavy metals also, it has been observed that soil texture, depth and PH determine the efficacy of otherwise silica encapsulation in remediating contaminated soil.

# RECOMMENDATIONS

This treatment train should be carried out in-situ to confirm the success or otherwise of silica encapsulation. Due to heterogeneity of the soil that caused increase in solution requirement, there should be control of mobility by the addition of polymers to stem the problem. For contaminants such as NAPLs that are extremely insoluble and cannot be mobilized by surfactants application, bio surfactants in addition to silica can facilitate their mobilization and eventual encapsulation. Following the success of the technology ex - situ, and to optimize its performance, it is suggested that further research be done to ensure effective mixing on-site equipment to encourage speedy reactive silica process. It is also recommended that this technology be applied to other soil or water contaminants. Urgent attention is needed to devise and implement appropriate means of monitoring hydrocarbon and heavy metal contaminants in food crops grown in all hydrocarbon spill areas to prevent their excessive build up in food crops. The qualitative and quantitative objective data obtained from this study are recommended for suitable remediation activity. Further development of the technology would require additional research evaluation its long-term performance under a range of environmental condition Field-based experiments and studies investigating potential adverse effects of silica treatment are necessary to demonstrate the safety, efficacy, and reliability of silica treatment of contaminated soil. Further development of the technology should require additional research evaluating its long-term performance under a range of environmental conditions.

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