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The Effect of Electrokinetic Remediation on the Index Properties of Lead-Contaminated Lateritic Soil

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ARTICLE INFORMATION

ABSTRACT

Soil contamination is a global concern that has great effects on human health, groundwater, and the engineering properties of soil. The aim of this paper is to use the electrokinetic remediation technique to remove lead contaminants from lateritic soil and evaluate the index properties of the remediated soil. Contaminated soil specimens were prepared by mixing the lateritic soil with a maximum 1% lead nitrate concentration in a stepwise increment of 0.2% by weight of dry soil. Thereafter, the soil was remediated in an electrokinetic cell using graphite electrodes with acetic acid and ethylenediaminetetraacetic acid (EDTA) as electrolytes. The results obtained shows a liquid limit and the plasticity index of electrokinetic lead remediated soil decreased from 52.00 to 39.60% and 29.09 to 17.59%, respectively. The cation exchange capacity (CEC) of the electrokinetic lead remediated soil increased from 5.31 to 8.20 Cmolkg-1 after the electrokinetic process, while the liner shrinkage decreased from 12.14 to 5.0 %. The percentage passing the BS NO 200 sieve ranges from 7.68 to 5.85 for British Standard Light (BSL), 5.09 to 5.50 for West African Standard (WAS), and 10.4 to 5.60 for British Standard Heavy (BSH). Furthermore, the percentage of gravel for BSL ranges from 1.6 to 14 for BSL, 1.6 to 13.4 for WAS, and 1.7 to 3.5 for BSH. Overall, the electrokinetic technique proved to be effective in removing lead from contaminated lateritic soil and improving its index properties. Results obtained provide valuable insight into the remediation of contaminated soil using the electrokinetic technique.

1. INTRODUCTION

Soil pollution will exist as long as humans interact with the soil. This interaction includes mining, ore processing, and smelting, agricultural additives, chemical storage, manufacturing, use of synthetic products like

insecticides, paints, and batteries; land application of industrial or domestic sludge and improper waste management (USDA NRCS, 2000). Heavy metals, radionuclides, and organic compounds are just a few of the toxic pollutants that have been discovered as contaminants. Through a variety of exposure pathways, the public and the environment are exposed to these pollutants at levels that are intolerable and have an unacceptably negative impact on both public health and the environment. Environmentalists and regulatory agencies now view the remediation of these sites as a top priority (Reddy and Camesella 2009).

Lead is one of the most common heavy metals in nature. It is very dangerous to living things and has never been an essential nutrient. Because of this, it is very important for human health to keep an eye on its level in the environment. When lead builds up in the body, it becomes more dangerous and hurts the nervous system. Batteries, cable sheathing, lead sheets, and pipes are the main sources of lead in the environment (Li et al., 2019). Although in some cases lead can be recovered and recycled, in most cases it is in compound form, as in paints and petrol additives, and is lost to the environment, landing in the aquatic environment (Wuana and Okieimen 2011). These lead salts enter the environment through the exhausts of cars. The larger particles will drop to the ground immediately and pollute soils or surface waters; the smaller particles will travel long distances through the air and remain in the atmosphere. Part of this lead will fall back to earth when it rains. The lead cycle that results from human production is much longer than the natural lead cycle. Lead pollution is a worldwide issue, with Nigeria having her fair share recently experienced lead poisoning in Zamfara State (North-West Nigeria) in 2010. The outbreak was described as the deadliest in contemporary history, killing 400 youngsters and gravely injuring perhaps 300 more (Dooyema et al., 2012; Elom et al., 2018). Additionally, lead poisoning was reported in Niger State (Central Nigeria), which resulted in the deaths of 28 children and the hospitalization of 65 others (Nigerian Vanguard 2015)

Heavy metal-contaminated soil has been cleaned up in the past in many different ways. soil washing (Ng et al., 2015; Jiang et al., 2015; Jiang et al., 2017; Jiang et al., 2018), Solidification/stabilization (Guo et al., 2017; Dai et al., 2018), phytoremediation (Bian et al., 2017), conventional chemical reduction and electrokinetic remediation (EKR) (Ferrucci et al., 2017). Electrokinetic remediation is a green technology and has numerous advantages, such as its flexibility for use ex-situ or in-situ, minimal soil disruption, applicability to low permeable and heterogeneous soils, applicability to saturated and unsaturated soils, and applicability for contaminant mixtures such as heavy metals (Han et al., 2021). The EKR technique has recently gained popularity among geo-environmentalists and the larger scientific community (Reddy and Camesella, 2009). Electrokinetic remediation is a process that involves the application of a direct current (DC) electric field across a wet mass of contaminated soil via electrodes positioned below the surface to drive contaminants to the negative electrode and then extract them from the soil. Electrokinetic remediation has been reported to be particularly beneficial when treating soils with low permeability, which are typically difficult to treat (Iannelli et al., 2015; Hassan et al., 2015; Yuan et al., 2017; Sun et al., 2019). Numerous electrokinetic phenomena that occur during the electrochemical remediation process, such as electro-osmosis, electromigration (ionic migration), and electrophoresis, involving the movement of soil pore fluid, charged contaminants in the dissolved phase, and colloidal size solid particles, require careful investigation in order to determine their effect on the EKR process. Similarly, the various components and physical designs of the EKR system continue to reveal previously unknown details about the remediation process's dependence on factors such as electrodes and electrolyte types and dimensions; chelating agent type and quantity; fixed and variable voltage and current; and the constraint of basic pH in the soil near the cathode caused by heavy metal precipitation (Tian, et al., 2017; Yan, et al., 2018; Gobadi et al., 2021). Electrokinetic remediation as a relatively new technique requires additional research in laboratory studies to be conducted on various problematic soils in order to investigate multiple changes in soil composition, the effects and degree of improvement of the soil with and without permeating different enhancement (stabilizing) agents through the soil, multiple changes in soil composition and chemistry, to characterize the soil's physicochemical and engineering properties after EK treatment, and to determine if the remediated soil may be used for engineering. The aim of the work is to apply the electrokinetic technique to remove lead ions from contaminated lateritic soil and thereafter determine the index properties.

2. MATERIALS AND METHODS

2.1. Materials

The lateritic soil used in this study was collected by the method of disturbed sampling from a lateritic soil formation located in Shika, Zaria, Kaduna State, Nigeria (Latitude 11°15'and longitude 7°45'E). The soil sample was collected at depths between 1.5 m and 2.0 m corresponding to the B-horizon usually characterized by accumulation of material leached from the overlying A-horizon. The soil samples collected were then air-dried before being pulverized to obtain particles passing BS No Sieve (4.76 mm aperture). The following high-quality reagents were used in this experimental investigation: lead nitrate Pb $(NO_3)_2$ (99% purity) supplied by Guanghua Sci-Tech Co., Ltd, disodium ethylenediaminetetraacetate ($Na_2C_{10}H_{14}O_8$ $N2.2H_2O$) (molecular weight 372.24) supplied Merck KGaA Germany, aqueous nitric acid (HNO_3) supplied by EM Science, Gibbstown, NJ, USA and acetic acid (CH_3COOH) (98.9% analytical reagent grade) supplied by Guanghua Sci-Tech Co., Ltd. The following equipment were used: KPS605D DC power supply, supplied by E-CrossStu GmbH China, PHS-25C Microcomputer PH meter supplied Hangzhou Qiwei Co., Ltd, NITRON 3000 X-ray fluorescence supplied by Thermo Fisher Scientific and HP 02 Peristaltic pump supplied by Grothen China.

2. Methods

2.2.1. Artificial contamination of soil sample

Soil contamination with lead was carried out at 0%, 0.2%,0.4%, 0.6%, 0.8% and 1% by weight which correspond to 2000 mg/kg, 4000 mg/kg, 6000 mg/kg, 8000 mg/kg, and 10000 mg/kg according to past works by previous researchers (Udiba et al., 2020; Tirima et al., 2021). Five separate plastic containers were filled with soil weighing a total of 117 kg each. The soil sample in each bucket was thoroughly mixed with lead nitrate at a stepwise increment of 0.2% ranging from 0% to 1% based on the sample's dry unit weight in accordance with Resmi et al. (2011) and Karkush et al. (2013). The mixture was left for a month in the laboratory to achieve a homogeneous saturation of lead nitrate. After 30 days, the contaminated soil was stirred for 10 minutes in a mixer to evenly distribute the lead nitrate adequately throughout the soil (Li et al., 2021). The soil was then air-dried in large trays at room temperature until they were dried then pulverized and stored in polyethene bags after passing through a 2 mm sieve.

2.2.2. Set-up and calibration of electrokinetic cell

The test equipment consists of an acrylic plastic electrokinetic remediation (EKR) cell, a DC power supply, a peristaltic pump, an overflow collection system, and a data acquisition system. The EKR cell measures 40 cm (length), 20 cm (width), and 30 cm (height). The dimensions of the soil compartment (30 cm x 20 cm x 30 cm) and the electrolyte chambers (5 cm x 20 cm x 30 cm). The graphite electrode measured 8 mm in diameter and 30 cm in length. There was a total of six electrodes. Porous plexiglas separated the soil compartment from the electrolyte compartment. On the porous plate, the holes were 5 mm in diameter and 2 mm apart. In order to prevent soil particles from entering the electrolyte chamber during the test, 40 cm diameter Whatman filter paper was placed between the porous plate and the soil. The top and bottom of the electrolyte chamber were equipped with two control valves. The top valve was designed to collect the electromotive flow fluid, while the bottom valve pumped the electrolyte into the electrolyte chamber. The entire setup is depicted in Plate 1.

Plate 1: Electrokinetic setup

2.2.3. Soil sample preparation

A digital scale was used to measure three numbers of 15 kg of the contaminated soil for each of the three cells that were constructed. The soil was mixed with the amount of water content of the liquid limit and was mixed properly for homogeneity and hereafter compacted into electrokinetic cells with a plastic rod 50 mm in length and 10 mm in diameter. A DC power supply was used to apply a voltage of 30 volts across the electrodes to achieve 1 volt per centimetre according to Yeung (2011). Both the anolyte and the catholyte consisted of 0.1 m acetic acid. After the test, four thin metal sheets were inserted with a 6-centimetre gap between them into the chamber containing the soil sample, and the soil was divided into five equal parts. Each soil part was carefully excavated with a spatula, the areas from the cathode to the anode were sequentially labeled S1–S5, and the soil was dried for testing.

2.2.4. Laboratory tests

The dried and sieved lead-contaminated, and electrokinetic lead-remediated soil was used for particle size analysis, and Atterberg limits, the above tests were carried out according to British Standards (BS 1377, 1990, Part 2) and (BS 1924 1990) respectively. The cation exchange capacity (CEC) was carried out according to Peck et al. (1974). The pH of the electrokinetic lead remediated soil was carried according to (ASTM 2012) and the oxide composition using Nitron 3000 X-ray fluorescence was supplied by Thermo Fisher Scientific.

3. RESULTS AND DISCUSSION

3.1. X-Ray Fluorescence

The XRF test results are given in Table 1. X-ray fluorescence tests were carried out on the natural and electrokinetic lead-remediated soils. Silicon dioxide (SiO_2), iron (III) oxide (Fe_2O_3), aluminum oxide (Al_2O_3), calcium oxide (CaO), and manganese (MnO) increased as the level of lead concentration increased. In addition, new compounds such as lead oxide (PbO), rubidium (RbO), copper (II) oxide, and tungsten trioxide (WO_3) are observed in the electrokinetic lead-remediated soil. This could be attributed to the fact that when soil is subjected to electrokinetic processes under varying chemical conditions, a complex combination of geochemical and dynamic processes develops within the porous media. These processes in the system chemistry alter the soil structure (arrangement, aggregation, orientation, and distribution of clay particles and pore spaces within a soil mass), as well as variations in the diffuse double layer, ionic concentration, and the subsequent formation of new compounds (Azhar et al., 2017).

3.2. pH

The pH of the soil is one of the most influential factors on the movement and transport of pollutants. Maintaining the pH level of the soil appears crucial. The changes in soil pH following remediation are depicted in Figure 1. From the anode to the cathode, the pH of the soil increased. The anolyte produced H+, which acidified the soil surrounding the anode, while the catholyte produced OH., which alkalinized the soil surrounding the cathode. The pH values of the soil were kept between 6.5 and 7.0. However, the final pH values were greater than the soil's initial pH value prior to remediation. During the remediation process, H+ produced by the anode migrated from the anode to the cathode as a result of electromigration. This process resulted in a gradual decrease in soil pH levels. It aided in the desorption and elimination of heavy metals (Kamal et al., 2021).

3.3. Cation Exchange Capacity

Figure 2 show that the cation exchange capacity of remediated soil in the range of 0.2 to 1.0% concentration of the lateritic soil increased from 5.31 cmol.kg-1 to a peak value of 8.20 (cmol.kg^{-1}). This could be attributed to an exchange of ions occurring in the diffuse double layer of the clay mineral (Sen et al., 2016). Ions with a valency of 1 are exchanged by ions of higher valency like calcium, magnesium, aluminium, and iron. This causes a reduction in the hydration of the ions and hence a reduction in the thickness of the double layer, which improves the properties of the soil. (Megur and Rakaraddi 2014).

3.4. Particle Size Distribution

The lateritic soil is reddish brown both the natural (0%) and the electrokinetic lead remediated soils fall under the A-7-6 soil group of the AASHTO soil classification system. (AASHTO 1986). For Unified Soil

Classification System (ASTM, 1992), the natural soil (0%) falls under CH, while 0.2 %, 0.4 %,0.6%, 0.8%, and 1.0 % were classified as CL group. Table 2 provides a summary of the characteristics of both natural and electrokinetic lead-remediated soil. Figures 3, 4, and 5 show the natural and electrokinetic lead remediated soil's particle size distribution curve. It was observed that after remediation at British Standard Light (BSL), West African Standard (WAS), and British Standard Heavy (BSH), it shows 82.15% of fines passing through the number 200 sieve for the natural soil and for the electrokinetic lead remediated soil at the various percentages. There was a drastic reduction in the percentage of fines passing through the same sieve and this could be attributed to the flocculation of the clay particles, and the change in soil fabric and structure.

Table 1: Oxide composition of the natural soil and electrokinetic lead remediated soil

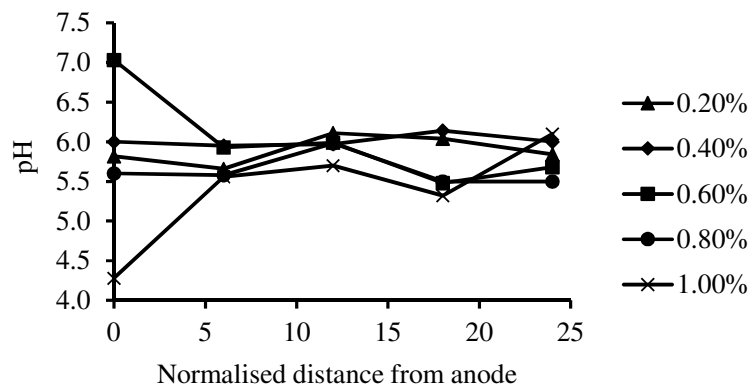| | Lead concentration | | | | | | | | | | | | | | | |
| | 0 (%) | 0.2 (%) | | | 0.4 (%) | | | 0.6 (%) | | | 0.8 (%) | | | 1.0 (%) | | |
Oxide		S1	S3	S5	S1	S3	S5	S1	S3	S5	S1	S3	S5	S1	S3	S5
SiO_2	52.309	55.878	53.124	53.357	50.091	53.375	52.795	50.234	54.225	55.095	53.378	55.181	55.493	54.967	53.521	46.401
V_2O_5	0.081	0.113	0.083	0.110	0.109	0.110	0.101	0.105	0.100	0.091	0.094	0.088	0.102	0.111	0.119	0.104
Cr_2O_3	0.004	0.014	0.011	0.021	0.025	0.021	0.029	0.020	0.025	0.022	0.026	0.020	0.025	0.017	0.021	0.018
MnO	0.054	0.066	0.053	0.056	0.071	0.056	0.077	0.057	0.061	0.053	0.054	0.062	0.065	0.054	0.060	0.079
Fe_2O_3	11.499	11.324	12.765	11.510	12.126	11.510	12.330	11.796	11.985	11.000	11.818	11.330	11.884	11.257	12.010	11.399
CO_3O_4	0.054	0.045	0.055	0.051	0.055	0.051	0.063	0.051	0.049	0.045	0.048	0.055	0.051	0.048	0.051	0.040
NiO	0.000	0.003	0.005	0.000	0.003	0.000	0.001	0.004	0.001	0.000	0.002	0.002	0.000	0.002	0.000	0.004
CuO	0.043	0.044	0.062	0.045	0.057	0.045.	0.047	0.059	0.040	0.036	0.046	0.040	0.047	0.034	0.041	0.073
Nb_2O_3	0.017	0.012	0.034	0.018	0.024	0.018	0.013	0.028	0.012	0.017	0.022	0.012	0.016	0.017	0.013	0.021
MoO_3	0.004	0.005	0.005	0.003	0.016	0.003	0.004	0.010	0.003	0.001	0.005	0.007	0.000	0.004	0.002	0.003
WO_3	0.000	0.001	0.004	0.000	0.007	0.000	0.000	0.002	0.004	0.005	0.001	0.000	0.007	0.000	0.000	0.001
P_2O_5	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.049	0.000	0.00	0.000	0.000	0.00	0.000
SO_3	0.000	0.000	0.000	0.000	0.000	0.000	0.039	0.000	0.000	0.000	0.000	0.00	0.000	0.080	0.00	0.000
CaO	0.049	0.077	0.118	0.035	0.105	0.035	0.265	0.086	0.023	0.123	0.019	0.055	0.101	0.043	0.027	0.265
MgO	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
K_2O	0.527	0.524	0.525	0.497	0.535	0.497	0.538	0.423	0.506	0.523	0.473	0.442	0.476	0.541	0.480	0.487
BaO	0.014	0.002	0.000	0.000	0.023	0.000	0.062	0.028	0.040	0.000	0.055	0.044	0.000	0.000	0.018	0.045
Al_2O_3	32.990	27.696	29.439	31.072	29.583	31.072	30.674	28.241	29.780	30.059	30.454	29.193	28.272	29.793	30.581	23.984
Ta_2O_5	0.028	0.025	0.018	0.025	0.023	0.025	0.022	0.004	0.009	0.017	0.021	0.014	0.001	0.013	0.025	0.000
TiO_2	1.502	2.014	1.955	1.738	1.944	1.738	1.892	1.718	1.691	1.633	1.829	1.732	1.923	1.667	1.814	1.797
ZnO	0.002	0.005	0.005	0.007	0.012	0.007	0.003	0.008	0.004	0.004	0.007	0.005	0.174	0.018	0.001	0.007
Ag_2O	0.014	0.041	0.022	0.020	0.036	0.020	0.022	0.023	0.043	0.034	0.031	0.036	0.037	0.036	0.010	0.041
Cl	0.650	0.641	0.934	0.616	0.744	0.616	0.651	0.593	0.532	0.529	0.624	0.581	0.657	0.580	0.607	0.701
ZrO_2	0.157	0.266	0.346	0.265	0.352	0.265	0.274	0.313	0.258	0.243	0.236	0.261	0.289	0.256	0.277	0.354
RbO		0.011	0.015	0.011	0.016	0.011	0.007	0.008	0.009	0.007	0.007	0.004	0.007	0.007	0.005	0.011
PbO		0.033	0.421	0.543	4.040	0.543	0.091	6.186	0.603	0.415	0.749	0.834	0.425	0.438	0.317	14.164

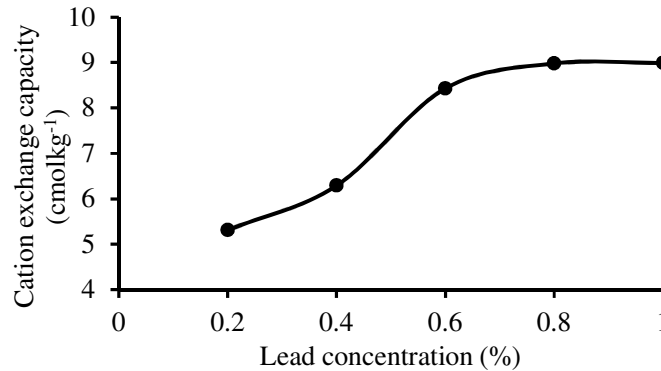Figure 1: Soil pH with normalized distance from the anode

Figure 2: Variation of cation exchange capacity of electrokinetic soil with lead concentration

Table 2: Index properties of natural and electrokinetic lead remediated soil

Engineering properties	Natural soil	0.2% EKR	0.4% EKR	0.6% EKR	0.8% EKR	1.0 EKR
Natural moisture content (%)	22.70					
Liquid limit (%)	52.00	49.40	48.00	47.60	47.00	39.60
Plastic limit (%)	22.93	22.01	29.02	29.49	29.24	22.01
Plasticity index (%)	29.07	27.39	18.92	18.12	17.76	17.59
Linear shrinkage (%)	12.14	11.43	10.00	9.00	7.60	5.00
Percentage passing BS No. 200 sieve (BSL)	82.15	7.68	8.50	9.75	4.60	5.85
Percentage passing BS No. 200 sieve (WAS)	82.15	5.09	9.78	9.06	10.30	5.50
Percentage passing BS No. 200 sieve (BSH)	82.15 A-7-6	10.4	14.8	7.73	10.30	5.60
AASHTO classification, USCS classification specific gravity MDD (Mg/m^3)	1.68					
Standard Proctor	1.76	1.78	1.78	1.79	1.74	1.86
West African Standard	1.84	1.87	1.84	1.61	1.86	1.86
Modified proctor		1.96	1.97	1.97	1.96	1.94
OMC (%)	19.00					
Standard proctor	16.80	13.20	14.40	14.60	14.98	15.60
West African Standard	14.00	13.10	13.40	13.00	11.20	13.40
Modified proctor	4.20	10.40	9.40	10.60	10.80	11.66
pH	Brown	5.83	6.0	7.03	5.6	4.28
Colour	Muscovite					
Dominant clay mineral						

3.5. Atterberg Limits

Liquid, plastic, and shrinkage limits were determined for contaminated and electrokinetic remediated soil samples. The plasticity index was obtained by subtracting the plastic limit from the liquid limit. Figure 6 shows liquid, plastic, and liner shrinkage for the electrokinetic lead remediated soil, there was a great improvement after the process. The liquid limit value was 52.00, 49.40, 48.00, 47.60, 47.00, and 39.60 % and the plastic index values were 29.07, 27.39, 18.92, 18.12, 17.76, and 17.59 % respectively for soil samples 0, 0.2, 0.4, 0.6, 0.8, and 1.0%. The decrease in liquid limit and the plasticity index could be a result of new compounds formed, which could be attributed to changes in physical properties ascribed to electrochemical effects under the influence of an electrical gradient, such as electroosmosis, electromigration, electrolysis, hydrolysis, and electrode degradation (Lefebvre and Burnotte 2002). These processes are consistent from anode to cathode in all soil samples and contribute to the physicochemical alterations of treated soil. This electrochemical effect can also be linked to alterations in the clay-water-electrolyte system, which result in the dissolution of chemical compounds or clay minerals, ion exchange capacity, adsorption, desorption, complexation, precipitation, and mineralization (secondary mineral). (Kamal et al., 2021)
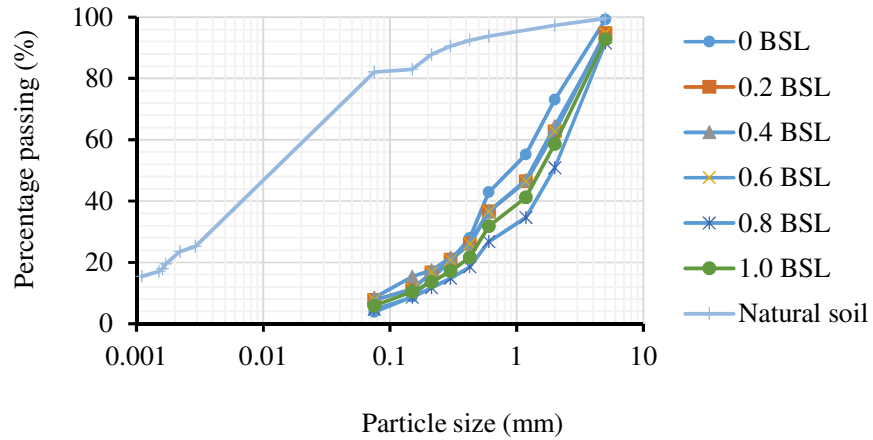
Figure 3: Variation of particles size distribution curve at the optimum moisture content at BSL
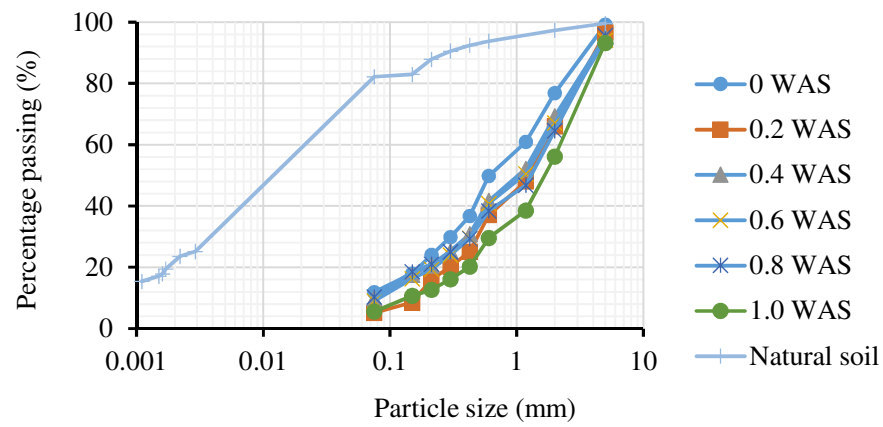

Figure 4: Variation of particles size distribution curve at the optimum moisture content at WAS
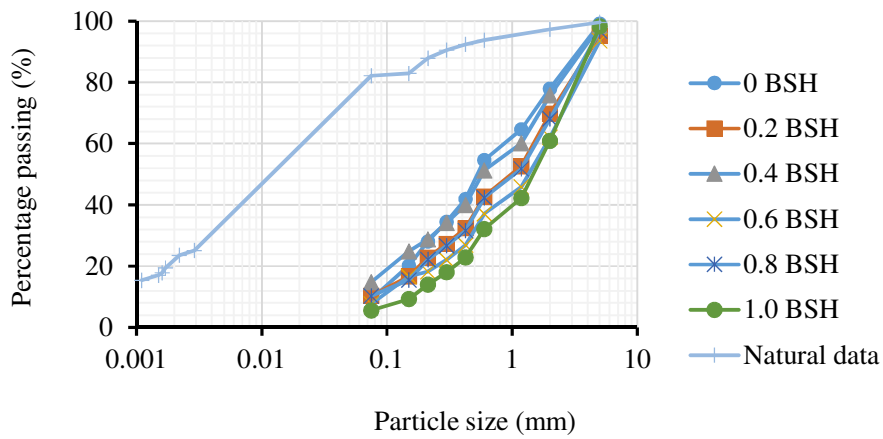

Figure 5: Variation of particles size distribution curve at the optimum moisture content at BSH
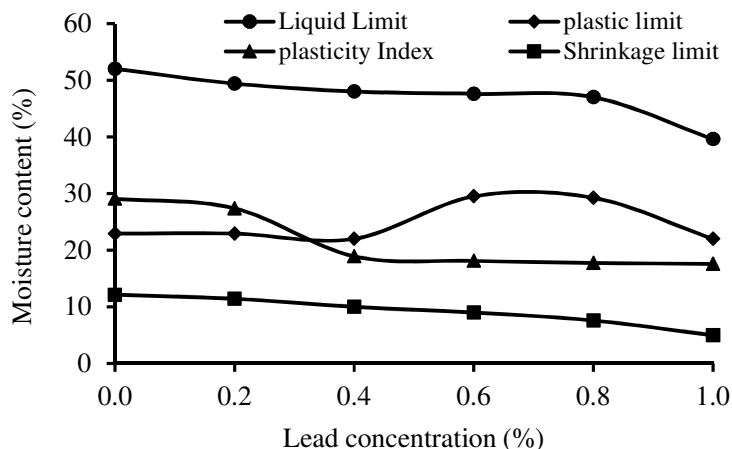
Figure 6: Atterberg limits for the electrokinetic lead remediated soil

4. CONCLUSION

Electrokinetic remediation is a green technology that can be used both to remove contaminants and improved the index properties of the soil. A decrease in the liquid limit will bring an improvement in the soil. In this present study, the liquid and the plastic limit decreased from 52.00 to 39.60 % and 29.07 to 17.59% respectively for 0.2 to 1.0 % electrokinetic lead remediated soil. The results obtained show that the soil could be used as a hydraulic barrier in a landfill since it has a plasticity index than 7%. The liquid and plasticity index of the studied soil is both greater than the recommended value which suggests that the liquid should be less than or equal to 30 % and the plasticity index should be less than or equal to 10% for suitability material for subgrade.

5. ACKNOWLEDGMENT

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6. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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