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NIGERIAN LATERITIC CLAY SOILS AS HYDRAULIC BARRIERS TO ADSORB METALS. GEOTECHNICAL CHARACTERIZATION AND CHEMICAL COMPATIBILITY

The suitability of using lateritic clays from Aviele and Igarra has been investigated both in the Northern part of Edo state, Nigeria as liners of an engineered landfill and to adsorb metals in leachates. Geotechnical characteristics, pH, and elemental composition for the lateritic clay samples were determined. The chemical composition, pH, total dissolved solids and electrical conductivity were determined for leachates collected from two dumpsites. The capacities of the lateritic clay soils to adsorb heavy metals in the leachates were determined using the batch equilibrium adsorption technique. The unconfined compressive strength (UCS) of soils were found to be sufficient to resist damage. By both the standard and modified Proctor compaction tests, it was found that the coefficients of permeability for the soil samples were lower than 1×10^{-9} m/s that is widely recommended for soils that are to be used as landfill liners. Pb²⁺, Zn²⁺ and Cr²⁺ were the heavy metals in the leachates. The sorption selectivity order for tested soils depended on the soil type and properties.

1. INTRODUCTION

Large quantities of solid wastes generated in many developing countries are indiscriminately discarded in dumpsites, which are environment-unfriendly. Over time, some of these wastes get decomposed, oxidized and corroded, releasing toxic substances (leachates and harmful gases) that contaminate underground water, air and soil. This contamination of the environment may lead to various human health implications and even, loss of lives.

An engineered landfill is a waste disposal technique that is most environment-friendly. It involves the use of clay liners that serve as hydraulic barriers to protect

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groundwater. In order to keep the cost of constructing a landfill as low as possible, it is important to use locally available materials as landfill liners. Though compacted clays are usually used as landfill liners, Guney et al. [1] stated that not all natural clays can provide good contaminant containment properties.

Wide acceptance of the use of compacted clays as landfill liners is largely based on experience in North America and Europe, with less investigation on the use of lateritic clayey soils found in other parts of the world [2, 3]. Lateritic soils are widespread in some countries in Africa such as Nigeria [4, 5]. Consequently, their use as landfill liners during the construction of engineered landfills will lower a landfill construction project cost. This is recommended wherever such lateritic soils are available. The aim of this research is to investigate the suitability of using lateritic clays from Aviele and Igarra in Edo state, Nigeria as liners of an engineered landfill and to adsorb metals in leachates from municipal solid wastes.

2. MATERIALS AND METHODS

The lateritic soil samples were collected from Aviele (latitudes 07°11′N and 07°15′N, and longitudes 06°29′E and 06°32′E) in Etsako West Local Government Area (LGA) (samples A) and Igarra (latitudes 07°27′N and 07°30′N, and longitudes 06°08′E and 06°11′E) in Akokoedo LGA, Edo State (samples B), Nigeria. The samples were collected at a depth of 1.5 m below the ground surface. Samples for determination of natural moisture content were collected in water-tight containers. The bulk samples (A and B) were taken altogether from 4 different geographical sample locations. Four different samples weighing about 2000 g each were taken from each location and thereafter homogenized together to have a soil sample with uniform composition.

Leachates, collected from two municipal solid waste dumpsites (Otofure and Iguomo dumpsites) in Benin City, Edo State, Nigeria, were used to evaluate the capacity of the lateritic soils to adsorb the heavy metals in the collected leachates. These dumpsites do not have leachate collection facilities but the leachates were collected using perforated PVC pipes placed at four different points at the base of each of the dumpsites. For each of the dumpsites, the collected leachate samples of about 1 dm³ in volume were mixed prior to its analysis. Heavy metals (lead, zinc and chromium) in the form of powdered oxides (PbO, ZnO, and CrCl₂) were weighed in varying quantities (ranging from 0.5 to 5 g/dm³ of PbO, ZnO and CrCl₂) and added to the collected leachates to increase metal concentrations.

The chemical composition, pH, total dissolved solids and electrical conductivity were determined for leachates collected from two dumpsites. The elemental composition of the soil samples was determined using a S1 TITAN Handheld XRF (X-ray fluorescence) spectrometer, produced by Bruker Corporation. A laboratory oven-drying method was used to determine the natural moisture content of the soil samples. The

particle size distributions of the soil samples were determined from sieve and hydrometer analyses. All other geotechnical tests, including specific gravity, Atterberg limits, compaction and unconfined compressive strength (UCS) tests were determined using a pycnometer, a Cassangrade's liquid device, a compaction mould and a rammer, and an unconfined compression tester (Proving Ring Type) – (UCA-05) according to procedures described in [6]. The permeabilities of the soil samples were determined using the falling head permeameter and in accordance with the procedure described in [7].

Batch equilibrium adsorption tests (BEATs) were performed in order to determine the capacity of the soil to adsorb the predominant cations in the leachate. The procedure used for carrying out the BEATs is similar to that used by Bello and Osinubi [8]. A soil-leachate ratio (by dry mass of soil in g/dm^3) of 1:4, which is the highest ratio recommended by USEPA [9] was employed. This ratio 1:4 was maintained by adding 50 g of soil and 0.2 dm³ of the leachate into a plastic container that has been rinsed with distilled water. The mixtures were subjected to shaking and a soil-leachate contact period of 48 h. The soil and leachates were afterward separated using filter papers. Cation concentrations in the leachates before and after this test were measured using iCE 3400 AAS atomic absorption spectrometer produced by Thermo Fisher Scientific. The uptake of each of the cations in the leachate, q (in mg/g) was calculated using equation

$$q = \frac{\left(C_0 - C_{\text{eq}}\right)V}{m} \tag{1}$$

where C_0 and C_{eq} represent initial and equilibrium (residual) concentrations of the considered metals in the leachate (mg/dm³), respectively, V represents the volume of the leachate (dm³), and m represents the mass of the lateritic soil in contact with the leachate (g).

The equilibrium adsorption isotherms for each of the metals were produced by plotting the metal sorption uptake (q) against the equilibrium concentration of the metal in the leachate (C_{eq}) . The slope of the adsorption isotherm, which is called the partition coefficient, K_p (dm³/g), was determined using equation

$$K_{p} = \frac{\Delta q}{\Delta C_{eq}} \tag{2}$$

The partition coefficient was used to determine the retardation factor, R_d , using equation:

$$R_d = 1 + \frac{\rho_d}{n_e} K_p \tag{3}$$

where ρ_d and n_e is the bulk density (g/dm³) and effective porosity of the in situ soil, respectively. The effective porosity represents the pore space available for liquid to flow.

3. RESULTS AND DISCUSSION

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The main elemental composition of the soil samples and leachates used in this study are presented in Tables 1 and 2, respectively. Aluminium and iron were found to be the major elements in both soil samples, which is characteristic of lateritic soils.

Table 1 Elemental composition of the soil samples [wt. %]

Element	Cor	Content			
Element	Soil A	Soil B			
Aluminum (Al)	80.6	86.3			
Titanium (Ti)	1.1	_			
Silica (Si)	3.5	3.3			
Iron (Fe)	13.6	7.5			
Zinc (Zn)	0.02	0.05			
Zirconium (Zr)	_	0.04			
Molybdenum (Mo)	0.03	0.07			
Manganese (Mn)	0.74	0.52			

 $\label{eq:Table 2} Table \ 2$ Elemental composition of the leachates [mg/dm³]

Element	Content			
Element	Leachate 1	Leachate 2		
Calcium (Ca)	105.20	80.54		
Potassium (K)	450.40	123.30		
Magnesium (Mg)	58.39	24.40		
Manganese (Mn)	0.30	0.30		
Lead (Pb)	0.22	0.10		
Zinc (Zn)	0.54	0.37		
Sodium (Na)	359.07	132.42		
Iron (Fe)	3.19	1.96		
Chromium (Cr)	0.05	0.04		

The predominance of aluminum in the soil samples confirms that soils have experienced laterization. The soil samples are likely to be old and highly weathered soils. Zinc, zirconium and molybdenum are other elements that were found in much lower quantities. Table 2 presents potassium, sodium, calcium, magnesium and iron as the predominant elements found in the leachates collected from the two dumpsites.

pH of leachates 1 and 2 were 5.3 and 5.9, electrical conductivities were 10.2 and 13.7 mS/cm and total dissolved solids - 3219 and 683 mg/dm³, respectively. Heavy

metals found in the leachates, including lead, zinc and chromium, were in small quantities (Table 2) because of the leachate dilution during the wet/rainy season when the samples were collected. Table 3 shows the concentrations of the Pb, Zn and Cr in the leachates (L1 and L2) after the addition of the heavy metal oxides) increasing metal concentrations.

 $T\,a\,b\,l\,e\,\,3$ Concentrations of the heavy metals in leachates [mg/dm 3]

C ₀ of heavy metal	Concentration after addition					
in oxide added to leachate	Pb in L1	Pb in L2	Zn in L1	Zn in L2	Cr in L1	Cr in L2
$[g/dm^3]$	[mg/dm ³]	[mg/dm ³]	[mg/dm ³]	[mg/dm ³]	[mg/dm ³]	[mg/dm ³]
5	5014	4938	5045	4986	5002	4994
4.5	4539	4496	4486	4537	4502	4500
4	4033	3996	3980	4007	4002	3990
3.5	3519	3502	3853	3731	3504	3485
3	3006	2997	3034	3000	2996	2990
2.5	2511	2506	2532	2495	2502	2493
2	2002	2001	2013	2009	2001	1990
1.5	1520	1469	1524	1503	1504	1501
1	996	921	1051	995	1005	996
0.5	508	496	548	513	497	497

Table 4
Properties of the soil samples

Durante	Soil samples			
Property	A	В		
Natural moisture content, % d. m.	10.0	8.0		
pH	5.4	5.7		
EC, mS/cm	10.7	13.2		
Specific gravity	2.7	2.6		
Liquid limit, %	51.5	54.0		
Plastic limit, %	27.0	26.7		
Plasticity index, %	24.5	27.3		
Linear shrinkage, %	12.1	11.0		
Bulk density, g/dm ³	1.9476	1.8559		
Effective porosity	0.41	0.46		
AASHTO classification	A-7-6	A-7-6		
USCS classification	CH	CH		

The characteristics of the soil samples are summarized in Table 4. The plasticity indices and the liquid limits were used to classify the soil samples by means of the

plasticity chart (Fig. 1). The particle size distribution obtained from the sieve and hydrometer analyses of the soil samples are presented in Table 5.

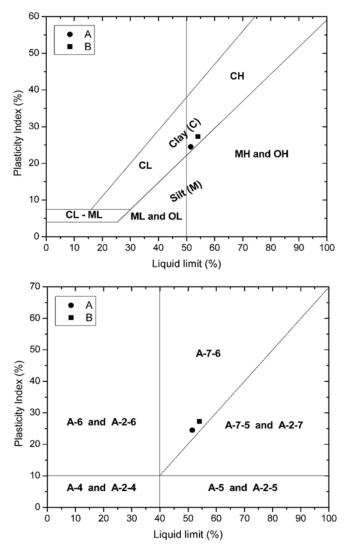


Fig. 1. Plasticity charts showing classification of the soil samples according to USCS (upper) and AASHTO (lower) system

The soil samples are predominantly fine-grained, having the percentages passing the BS No. 200 sieve (0.075 mm) to be greater than 50% for both samples A and B. The clay fraction of the soil samples will mostly influence their plasticity, deformation, strength, permeability and adsorption characteristics.

Table 5
Particle size classification of the soil samples

Soil A		Soil B				
Particle size [mm]	Passin	g [%]	Particle siz	ele size [mm] Passing [%		
Sieve analysis						
4.75	88	8	4.75	5	91.4	
2.36	8	1	2.36	5	87.7	
1.7	75	.8	1.7		84.8	
1.18	71	.5	1.18	}	80.4	
0.6	68	8	0.6		76.1	
0.5	66	.5	0.5		73.6	
0.425	65	.4	0.42	5	71.5	
0.212	63	.8	0.21	2	69.2	
0.15	62	.3	0.15	5	67	
0.075	59	.7	0.075 64.5		64.5	
	Hydrometer analysis					
0.0573	57.5		0.0573		62.2	
0.0416	53.2		0.0416		58.8	
0.0301	49.7		0.0301		56.4	
0.0194	45.3		0.0194		55.0	
0.0113	41	.8	0.0113		53.5	
0.0081	39	.4	0.0081		51.1	
0.0058	37	.0	0.0058		45.6	
0.0041	32	.1	0.0041		42.1	
0.0029	27	.6	0.0029		37.6	
0.0012	23	.2	0.001	2	34.9	
Composition, %		\$	Soil A		Soil B	
Gravel (2–60 mm)	el (2–60 mm)		22.0		15.0	
Sand (0.06-2.00 mi	n)		19.5 22.5		22.5	
Silt (0.002-0.060 m	nm)		33.0 27.0		27.0	
Clay (<0.002 mm)			25.5		35.5	

Calculations of the soil activity for soil samples A and B were found to be 0.98 and 0.78, respectively. These indicate that they are of average activity clays (between 0.75 and 1.25), neither inactive nor active. According to Unified Soil Classification System (USCS) (Fig. 1 upper) and American Association of State Highway and Transportation Officials (AASHTO) (Fig. 1 lower) soil classification systems, these soil samples are both classified as clay of high plasticity (CH) and A-7-6 with a group index of 14, respectively.

The results of standard Proctor and modified Proctor compaction tests on the soil samples are presented in Fig. 2. Expectedly, the optimum moisture contents (OMC) for the soil samples compacted using the standard Proctor compaction energy were higher than those obtained for the samples using the modified Proctor compaction energy but their maximum dry unit weights (MDUW) were lower. Generally, the OMC and MDUW of

sample A were found to be higher than those of sample B. The compaction characteristics suggest that adequate compaction of the soil samples can be achieved, when they are used as landfill liner materials.

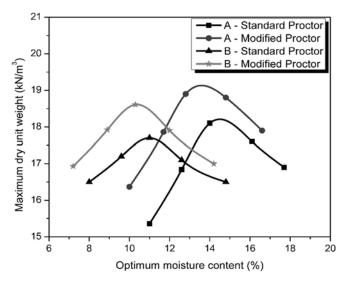


Fig. 2. Compaction characteristics of the soil samples

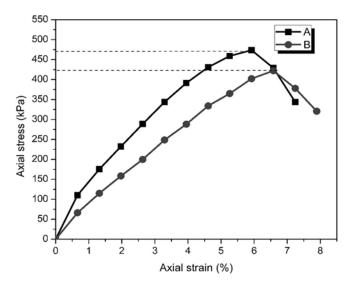


Fig. 3. Stress versus strain plot for the soil samples

The stress versus strain relationship exhibited by the soil samples is illustrated in Fig. 3. The unconfined compressive strength (UCS) of soil samples A and B were 470 kPa and

420 kPa, respectively. The coefficient of permeability from falling head test for sample A was 7.95×10^{-10} m/s (standard Proctor compaction) and 3.80×10^{-11} m/s (modified Proctor compaction). For sample B, the coefficient of permeability was 6.90×10^{-9} m/s (standard Proctor compaction) and 4.10×10^{-10} m/s (modified Proctor compaction). The coefficients of permeability were reduced as expected by an order of magnitude with a greater modified Proctor compaction energy (2703 kJ/m³), however the values obtained using the lower standard Proctor energy (596 kJ/m³) satisfied the upper acceptable limit requirement for bottom liners (1×10^{-9} m/s) [10].

 $\label{eq:Table 6} Table~6$ Initial and equilibrium concentrations of heavy metals in leachates after batch adsorption [mg/dm³]

[Dk]. : I 1	[Pb] _{eq}	in L1	[Dh], in I 2	[Pb] _{eq} in L2		[7n]. in I 1	[Zn] _{eq} in L1	
[Pb] ₀ in L1	Soil A	Soil B	[Pb] ₀ in L2	Soil A	Soil B	[Zn]₀ in L1	Soil A	Soil B
5014	96.5	190.5	4938	98	189.5	5045	188.5	294
4539	90	179	4496	93.5	179.5	4486	172.5	273.5
4033	83.5	145.5	3996	81	146	3980	153	256
3519	79.5	135	3502	77	136	3853	133.5	225
3006	64.5	109	2997	59.5	106.5	3034	103.5	204.5
2511	41	81	2506	39.5	79.5	2532	90.5	185
2002	24	63.5	2001	26	65	2013	75	152
1520	13	45	1469	15.5	45.5	1524	33.5	114.5
996	9	24.5	921	8	22.5	1051	17	73
508	3.5	12	496	4	18.5	548	7.5	51.5
[Zn] ₀ in L2	[Zn]eq	in L2	[Cr] ₀ in L1	[Cr] _{eq} in L1		[Crl. in I 2	[Cr] _{eq} in L2	
	Soil A	Soil B	[CI]0 III LI	$[Cr]_0 \text{ in L1} \qquad Soil A \qquad Soil B \qquad [Cr]_0 \text{ in L2}$	Soil A	Soil B		
4986	188	295.5	5002	136	156	4994	135	154
4537	175.5	272.5	4502	124	145.5	4500	125	145.5
4007	153.5	251.5	4002	115.5	125.5	3990	119.5	129
3731	133.5	218.5	3504	102.5	117	3485	101.5	120
3000	106.5	193.5	2996	86.5	112.5	2990	87	111
2495			2502	70	92	2493	69.5	94.5
2773	91	178	2502	70	92	2173	07.0	,
2009	91 72	178 151.5	2502	55	78	1990	59	75.5
				, ,				
2009	72	151.5	2001	55	78	1990	59	75.5

From the batch adsorption test, the results of the equilibrium concentration of each of the considered heavy metals in the leachate are presented in Table 6.

The plots of the results of batch equilibrium adsorption test showing the capacities of the two lateritic soil samples to adsorb heavy metals from the leachate 1 are shown in Figs. 4 and 5, respectively, and from leachate 2 in Figs. 6 and 7. All the adsorption isotherms exhibit a positive linear trend throughout for the cations.

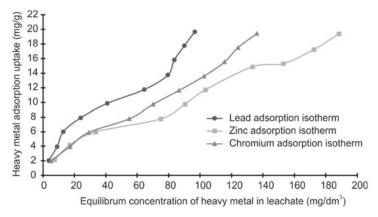


Fig. 4. Adsorption isotherm for soil sample A in leachate 1

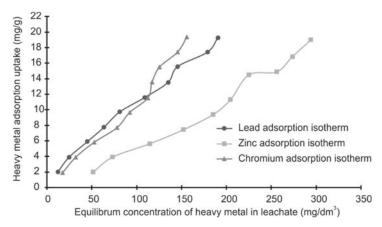


Fig. 5. Adsorption isotherm for soil sample B in leachate 1

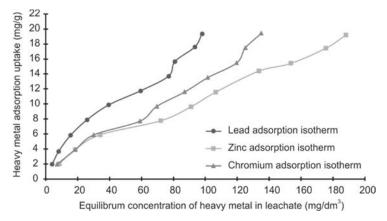


Fig. 6. Adsorption isotherm for soil sample A in leachate 2

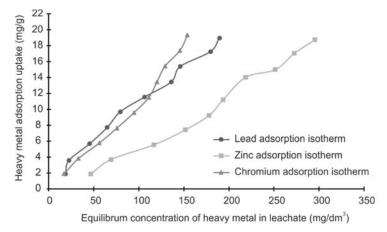


Fig. 7. Adsorption isotherm for soil sample B in leachate 2

Figures 4–7 show that the adsorption of the heavy metals from leachates by the soil samples was similar in character for each soil sample even in various leachates. This further proves the homogeneity of the soil samples that were collected, as even the partition coefficient is similar for each soil even in both leachates as shown in Table 7.

 $$\operatorname{\texttt{Table}}$\ 7$$ Retardation factors for heavy metals in tested soil samples

Heavy for soil sample in leachate	$K_p [dm^3/g]$	P_d [g/dm ³]	n_e	R_d
Lead, soil A in L1 Lead, soil A in L2	0.16			1.76
Zinc, soil A in L1 Zinc, soil A in L2	0.09	1.9476	0.41	1.43
Chromium, soil A in L1 Chromium, soil A in L2	0.13			1.62
Lead, soil B in L1 Lead, soil B in L2	0.09			1.36
Zinc, soil B in L1 Zinc, soil B in L2	0.07	1.8559	0.46	1.28
Chromium, soil B in L1 Chromium, soil B in L2	0.12			1.48

Unusual shapes of the adsorption isotherms were found to be due to implicit combination of type II isotherm behavior (this type of isotherm indicates an indefinite multi-layer formation after completion of the monolayer and is found in adsorbents with a wide distribution of pore sizes), increase in the equilibrium concentration of heavy metals in the leachate and possibly heat effects (from shaking in the rotary tumbler).

However the research has shown that many adsorbents and adsorbates exhibit more complicated isotherm shapes due to pore-filling, complexation or binding anomalies [11].

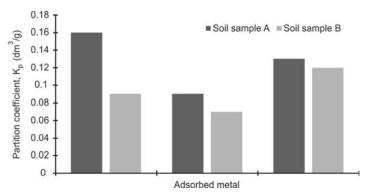


Fig. 8. Partition coefficients for the metal uptake by the soil samples

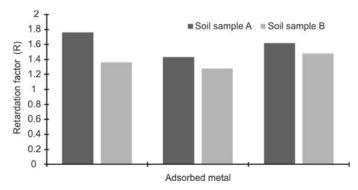


Fig. 9. Retardation factors for the metal uptake by the soil samples

The partition coefficients and retardation factors for each of the metals and each of the soil samples as shown in Table 7 are in Figs. 8 and 9. Since each tested soil showed the same partition coefficient and retardation factor for each of the metals in different leachates, the graph was limited to metal adsorption of soil samples in any one of the two leachates, to avoid repetition. The figures show that K_p and R_d for the soil sample B are generally lower than those for soil sample A.

The proportions of fine-grained particles in soils play a vital role towards their adsorptive behavior. Consequently, the plasticity product of soil samples can be used to correlate the effective contribution of the plasticity of the fine-grained particles of the soils to their adsorptive behavior. The plasticity products for samples A and B were calculated and found to be 1592.5 and 1583.4, respectively. This may explain the reason why the amount of the metals adsorbed on soil sample B were found to be lower than those adsorbed on soil sample A.

The K_p and R_d coefficients of the lateritic clays, which were both classified as A-7-6 with a group index of 14 and CH, decreased upon decreasing the plasticity product. However, the sorption selectivity order for both soil samples was slightly different from each other. For soil sample A the sorption selective order was as follows: $Zn^{2+} < Cr^{2+} < Pb^{2+}$ while for soil sample B it was: $Zn^{2+} < Pb^{2+} < Cr^{2+}$.

The results of the soil classification tests, from which the soil samples were classified as clay of high plasticity (CH), indicate that the soil samples have the potential of being used as hydraulic barriers for waste containment [12].

The maximum dry unit weights of the soil samples using both the standard and modified Proctor compaction tests were found to be high. These, along with the unconfined compressive strength (UCS) of the soil samples, suggest that the samples have appreciable strength and resistance to damage.

The coefficients of permeability for both tested soil samples were found to be lower than 1×10^{-9} m/s, which is widely recommended for a soil that is to be used as a landfill liner [10, 13–19].

4. CONCLUSIONS

The geotechnical properties of two lateritic clay soils have been evaluated for potential use as landfill liners capable of adsorbing metal contaminants from municipal solid waste leachates. The soil samples, which were of high plasticity, were both classified as A-7-6 and CH, according to American Association of State Highway and Transportation Officials (AASHTO) and Unified Soil Classification System (USCS). The unconfined compressive strengths (UCS) of tested soils which were in the range of 420–470 kPa, were found to be sufficient to resist damage. Based both on the standard and modified Proctor compaction tests, the coefficients of permeability for the tested soils were lower than the 1×10^{-9} m/s that is widely recommended for soils to be used as landfill liners. The amounts of heavy metals (lead, zinc and chromium) adsorbed on tested soils depended on the plasticity of the fine-grained of the tested soils. The sorption selectivity order for both tested soils depended on the soil type and properties.

The lateritic clay soils under examination and soils with similar geotechnical properties can be suitably used as landfill liners. They have the potential of preventing the migration of contaminants in landfill leachates and consequently protecting underground water.

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