



CEMENT STABILIZATION TREATMENT OF LEAD AND NAPHTHALENE CONTAMINATED LATERITIC SOILS

Opeyemi E. OLUWATUYI^{1*}, Edwin C. ASHAKA², Oluwapelumi O. OJURI²

¹Department of Civil Engineering, College of Engineering, Landmark University, Omu-Aran, Nigeria

²Department of Civil and Environmental Engineering, School of Engineering and Engineering Technology, Federal University of Technology, Akure, Nigeria

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Abstract. This article presents an investigation on the influence of Ordinary Portland Cement (OPC) as a binder in the stabilization treatment of lateritic soil contaminated with lead or naphthalene. To evaluate the performance of the binder, the contaminated soils were tested for mechanical strength and environmental performance before and after the stabilization treatment. Results showed that the strength as inferred from the unconfined compressive strength (UCS) and cohesion values increased with the addition of the binder. Cement stabilization of the lead contaminated samples also prompted a reduction in the release of lead below the admissible limit during the leaching test. Cement stabilization of the naphthalene contaminated samples, on the other hand, could not contain the release of naphthalene below the regulatory level during the leaching test. The batch equilibrium adsorption test (BEAT) showed that cement stabilization increased the adsorption capacity of the soil for the contaminants.

Keywords: batch adsorption, cement, compressive strength, leaching, soil contamination, stabilization.

Introduction

There is growing concern that Nigerian soils are being unabatedly degraded with diverse known and emerging organic and/or inorganic contaminants/chemicals of concern (COCs) from poor environmental management of mining activities, industrial development and weak or non-existent legal framework. Inorganics (heavy metals e.g. lead) and certain types of organics (e.g. naphthalene) are extremely insistent in soils typically causing austere insalubrity to people and receptors (F. Wang, H. Wang, Jin, & Al-Tabbaa, 2015). Due to its fixed and non-breakable structure, lead present possible risks because of its bioavailability (Ragnvaldsson, Berglind, Tysklind, & Lefler, 2007). Most soils especially fine-grained soils easily absorb lead onto themselves (Ojuri, Akinwumi, & Oluwatuyi, 2017), this attribute heightens with an increase in the soil's organic content, CEC (cation exchange capacity) and pH (Zhang et al., 2015). On the other hand, polycyclic aromatic hydrocarbons (PAHs) are a group of organic compounds that consist of two or more fused benzene ring structures in various arrangements (Environment Agency, 2003). From the United States Environmental Protection Agency [USEPA] (2009) list of 16

priority PAHs contaminants (Bojes & Pope, 2007; Wang et al., 2007), naphthalene (C₁₀H₈) is deemed the simplest partly because of its low molecular weight (Yan & Lo, 2013). At room temperature, naphthalene is a white crystalline solid with a strong tar-like odour (Defra & Environment Agency, 2003). It has been extensively used in the study of PAH elimination/reduction in soil (Liang & Guo, 2010; Oluwatuyi & Ojuri, 2017).

In Nigeria, the rate at which these two contaminants are found in the soil is gradually becoming alarming. Lead mainly through lead batteries and other anthropogenic activities, naphthalene through mothballs (also known as camphor) and other hydrocarbon sources. The concentrations of heavy metals like lead on Nigerian highways need to be curtailed before they start affecting the human health and environment (Ojuri, Taiwo, & Oluwatuyi, 2016). Research on the hydrocarbon contaminated site of Baruwa community in Lagos State, Nigeria showed that more than 200 wells were contaminated with floating petroleum product (including naphthalene). Groundwater was about 25 m below ground surface in the wells of between 0.75 m and 2.00 m in diameter. Remediation using free hydrocarbon recovery, in-situ chemical oxidation and bioremediation started in 2014 with a pilot scheme

*Corresponding author. E-mail: oluwatuyi.opeyemi@lmu.edu.ng

area (100×100 m), 10 existing wells were within the pilot scheme area (Ojuri, Ola, Fadugba, & Uduebor, 2014; Uduebor & Ola, 2016).

Geo-environmental engineers have a huge responsibility during the early stages of the construction process, particularly “where potentially adverse impacts are highest” (Holt, Jefferson, Braithwaite, & Chapman, 2010). Contaminated soils in their untreated state usually lack strength and firmness, making them unfit for construction purposes and a potential source of danger to the environment. When such contaminated soil is encountered, the Engineer has the options of leaving the site for a suitable one, replacing the soil with an appropriate one or improving the properties of the soil through stabilization. Stabilization is different from other forms of remediation treatments in that instead of destroying the contaminant, their movement in the soil is limited through the chemical reaction between the soil and binder thereby minimizing their discharge to the environment (Siebielec & Chaney, 2012). Lime (Mckinley, Thomas, Williams, & Reid, 2001; Wang, Abriak, Zentar, & Xu, 2012) and cement (Du, Jiang, Liu, Jin, Singh, & Puppala, 2013; Akinwumi, Booth, Diwa, & Mills, 2016) are the most common forms of binder used in stabilization treatment. They could be used alone or used along with other pozzolans like rice husk ash (Yin, Mahmud, & Shaaban, 2006; Oluwatuyi & Ojuri, 2017), fly ash (Kogbara, Al-Tabbaa, Yi, & Stegemann, 2013; Wang, Abriak, & Zentar, 2013; Falciglia, Al-Tabbaa, & Vagliasindi, 2014), pulverized fuel ash (Tang, Yan, Lo, & Liu, 2015), milled eggshell (Oluwatuyi et al., 2018), sawdust ash (Ojuri & Oluwatuyi, 2018) etc.

This scientific article examines the outcome of using cement as a binder on the mechanical strength and environmental performance of lateritic soils contaminated with lead and naphthalene. The strength of cement stabilized lead and naphthalene contaminated lateritic soils was compared to that of lime-rice husk ash stabilized (a previous study). The suitability of the cement stabilized lead and naphthalene contaminated lateritic soil as a possible construction material is also determined.

1. Materials and methods

1.1. Sample collection, preparation and treatment

The lateritic soil used in this work was sampled from the same geographical area as those used by Ojuri and Oluwatuyi (2014). The sampling location was within Latitude 7° 10'N and 7° 20'N and Longitude 5° 07'E and 5° 17'E inside the campus of Federal University of Technology, Akure (FUTA), Nigeria. A 10 kg bulk relatively undisturbed sample of soil was collected at a trial pit depth of about 1.0 m and transferred to the laboratory for further treatment. Treatment of the soil samples which include pulverization and homogenization, artificial contamination with lead and naphthalene and subsequent binder addition was all done in a similar procedure and quantity as those of Oluwatuyi and Ojuri (2017).

1.2. Testing protocol

Basic geotechnical tests which include particle size distribution, specific gravity, natural moisture content, and Atterberg limits were conducted on the uncontaminated soil sample. Strength tests which include unconfined compressive strength (UCS) and direct shear (DS) tests were conducted on the relatively undisturbed samples of the soil, 2000 ppm lead and 3% naphthalene contaminated soil (which are the maximum quantity for each contaminant) and cement stabilized contaminated soil. The basic geotechnical tests and strength tests were conducted in accordance to the steps configured in British Standards Institution, BSI (1990). The chemical (elemental) composition of the soil and cement was determined using S1 TITAN Handheld X-ray Fluorescence Spectrometer, produced by Bruker Corporation. Environmental performance tests which include leaching and batch equilibrium adsorption tests were performed on the contaminated soil and cement stabilized soil samples.

The procedures for the leaching were the same as the one used by Oluwatuyi and Ojuri (2017), the exception was that all soil samples were prepared at natural pH only and tumbled at different contact times of 30, 60, and 90 days. The contact time is the period of contact between the contaminated soil and the liquid (termed “leachant”). The contaminated soil and the liquid are mechanically mixed during this period. At the completion of this period, mass (contaminant) transfer from the contaminated soil to the liquid is measured by filtering the liquid (termed “leachate”) from the liquid-contaminated soil mix and determining its contaminant concentration. The contact time depict a significant factor that affects the discharge of contaminants during leaching test. It may affect the amount of leached contaminant, except equilibrium terms have been fixed (Galvín, Ayuso, Jiménez, & Agrela, 2012). The inorganic contaminant content was analyzed using the iCE 3400 AAS atomic absorption spectrophotometer produced by Thermo Fisher Scientific. The naphthalene contaminant content in the leachate was analyzed using the gas chromatography and electrical conductivity meter (indirect approach). Batch equilibrium adsorption tests (BEATs) were also performed as described in Oluwatuyi and Ojuri (2017).

2. Results and discussion

2.1. Material characterization and soil geotechnical properties

The percentage composition of chemical oxides in the soil and cement samples used in this study is presented in Figure 1. A ternary plot of the oxides of silicon (SiO_2), iron (Fe_2O_3) and aluminium (Al_2O_3) existent in the soil sample is presented in Figure 2. The degree of laterization in the soil sample, calculated from the ratio of silica to sesquioxides ($\text{SiO}_2 / (\text{Fe}_2\text{O}_3 + \text{Al}_2\text{O}_3)$) was 0.02, an indication the soil sample was laterite. The natural soil sample was

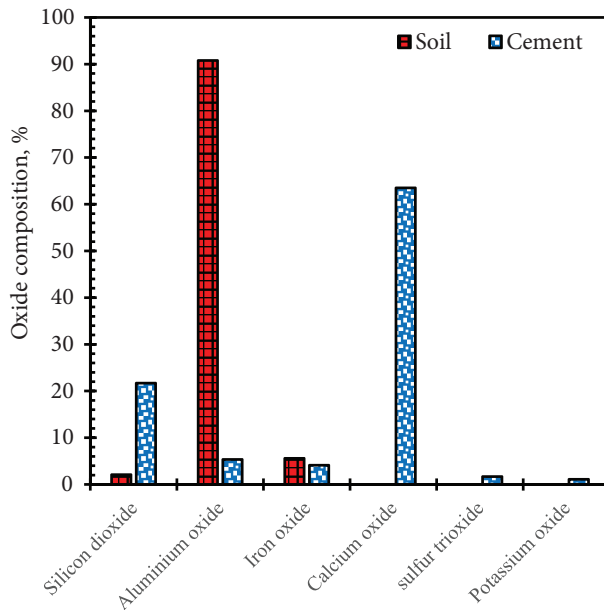


Figure 1. Chemical composition of soil and cement

plotted in the strong laterization profile (Figure 2), according to the classification of weathering products.

The summary of the basic geotechnical tests conducted on the natural uncontaminated soil and the (lead and naphthalene) contaminated soil is presented in Table 1. The table shows that the Atterberg limit values all reduced with the addition of the contaminants to the soil. The natural soil sample was classified as clay of high plasticity (CH) while the lead and naphthalene contaminated samples were both classified as clayey sand (SC) according to the USCS classification. The percentage of particles passing through a 75 μm sieve (sieve No. 200) was approximately 52%, 40% and 36% for the natural soil, lead and naphthalene contaminated samples respectively. There was a noticeable change in the grain size distribution of the natural soil compared to the contaminated soil (Table 1). The change (decrease) in the percentage gravel was due to the pulverization of the soil sample before artificial contamination. On the other hand, change (decrease) in the percentage fines (silt and clay) may be due to the bonding of the fine sizes with each other to form pseudo-sand sizes and with other sand sizes to form larger sand or clog sizes. The natural moisture content of soil sample was 25.5%. After contamination, samples were rehydrated by adding deionized water to contaminated samples to yield a target water content of approximately 25% before the binder addition.

2.2. Strength of cement stabilized contaminated soil

Figure 3 showed that the UCS values increased as the cement content used in stabilizing the contaminated soil increased. The UCS values of the lead and naphthalene contaminated soil were increased by 562 kN/m² (122.2%) and 795 kN/m² (159%) respectively after stabilization with 10% cement. These UCS values were higher than those

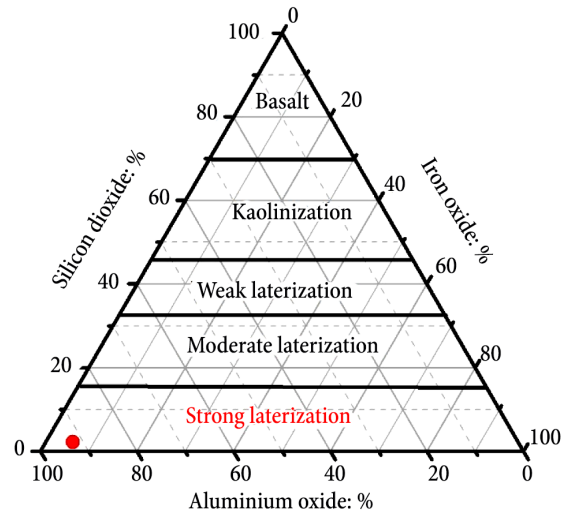


Figure 2. Ternary plot of natural soil sample

Table 1. Geotechnical properties of the uncontaminated and contaminated soil samples

Engineering & Physical Properties	Natural soil	2000 ppm of lead contaminated soil	3% (by dry weight of the soil) naphthalene contaminated soil
Physical			
pH	6.6	5.9	6.4
Specific gravity	2.72	2.71	2.72
Liquid limit (%)	52.3	37.6	32.8
Plastic limit (%)	24.5	20.2	19.5
Plasticity index (%)	27.8	17.4	13.3
Linear shrinkage (%)	11.5	10.4	10.1
Bulk density (g/m ³)	1.64	1.69	1.70
Organic matter (g/kg)	6.2	5.6	5.3
Strength			
UCS (kN/m ²)	223	460	500
Cohesion (kN/m ²)	90	170	120
Soil Composition summary			
% Gravel (2–60 mm)	12.04	9.68	9.03
% Sand (0.06–2.00 mm)	36.16	50.45	54.62
% Silt (0.002–0.060 mm)	23.39	19.52	18.46
% Clay (<0.002 mm)	28.41	20.35	17.89
Soil Classification			
AASHTO Classification	A-7-6	A-6	A-6
USCS Classification	CH (clay with high plasticity)	SC (clayey sand)	SC (clayey sand)

of Crane, Cassidy, and Srivastava (2014) who used a 5% cement content and those of the previous study by Ojuri and Oluwatuyi (2014). Normally the contaminants used are known to cause deleterious effects on the UCS of the soil (Trussell & Spence, 1994). Contamination of the soil with lead and naphthalene (which were in granular form) increased the density and inter-particle cohesion of the soil leading to a rise in the UCS value of the soil. Contamination of high plastic clay soil with organic compounds or electrolyte solutions having a high salt concentration is expected to produce an improvement of mechanical behaviour and not a detriment.

The UCS samples though not cured had values that surpassed the regulatory waste disposal limit of 340 kN/m²

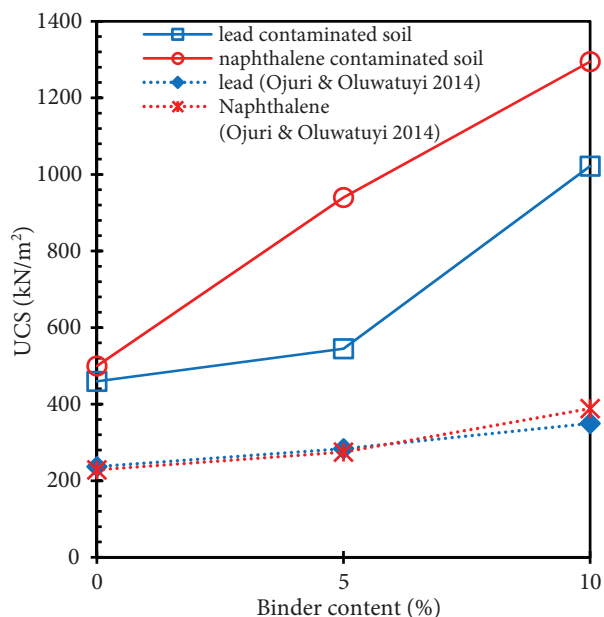


Figure 3. Variation of UCS with binder content

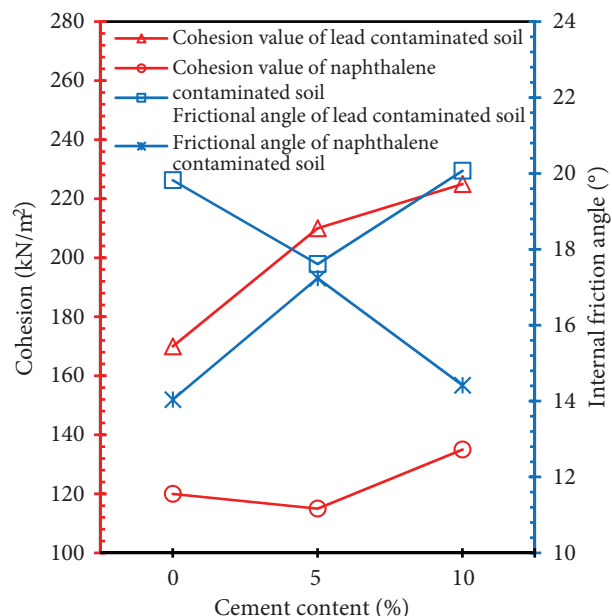


Figure 4. Variation of DS with binder content

in the United Kingdom (Sollars & Perry, 1989). The values were also greater than the UCS design values of 345 kN/m² commonly required for in-situ stabilization (USEPA, 2009; Interstate Technology and Regulatory Council [ITRC], 2011), 350 kN/m² applied in United Kingdom (Wheeler, 1995), and 440 kN/m² used in Canada (Stegemann & Cote, 1996). The cohesion values also had the same trend as the UCS values. Figure 4 displays the cohesion (forces that cement particles of soil) and internal friction angle (shear strength of soil due to friction) values acquired from the DS test conducted on the lead and naphthalene stabilized contaminated soil with cement content. The cohesion values of the lead and naphthalene contaminated soil were increased by 55 kN/m² (32.4%) and 15 kN/m² (12.5%) respectively after stabilization with 10% cement.

2.3. Leaching properties of cement stabilized contaminated soil

The main goal of stabilization treatment is to decrease drifting of the contaminant at a proportion of no consequential adverse effects to human and environment. Leaching test is a way of evaluating this goal, the stabilization treatment must be designed so that environmental (water & soil) quality is unaffected by contaminant concentration that surpasses an admissible limit (ITRC, 2011). The contaminant concentration in groundwater at a specific period must remain below the established standard (Antemir, Hills, Carey, Magnié, & Polettini, 2010). Ameliorating the strength of the contaminated soil is not enough, there is a need to also evaluate the environmental performance of the stabilized contaminated soil. Leaching is an environmental performance test and one of the two predominant criteria (the other is UCS) used in assessing the effectiveness of stabilization treatment in the United States (USEPA, 2000). The results of the leaching procedure conducted on the lead and

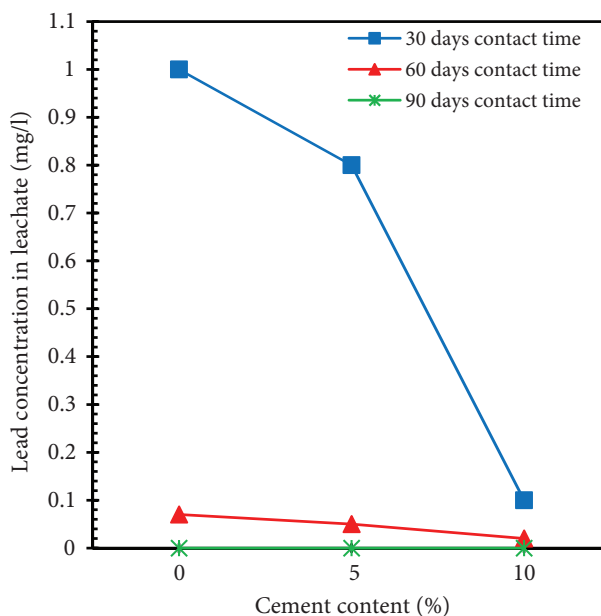


Figure 5. Leachate lead concentration

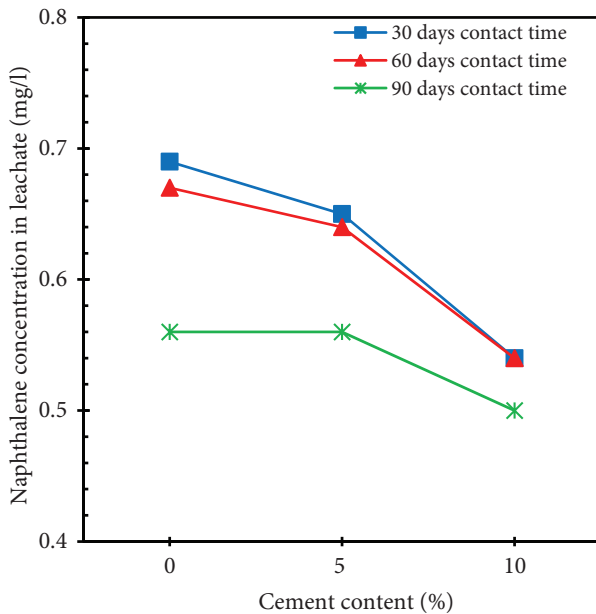


Figure 6. Leachate naphthalene concentration

naphthalene contaminated soil at different contact times are presented in Figures 5 and 6 respectively. The contact time or leaching period is the period of contact between the soil sample and the liquid leachant (water) during the leaching test. The figures showed that both contact time and cement content had an effect on the leaching properties of the lead contaminated soil. The concentration of lead and naphthalene decreased as cement content increased and also as the contact time lengthened. After 90 days of contact time, no lead was detected in the leachate. The results for lead contaminated soils were in the same range as those reported by Du et al. (2014) who used a lesser contact time. The results for naphthalene contaminated soils were in the same range as those reported by Hebatpuria, Arafat, Bishop, and Pinto (1999).

The decrease in the contaminant concentration can be attributed to the cement stabilization treatment which solidifies the soil matrix ultimately decreasing the mobility, solubility and toxicity of the contaminants. The decrease in concentration with increase in contact time could also be attributed to the contact time acting as a curing period for the contaminated samples treated with cement, these result in samples with more stable linkages leading to more decrease in the mobility of the contaminants. In this study, the lead concentration in the leachates from the cement stabilized lead contaminated soil were all well below 5 mg/l, which is the standard admissible limit for lead in the soil (USEPA, 1998). For the naphthalene contaminated soils, the naphthalene concentration in the leachates from cement stabilized naphthalene contaminated soil were all above 0.2 µg/l, which is the minimum reporting level for most volatile organic compounds including naphthalene in groundwater (Squillace, Moran, Lapham, Price, & Clawges, 1999).

2.4. Adsorption properties of cement stabilized contaminated soil

From the batch adsorption test, the results of the equilibrium concentration of lead and naphthalene contaminant in soil samples at varied cement content are presented in Figures 7 and 8 respectively. All the adsorption isotherms showed a positive linear drift for lead ions and naphthalene. The noticeable drifts connote that for lead ions, adsorption was more efficient at greater leachate concentrations and that a little more of lead ions was absorbed out of the solution as the cement content was increased. For naphthalene, the drifts imply more of naphthalene were absorbed out of the solution as the cement content was

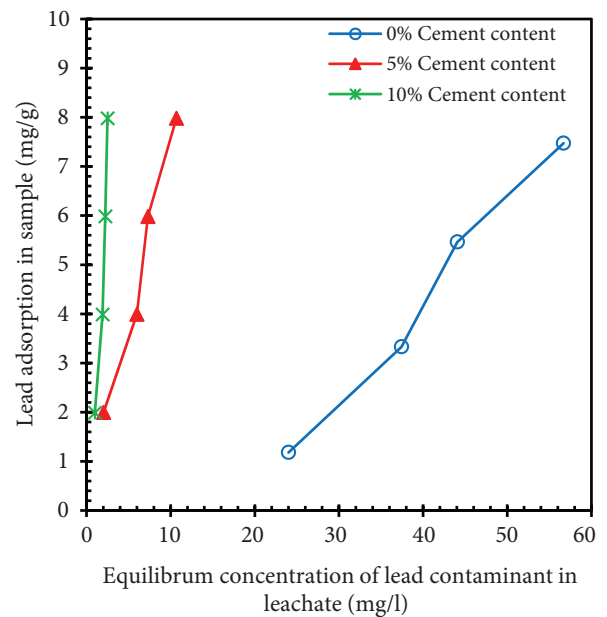


Figure 7. Adsorption isotherms for lead ions

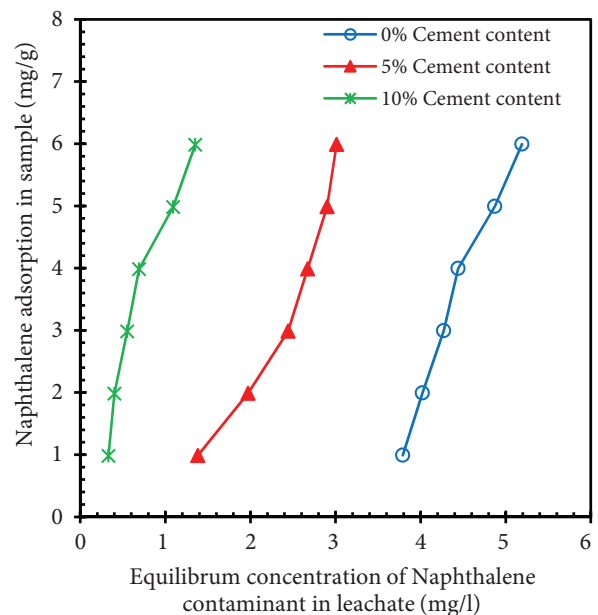


Figure 8. Adsorption isotherms for naphthalene

Table 2. Partition coefficient and retardation factors for contaminants in soil samples

Soil Sample Binder Addition level	Effective porosity (n_e)	Bulk Density (P_d) g/l	Partition Coefficient (K_p) l/g		Retardation factor (R_d)	
			Lead	Naphthalene	Lead	Naphthalene
0%	0.38	1.64	0.20	3.53	1.86	16.23
5%	0.34	1.76	0.71	2.90	4.68	16.01
10%	0.29	1.90	3.74	4.48	25.50	30.35

increased. The unusual shapes of the adsorption isotherms found in Figures 7 and 8 may be due pore-filling, complexation or binding anomalies (Park & Knaebel, 1992).

The values of the partition coefficient (K_p) and retardation factor (R_d) for each of the contaminants in the soil samples were presented in Table 2. The retardation factor for the lead in the sample not stabilized was the lowest followed by the 5% and 10% cement stabilized contaminated sample. This means that lead had the highest mobility (associated to the lowest adsorption) in the contaminated sample not stabilized. The mobility of lead subsequently decreased as the cement content in the sample was increased. The values of the retardation factor for lead in the soil samples were in the same range as those obtained by De Matos, Fontes, Da Costa, and Martinez (2001). The retardation factor for naphthalene in the soil samples was higher than those obtained by Kan and Tomson (1990) which implies a higher adsorption capacity of soil samples for naphthalene. Although unlike the lead contaminant the 5% cement stabilized contaminated sample had the lowest retardation factor for naphthalene followed by contaminated sample not stabilized and 10% cement stabilized contaminated sample. This might be due to binding anomalies from the 5% cement content on the naphthalene contaminated soil.

Conclusions

The scientific article examined the effect of cement as a stabilizing binder on the strength, leaching and adsorption properties of lead and naphthalene contaminated soils. The following conclusions were drawn.

The contamination of soil with lead and naphthalene (both in granular form) reduced the plasticity and in turn increase its strength. Liquid limit reduced from 52.3 for natural soil to 37.6 and 32.8 for lead and naphthalene contaminated samples, respectively. The UCS value of natural soil at 223 kN/m² increased to 460 and 500 kN/m² for lead and naphthalene contaminated samples, respectively.

Treatment of the contaminated soil with Portland cement shows further improvement in the strength (mechanical properties of the soil) with both the UCS and cohesion increasing with an increase in the cement content. The UCS values of the lead and naphthalene contaminated soil were 460 and 500 kN/m², by increasing the cement content from 0 to 10%, UCS values of stabilized samples increased to 1022 kN/m² and 1295 kN/m² for lead and naphthalene contaminated samples, respectively.

The leaching test on the samples showed that the cement stabilization was effective for the lead contaminated samples as their leachability was reduced beneath the standard admissible limit (5 mg/l) for lead in soil. The lead concentration in leachate after 30 days of contact time was 1 mg/l, by increasing the cement content from 0 to 10%, it reduced to 0.1 mg/l.

The leachability for naphthalene contaminated soil also decreased with cement stabilization but it was not below the minimum reporting level (0.2 µg/l) for naphthalene in the soil's groundwater. The naphthalene concentration in leachate after 30 days of contact time was 0.69 mg/l, by increasing the cement content from 0 to 10%, it reduced to 0.54 mg/l.

The retardation factors from the batch equilibrium adsorption test also showed that cement stabilized soil had a good adsorption capacity for both lead and naphthalene contaminants. For lead, by increasing the cement content from 0 to 10%, the retardation factors increased from 1.86 to 25.50. For naphthalene, by increasing the cement content from 0 to 10%, the retardation factors increased from 16.23 to 30.35.

Finally, Portland cement is recommended for improving the mechanical strength and environmental suitability of lead contaminated soils.

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