

Cement Stabilisation of Crude Oil-Contaminated Soil

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Abstract

Crude oil-contaminated soils are usually considered as unsuitable construction materials for earthworks. This study presents an experimental investigation of the effects of applying Portland cement on the plasticity, strength and permeability of a crude oil-contaminated soil, in order to ascertain its suitability for use as an earthwork construction material. A series of specific gravity, Atterberg limits, compaction, strength and permeability characteristics were determined for the natural soil, the soil after being artificially-contaminated with crude oil and for varying proportions of cement to the contaminated soil. The results indicate that the geotechnical properties of the soil became less desirable by its contamination with crude oil; whereas, the application of cement to the contaminated soil improved its properties by way of cation exchange, agglomeration and cementation. The cement stabilisation of crude oil-contaminated soil provides a stable supporting structure, as well as a capping layer, that prevents the crude oil from interacting with the construction materials above. Instead of disposing of such a contaminated soil, causing unnecessary waste and incurring costs, its stabilisation with cement, which is practically possible to undertake on site, makes it useful for supporting a structural foundation or a road pavement structure.

Keywords chosen from ICE Publishing list

Foundation; Pollution; Sustainability

1. Introduction

Crude oil spills resulting from the production, transportation and storage of crude oil have become common incidence in and around oil-producing countries. If the predictions made many years ago that oil resources would be entirely depleted within the first decade of the twenty-first century (Peden, 2003) are anything to go by, crude oil spills should not be a concern today. Crude oil contaminated soils are widely reported and prominent worldwide examples include: USA (Leewis *et al.*, 2013; Horel *et al.*, 2014), Canada (Akbari and Ghoshal, 2014), Argentina (Rios *et al.*, 2014), Italy (Giampaolo *et al.*, 2014), Romania (Marinescu *et al.*, 2014), China (Fu *et al.*, 2014; Wang *et al.*, 2013), Malaysia (Hamzah *et al.*, 2014), India (Yenn *et al.*, 2014), Iran (Khamehchiyan *et al.*, 2007; Rad *et al.*, 2014) and Kuwait (Al-Mailem *et al.*, 2014). The problem is most notable in the Niger Delta region, Nigeria (Onojke and Frank, 2013), where an estimated average of 240,000 barrels of crude oil is spilled every year (Ordinoha and Brisibe, 2013) and is listed among the top ten toxic threats (Blacksmith Institute, 2015).

Many research efforts have been carried out towards finding means of reclaiming or remediating petroleum oil-contaminated soil. Mukherjee and Bordoloi (2010) investigated the use of exogenously seeded bacterial consortium as a bioremediation technique. Yateem (2013) focussed on the rhizoremediation of oil-contaminated sites. Kavitha *et al.* (2014) studied the removal of crude oil contamination from soil using *Bacillus licheniformis* MTCC 5514. However, the majority of research efforts have centred on remediating the soil in order to make them suitable for agricultural purposes. Few research works have been carried out to improve the engineering properties of oil-contaminated soils for use as construction materials or as an earth foundation.

Shah *et al.* (2003), Khamehchiyan *et al.* (2007), Kermani and Ebadi (2012) and Akinwumi *et al.* (2014) have reported that contamination of soils with petroleum oil alter their geomechanical behaviour. Khamehchiyan *et al.* (2007) investigated the effect of crude oil contamination on the Atterberg limits, compaction characteristics, uniaxial compressive strength and permeability of clayey and sandy soils. They reported that the liquid and plastic limits, maximum dry unit weight, optimum moisture content (OMC), uniaxial compressive strength and permeability reduced with increasing crude oil content. Kermani and Ebadi (2012) and Akinwumi *et al.* (2014) report that crude oil increases the liquid and plastic limits of contaminated soil samples. Akinwumi *et al.* (2014) also state that crude oil reduces the maximum dry unit weight, optimum moisture content and permeability of a clayey soil. However, research works with the aim of making crude oil-contaminated soils become suitable for use as construction materials are rare, except Ochepo *et al.* (2013) who investigated the effect of spent (lubrication) oil contamination on the strength of lime and cement stabilized soil.

The role played by geotechnical engineers during the early stages of the process of construction cannot be overemphasized, especially where 'where potentially adverse impacts

are highest' (Holt *et al.*, 2010). Soil stabilisation is a common method adopted by geotechnical engineers to improve the engineering properties of weak or problematic soils in order to make them suitable for construction purposes. Portland cement is the most common and widely used soil stabilizer (Al-Aghbari *et al.*, 2009; Jegandan *et al.*, 2010; Ciancio and Boulter, 2012).

Previous studies have not investigated the effect of stabilizing a crude oil-contaminated soil applied with Portland cement. Therefore, the purpose of this study is to examine the effects of ordinary Portland cement on the plasticity, strength and permeability of a clayey soil contaminated with crude oil, so as to appraise their use for infrastructure development by Foundation and Highway Design Engineers.

2. Materials and methods

2.1 Sample collection

Soils used in this study were obtained from a borrow pit, having a deep lateritic soil profile, at Agbara (latitude 06°30'36"N and longitude 03°05'33"E), Ogun State, Nigeria, which is typical of soils in tropical climates. It was collected from the sidewall of the soil profile at ~10m depth. Previous borrowing activities for various nearby construction projects exposed the deep soil profile at this borrow pit, making it easy to collect the soil sample from the sidewall using a digger and shovel. Bulk samples were stored in labelled and sealed sacks and transferred to the civil engineering laboratory at Covenant University, while field sub-samples were stored in watertight sealed containers for subsequent laboratory determination of their natural moisture content using laboratory oven-drying method.

The crude oil sample was collected from a station operated by the Nigerian Petroleum Development Company (NPDC), the upstream arm of the Nigerian National Petroleum Corporation (NNPC) at Ologbo, Edo State, Niger Delta area of Nigeria.

2.2 Laboratory techniques

Bulk sample soils were air-dried, disaggregated, sieved (<4.75 mm) and thoroughly mixed (AASHTO T 99-10 2010). The contamination of the soil with crude oil, by dry weight of the soil, was done artificially in the laboratory. A brand of ordinary Portland cement (ASTM Type I cement of grade 42.5) available in the Nigerian market was procured and used in this study.

The geochemical characterization of the soil was determined using atomic absorption spectrophotometer (Pelkin Elmer 3300) while the chemical composition of the cement was determined using X-ray fluorescence analyzer (Thermo Fisher ARL, 9900).

The specific gravity and American Petroleum Institute (API) gravity of the crude oil sample at 15.55°C (60°F) were determined in accordance with ASTM D1298 (2012). The specific gravity was determined by the hydrometer method while the API gravity was calculated using Equation 1:

$$1. \quad ^\circ\text{API} = \left(\frac{141.5}{\text{Specific gravity at } 60^\circ\text{F}} \right) - 131.5$$

2.3 Experimental design and sample treatments

Representative soil sample was selected by quartering and riffing. It was then oven-dried in the laboratory at 105 °C for 24 hours. Immediately after oven-drying, a 50-litre laboratory mixing drum was used to mix the crude oil and the soil. The contaminated soil was then stored in an air-tight plastic container for one month and allowed to equilibrate prior to commencement of laboratory tests (Khomehchiyan *et al.*, 2007). Sieve and hydrometer analyses were carried out on the uncontaminated soil sample in order to determine the particle size distribution of the soil. Specific gravity, Atterberg limits, compaction, unsoaked and soaked California bearing ratio (CBR), unconfined compressive strength (UCS) and permeability tests were conducted on the natural soil, contaminated (10% crude oil content, by dry weight of the soil) soil and on the cement-stabilized contaminated soil samples, respectively. The maximum percentage of crude oil used (10%) was selected based on inference from review of literatures (Osuji *et al.*, 2006; Osuji *et al.*, 2007; Osuji *et al.*, 2007; Okop and Ekpo, 2012; Tane and Albert, 2015) reporting crude oil contamination levels in soils of the Niger Delta region of Nigeria after crude oil spills, which show that contamination levels exceeding 50,000 mg of crude oil per kg of soil are rare though possible. Also, the maximum contamination level used was selected in order to avoid crude oil draining out of the soil sample during compaction (Khomehchiyan *et al.*, 2007). Various proportions of Portland cement (0, 2, 6 and 10%, by weight of the soil) were applied to the contaminated soil. These tests were performed in order to determine the effects of application of the cement on the geotechnical properties of the contaminated soil. The tests were carried out in triplicate and the mean value of each parameter and its standard deviation are presented.

The natural moisture content of the soil was determined using laboratory oven-drying method, which involved the drying of the natural soil sample in a laboratory oven at a temperature of 105 °C for 24 hours in order to determine its moisture content as a percentage of its dry mass (BSI, 1996 – BS 1377: Part 2: 1990, Clause 3.2). Sieve analysis was performed on the soil by wet sieving to remove its silt and clay-sized particles, oven-drying of the coarser fraction and dry sieving of this fraction using a set of sieves with diminishing apertures (BSI, 1996 – BS 1377: Part 2: 1990, Clause 9.2). Hydrometer analysis was conducted on the fine-grained fraction of the soil by the method of sedimentation using sodium hexametaphosphate as the dispersing agent (BSI, 1996 – BS 1377: Part 2: 1990, Clause 9.5). The plasticity of the samples was determined from liquid and plastic limits tests. The liquid limits of the samples were determined using the Casagrande apparatus method. The fraction of the soil samples passing through 425 µm sieve were mixed to four consistencies, including some wet and dry of the liquid limit; for each consistency, the number of blows that caused 13 mm of closure of the groove by the sample in the cup of the apparatus was recorded; by interpolation using graphical plot of

moisture content against number of blows, the liquid limit of the soil sample was determined as the soil moisture content corresponding to the groove closure by 25 blows (BSI, 1996 – BS 1377: Part 2: 1990, Clause 4.5). The plastic limits of the samples were determined as the moisture content of the remoulded and rolled samples that produced longitudinal and transverse cracks at a rolled diameter of 3 mm (BSI, 1996 – BS 1377: Part 2: 1990, Clause 5.3) while the plasticity indices were derived by determining the difference between the liquid and plastic limits of each sample (BSI, 1996 – BS 1377: Part 2: 1990, Clause 5.4). The specific gravities of the samples were calculated from determinations of the dry weight the samples and the weight of the same sample plus water in 50 ml density bottles (BSI, 1996 – BS 1377: Part 2: 1990, Clause 8.3). The procedure that was followed for the determination of the UCS using the load frame method is in alignment with that outlined in BSI (1990) – BS 1377: Part 7: 1990, Clause 7.2. The UCS test specimens (50 mm diameter x 100 mm length), prepared and extruded from a cylindrical mould, were cured in sealed plastic bags. The UCS was determined after 28 days of curing of each sample.

The modified proctor energy was used for preparing the specimen for compaction and CBR tests. The specimen diameter and height are 150 mm and 175 mm, respectively. While preparing the specimen, 4.5 kg rammer was used to provide 56 blows to each of 5 layers of compaction. Unsoaked CBR test specimens were cured for 7 days under controlled temperature ($25 \pm 2^\circ\text{C}$) and relative humidity (100%). For the determination of soaked CBR, specimens were cured for 5 days under the controlled temperature and the relative humidity before being immersed in water for 48 hours. The specimens were soaked for 48 hours in order to allow the highly plastic soil to get fully saturated instead of 24 hours soaking that is more suitable for low to moderate plasticity soils (Little, 2000), thereby simulating the likely moisture condition of soils under structural foundations and pavements in the Niger Delta area of Nigeria that sometimes experience long periods of rainfall. Falling head permeameter was used to determine the coefficient of permeability of the samples. The procedure for the determination of the permeability of the samples by falling head method is in accordance with Head (1994).

3. Results

3.1 Materials characterisation

From the results of the atomic absorption spectroscopy carried out on the soil sample, a ternary plot of the percentage composition of the oxides of silica (SiO_2), iron (Fe_2O_3) and aluminum (Al_2O_3) in the soil was obtained (Figure 1). This plot was produced in order to determine the level or extent of laterization of the soil used. According to the Schellmann (1986) scheme of classification of weathering products, this soil sample was taken from a kaolinized profile. The silica-sesquioxides ratio for this soil is 1.66, which indicates that the soil is lateritic. The specific gravity of the crude oil at 15.55°C is 0.841 and its API gravity at 15.55°C is 36.8 degree API. The water content of the crude oil was not determined but it was inferred from studies (Onajake *et al.*, 2013; Dickson and Udoessien, 2012; Odebunmi and Adeniyi, 2004; Odebunmi *et al.*,

2002) on the properties of crude oil from the Niger Delta region of Nigeria having similar density that the water content is equal to or less than 0.2% by volume of the crude oil, which should not make significant contribution to the moisture content of the oil-contaminated soil.

The chemical composition of the cement used, expressed in terms of oxides and calculated from elemental analysis determined by X-ray fluorescence, is presented in Figure 2.

From Figure 2, it can be seen that the cement is rich in CaO, while the soil is rich in SiO₂. The predominance of silica in the soil suggests that this soil was formed from laterite on an acidic rock and that it contains some quartz (Akinwumi, 2014).

3.2 Geotechnical properties of the natural soil and the crude-oil contaminated soil

The classification, index properties, permeability and strength of the natural soil and the crude oil-contaminated soil are summarily presented in Table 1. It shows that the natural and contaminated soil samples are classified as sandy clay and sandy organic silt, respectively. The liquid and plastic limits and plasticity index of the contaminated soil were found to be higher than those of the natural soil. On the other hand, the optimum moisture content (OMC), maximum dry unit weight, coefficient of permeability and soaked CBR of the natural soil are higher than those of the contaminated soil. The unsoaked CBR of the contaminated soil is higher than that of the natural soil. The natural soil is brown and has about 55% of its particles passing through the 75 µm sieve (Figure 3).

3.3 Effects of stabilisation of the crude-oil contaminated soil with cement

Consistency (Atterberg) limits of a soil helps to characterize its state based on its water content. Figure 4 shows a combined plot of each of the liquid limit, plastic limit and plasticity index of the stabilized-contaminated soil samples with their cement content. The plastic limit slightly increases with increasing cement content in contaminated soil while the liquid limit and plasticity index of the contaminated soil samples were decreased. The plasticity index gives a measure of the plasticity of a soil. The application of 10% cement to the contaminated soil reduced its plasticity index by $10.2 \pm 1.35\%$ (32.3%), when compared with the contaminated soil. There is a strong correlation ($r = -0.979$) of the decrease in plasticity indices of the stabilized-contaminated soil samples with the increase in cement content. However, the plasticity after the application of 10% cement to the contaminated soil is still higher than that of the natural soil prior to its contamination with crude oil. Consequently, the increase in plasticity of the soil that followed addition of 10% crude oil were never overcome by the addition of 10% cement.

The clay minerals in the soil exhibit surface charge imbalances and its negatively charged surface became balanced by hydrated cations, thereby forming a thin layer of water (diffuse double layer) bonded to its surface. The contamination of the soil with crude oil increased the

attractable hydrated cations by the clay minerals and consequently, increased the thickness of the diffuse double layer. This probably explains why the plasticity index of the crude oil-contaminated soil increased. The application of cement to the contaminated soil resulted in the exchange of monovalent (hydrated) cations in the contaminated soil with divalent cations (such as Ca^{2+} and Mg^{2+}) present in the cement. Beetham et al. (2014) stated that the valence of the charge-balancing cations, among several factors, primarily controls the thickness of the diffuse double layer. Consequently, such balancing of the clay surface charges with divalent cations (resulting in fewer monovalent cations) has been reported to reduce the thickness of the diffuse double layer (Bohn, 2002). This may be responsible for the reduction in the plasticity index of the cement stabilized-contaminated soil.

Figures 5 and 6 show plots of plasticity index against liquid limit with classification of soil according to the American Association of State Highway and Transportation Officials (AASHTO) system and Unified Soil Classification system (USCS), respectively. The plot of the contaminated soil (0% cement) fell within the portion of the AASHTO plasticity chart for clay soil of high compressibility (Figure 5). As the cement content for treatment of the soil increases, its plot progressively shifts closer to both the silt and the low compressibility portions of the chart. Similarly, the plot of the contaminated soil fell within the portion of the USCS plasticity chart for silts of high plasticity (Figure 6). With increasing cement content in the contaminated soil, the reduction in its plasticity becomes graphically evident. The reduction in its compressibility or plasticity indicates that cement treatment of the contaminated soil made it more workable.

A graphical illustration of the variation of the maximum dry unit weight and optimum moisture content (OMC) of the contaminated soil with its content of cement is presented in Figure 7. As the cement content in the contaminated soil sample increased, its OMC and maximum dry unit weight was decreasing and increasing, respectively. The correlation of each of the decrease in OMC ($r = -0.961$) and increase in maximum dry unit weight ($r = 0.995$) with the increase in cement content is strong.

As stated earlier, the size of the diffuse double layer on the surface of the clay minerals in the stabilized-contaminated soil decreases with increasing cement content. Therefore, the clay particles tend to approach each other more and get clumped-together with increasing cement content in the soil. As the clay particles of the soil agglomerate, the clay-sized particles in the stabilized-contaminated soil and consequently the surface area of the soil particles decrease. Therefore, the stabilized-contaminated soil requires less water to reach optimum (Akinwumi, 2014). The higher specific gravity of cement (typically 3.15), compared with that of the contaminated soil (2.51), is responsible for increasing maximum dry unit weight of the stabilized-contaminated soil as its cement content increases. As the cement content increases in the contaminated soil, there was increasing cementation of some of the particles of the

contaminated soil. This led to a decrease in the volume of the stabilized-contaminated soil and may also have contributed to the increase in its dry unit weight.

Figure 8 shows how the CBR of the stabilized-contaminated soil varies with its cement content. The unsoaked and soaked CBR values of these soil samples progressively increased with increasing cement content. The application of 10% cement to the contaminated soil increased its unsoaked and soaked CBR values by $103 \pm 6.22\%$ (468.2%) and $25 \pm 1.36\%$ (312%), respectively. The correlation of each of the increase in unsoaked CBR ($r = 0.984$) and soaked CBR ($r = 0.999$) values with the increase in cement content is strong.

The soil became more compressible (having a higher dry density) by the application of crude oil, thereby increasing its unsoaked CBR value (Table 1). The formation of cementitious compounds such as calcium-silicate-hydrates and calcium-aluminate-hydrates in the cement-stabilized contaminated soil is responsible for the improvement in its unsoaked and soaked CBR values (Prusinski and Bhattacharja, 1999).

Figure 9 shows the variation of UCS of the stabilized-contaminated soil with its cement content. The UCS slightly increases as the cement content in the stabilized-contaminated soil increases. The UCS of the contaminated soil became increased by $32 \pm 6.53 \text{ kN/m}^2$ (8.2%) after the application of 10% cement content. The correlation between the UCS of the stabilized-contaminated soil and its cement content is strong, $r = 0.988$.

The contamination of the soil with crude oil increased its hydrogen ion (monovalent cation) concentration and consequently, increased the thickness of the diffuse double layer. Therefore, the soil fabric gets deflocculated, thereby reducing its shear resistance and leading to the loss in its UCS (Table 1). As the cement used to stabilize the contaminated soil increased, the cementation between the soil particles also increased – leading to increased inter-particle cohesion and frictional resistance. This resulted in the increase in the UCS of the stabilized-contaminated soil. However, the UCS after the application of 10% cement to the contaminated soil is still lower than the UCS of the natural soil prior to its contamination with crude oil. It is likely that some of the cement particles must have been coated with the crude oil, thereby retarding or preventing their hydration.

A graphical illustration of the change in the permeability of the stabilized-contaminated soil with its cement content is shown in Figure 10. As the cement content increases, the coefficient of permeability progressively decreased. The coefficient of permeability of the contaminated soil became reduced by $2.8 \times 10^{-7} \pm 0.21 \times 10^{-7} \text{ cm/s}$ (56%) after the application of 10% cement content. The decrease in permeability of the stabilized-contaminated soil as its cement content increases is strongly correlated, $r = -0.981$.

The contamination of the soil with crude oil and the compaction of the mixture led to its particles being more closely-packed than the particle-packing of the natural soil. The void in the contaminated soil becomes thereby reduced and occupied by both the crude oil and water. The crude oil in the void reduces the effective void through which water will flow. This, consequently, resulted in the lesser value of coefficient of permeability of the contaminated soil (Table 1). As the cement content in the stabilized-contaminated soil increases, the progressively increasing cementation of the soil particles led to further reduction of the void in or the porosity of the soil. The decrease in the porosity of the stabilized-contaminated soil accounts for the decrease in the coefficient of permeability with increasing cement content (Akinwumi and Ukegbu, 2015; Akinwumi and Booth, 2015).

4. Discussion

The effect of the addition of cement on the UCS and CBR of the crude oil-contaminated soil is similar to that obtained by Nasr (2014). Nasr (2014) investigated the effect of cement kiln dust on the UCS and CBR of an oil-contaminated sand; and reported that the UCS and CBR values of the sand increased with the addition of cement kiln dust. Tremblay *et al.* (2002) reported that the presence of oil and hydrocarbons in a soil stabilized with cement only delays the hydration of the cement but that the oil does not affect the final (ultimate) strength of the stabilized-contaminated soil.

Comparing the results of the contaminated soil and the stabilized-contaminated soil with standard requirements of TRL (1993) shows that the geotechnical properties of the contaminated soil barely met the requirements for use as a subgrade material but became improved after being stabilized with cement such that it sufficiently satisfies the requirements for use as a subgrade material for road pavement construction. It, in fact, satisfies the strength requirement for use as sub-base material but did not meet the plasticity requirements. Consequently, the cement stabilisation of crude oil-contaminated soil that is similar in properties to that studied can provide a stable supporting structure as well as a capping layer that prevents the oil contaminant beneath the capping layer from interacting with the construction materials of structural foundation or pavement structure placed above it.

Caution, however, needs to be exercised while using the findings of this research to extrapolate to other levels of crude oil contamination and to other soils. In these cases, there will be need for an investigation to determine the geotechnical properties of the contaminated soil and the level of cement treatment that would produce desired properties.

5. Conclusions

The contamination of the soil with 10% crude oil increased its plasticity and generally reduced its strength and permeability while the treatment of the contaminated soil with Portland cement reduced its plasticity, increased its strength and further reduced its permeability. The stabilized-

contaminated soil with reduced plasticity becomes a more workable material for earthworks. The increase in CBR and UCS of the stabilized-contaminated soil means that the application of cement made the crude oil-contaminated soil more stable, having a higher bearing capacity. Thus, making a layer of such stabilized-contaminated soil to act as a capping layer and become better suited to receive a structural foundation or road pavement structure. Such a layer, due to its highly-reduced permeability, can also act as a hydraulic barrier preventing structures placed on it from being contaminated or weakened by crude oil or oil-contaminated water beneath it. The cement stabilization of a crude oil-contaminated soil is practically possible to undertake on site.

The potential implications of these findings indicate that the crude oil-contaminated soils with similar properties as that studied, which would have been excavated and disposed off, thereby causing unnecessary waste, prior to commencement of any proposed road or building construction, will become useful construction materials for earthwork. Furthermore, the cement stabilisation of a crude oil-contaminated site having soil properties similar to that of the contaminated soil studied will reduce the project costs associated with the excavation of the contaminated soil, and that of obtaining and transporting suitable soil materials to the construction site.

Finally, Portland cement is recommended for improving the geotechnical properties of crude oil-contaminated soil with similar properties as that studied in order to make them suitably used as a subgrade capping layer during road construction or as a stabilized earth foundation to receive the structural foundation of buildings. There is need for further research works, especially, involving longer soaking periods of the samples in order to determine whether they deteriorate over the time of being subjected to immersion in water and to simulate periods of longer saturation of the treated soil to take account of areas where the water table is close to or has the potential of affecting the treated soils.

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Figure captions

Figure 1. Al_2O_3 - SiO_2 - Fe_2O_3 ternary plot for the uncontaminated soil

Figure 2. Chemical composition of the soil and cement

Figure 3. Particle size distribution of the uncontaminated soil

Figure 4. Variation of Atterberg limits with cement content (error bars indicating the standard deviation)

Figure 5. AASHTO plasticity chart showing the plots of the mean values of the contaminated and stabilized soil samples

Figure 6. USCS plasticity chart showing the plots of the mean values of the contaminated and stabilized soil samples

Figure 7. Variation of compaction characteristics with cement content (error bars indicating the standard deviation)

Figure 8. Variation of CBR with cement content (error bars indicating the standard deviation)

Figure 9. Variation of UCS with cement content (error bars indicating the standard deviation)

Figure 10. Variation of permeability with cement content (error bars indicating the standard deviation)