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IMPACT OF VOLTAGE ON ELECTROKINETIC REMEDIATED LEAD CONTAMINATED SOIL FOR USE AS ROAD BASE MATERIAL

1 (Sani, John E., Department of Civil Engineering, Nigerian Defence Academy, Kaduna, Nigeria) 2 (Tijani, Seidu., Department of Civil Engineering, Baze University, FCT-Abuja, Nigeria) Corresponding Author: **seidu.tijani@bazeuniversity.edu.ng**

3 (Rotimi, Abdulazeez J., Department of Civil Engineering, Baze University, FCT-Abuja, Nigeria) 4 (Moses, G., Department of Civil Engineering, Nigerian Defence Academy, Kaduna, Nigeria)

ABSTRACT : Impact of voltage on electrokinetic remediated lead contaminated soil to determine the soil's suitability for use as road base material was investigated in this research. Although, electrokinetic remediation method have proven to be more effective than most traditional techniques used in remediating low permeability soils contaminated with lead, there is still the problem of "the optimal voltage of the electrokinetic remediation process" that is most likely to produce the best result. The lead contaminated soil was remediated at 15V, 30V, 45V and 60V. At each voltage applied, index and geotechnical properties tests were conducted on the soils after remediation. Generally, the soil is non-plastic belonging to the A-4 group of soil. There is general improvement in all the geotechnical properties of the remediated soil. These improvements are maximum at 30V, there is little or no further improvement beyond this point. The CBR values indicated that none of the materials is suitable for use as road base material as the results do not satisfy the minimum 80% required. But the materials compacted using BSH met the minimum requirement of 40% CBR value and can be used as sub-base materials. Other results showed that the durability with resistances of 89% and 90% to loss in strength was recorded at 30V and 45V respectively, this, when compared to the resistance to loss in strength of 71% in un-remediated soil has respectively 25.3% and 26.8% durability advantages. It is recommended that 30V is just suitable for remediation purpose since less energy is used during the entire remediation process compared to 45V and 60V. All the UCS values failed to satisfy the 1700kN/m2 and 3000kN/m2 recommended for sub-base and base course materials respectively. With WAS compaction effort, the material remediated at 15V, 30V, 45V and 60V met the requirement of 80% resistance to loss in strength making the soil suitable for use as base course material.

KEYWORDS: Electrokinetic remediation, heavy metal, contamination, California bearing ratio, unconfined compression strength, durability. --

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I. INTRODUCTION

Electrokinetic is an emerging remediation technological method that has attracted increased interest among engineers and researchers in the last decade due to several promising laboratory and pilot-scale studies and experiments. This method of remediation aims to remove heavy metal contaminants from low permeability contaminated soils under the influence of an applied Direct Current (DC). The metals would be desorbed under an acidic or low pH environment at the anode during the electrokinetic treatment. Lead (Pb) is a typical heavy metal which refers to a group of toxic elements that are both biologically and industrially important. The widespread contamination of soil with lead and other heavy metals causes one of the most complex

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environmental problems that can seriously affect environmental quality and soil properties. Heavy metals are released into the soils through both natural and anthropogenic processes. Although soil contamination of heavy metals started in ancient times, the problem was provoked after the industrial revolution owing to dramatic increase in the use of heavy metals in various modern technologies. Contaminants can be categorized as chemical, biological, physical, or radiological substances. Other heavy metals commonly present in soils include nickel (Ni), lead (Pb), cadmium (Cd), arsenic (As), chromium (Cr), copper (Cu), cobalt (Co), zinc (Zn), manganese (Mn), aluminum (Al) and mercury (Khalid et al.; 2017).

Soil is an important engineering material; they are used in construction of roads, buildings, dams, embankments, reservoirs etc. The soils must have and maintain good engineering properties in accordance to specifications to ensure adequate support and safety to the structures. Conventional remediation methods for heavy metal are generally based on physical, chemical and biological approaches, which may be used in combination with one another to clean-up heavy metal contaminated soils to an acceptable and safe level. Contrasting to organic contaminants, heavy metals are somewhat unique by the fact that they are extremely not easily degraded by either biological or chemical induced means. Therefore, total heavy metal contents of soil persist for a long time after being introduced into the soil. For example, Pb has a soil persistence period of 150–5000 years (Khalid etal., 2017, Haruna et al., 2023). Excessive accumulation of heavy metals in the soils may cause deterioration of the soil engineering properties and create other environmental issues. Soil physico-chemical properties such as pH, electrical conductivity, cation exchange capacity, soil mineralogy, and presence of soil inorganic and organic ligands greatly influence the bioavailable and mobile heavy metals in soil (Shahid et al., 2012, Minnikova et al., 2017). Many studies have reported heavy metals build up in soil and associated risks to soil quality and biochemical activities (enzymes and microbes). Heavy metals at higher concentration are known to influence the growth of microbes in soil and their activities may directly affect soil properties (Minnikova et al., 2017). Numerous soil remediation techniques have been developed during the last two decades (Murtaza et al., 2014, Sabir et al., 2015). These techniques aim to reduce the total and/or bioavailable fractions of heavy metals in soils. Despite the efficiency, most of these techniques are costly, destructive to the environment and time consuming. Issues of capital and technical complexities have made soil clean-up a difficult task. Practical implementation of these conventional soil remediation methods faces several shortcomings and may introduce some level of hazard. Electrokinetic process has been effectively applied in remediatng both heavy metals and organic compounds contaminated soil. Chelating agents such as EDTA, citric acid, NaNO3 etc can be used to enhance the efficiency of electrokinetic remediation. A low intensity Direct Current is applied across pairs of electrode that have been planted in the ground on either side of the contaminated soil volume. The electrical current triggers electro-osmosis, electromigration and electrophoresis, which move the aqueous phase contaminants below the soil surface from one electrode to the other. Contaminants in the aqueous phase or contaminants desorbed from the soil surface are transported towards respective electrodes depending on the charge. The results from many laboratory studies have shown excellent contaminant removal efficiencies by the use of the electrokinetic technique (Reddy et al., 2006, Haruna et al., 2023).

According to Malavika and Deepthy (2017), after studying the effectiveness of electrokinetic remediation in decontaminating lead contaminated soil. They evaluated the efficiencies of different chelating agents such as 0.1M EDTA, 0.1M citric acid and tap water. They used different electrode materials which include copper and graphite and conducted various tests to determine properties of the soil. Their setup consisted of an electrokinetic cell, two electrodes, 5 Ampere DC supply, multimeter. The cell has three (3) compartments, the middle been the largest containing the contaminated soil. The other two smaller compartments which is located on either side of the middle cell was meant to contain the chelates and the electrodes. A low voltage DC of 20V was applied throughout the cell. All the tests conducted lasted for 24 hours. The electrolytes used for the study were 0.1M EDTA, 0.1M Citric acid and tap water. The soil was partitioned into three regions namely S1, S2 and S3 representing cathode, middle and anode regions in that order. From their results, it was clear that $0.1M$ EDTA showed better result than 0.1M citric acid and tap water. Concentration of Pb was higher in region S1 and less in other regions which indicates metal migration from the soil. Thus, it was found that the migration Lead occurred from anode to cathode region. When tap water and citric acid were used as electrolyte, the Pb mobility was poor, which resulted in less removal efficiency of lead. The result showed that EDTA removed about 68% of Pb from the contaminated soil. In determining the efficiency of the different electrode materials (graphite and copper) used, test to determine concentration of lead at different sections after the electrokinetic process was conducted. From the results, the removal efficiency of lead in anode region is same in both cases, but graphite showed better migration of lead in all the three regions. The difference in removal efficiency with increase in time duration was also studied, they considered 24hrs and 48hrs from where they discovered that

more amount of lead was concentrated towards the cathode region as the time duration increased. Some authors used more than one washing chelators for sequential extraction/washing of heavy metals contaminated soil. In their work, Wei et al., (2016) reported that phosphoric-oxalic acid-Na2EDTA order-based soil washing enhanced heavy metals removal efficiency by 41.9% for As and 89.6% for Cd.

According to Douay et al. (2008), who carried out soil replacement remediation project in three sites near Metaleurop Nord smelter situated in the North of France, having high concentrations of Lead (Pb) and Cadmium (Cd) in the top soils. They delineated a surface area of about 50–100 m2 for each site, removed the contaminated soil and replaced it with a non-contaminated one. After soil replacement, cultivation of crops on the site between 2003 to 2005 showed a clear improvement of yields and soil quality. Soil replacement method can effectively isolate the contaminated soil and ecosystem, thus minimizing its effect on the environment. However, this technique is costly due to high labor work but it's appropriate for heavily contaminated soils over small area.

Dellisanti (2016) carried out an in-field Joule heating vitrification of tons of Zn and Pb rich ceramic waste by heating up to about 1850°C. He reported that the vitrification method was greatly efficient to clean-up tons of heavy metals contaminated waste materials and can be applied for cleaning huge volumes of soil. Temperature during vitrification plays a key role in the immobilization of heavy metal in soil samples. For example, Navarro et al. (2013) carried out vitrification of waste from Ag-Pb mines in Spain using solar technology. They showed that vitrification caused immobilization of Zn, Mn, Fe, Cu and Ni at 1350 °C, whereas Zn, Ni, Mn and Cu were mobilized at 1050°C.

Vitrification can be performed both in situ and ex situ, although in situ method is preferred due to comparatively low cost and energy requirements. Ex situ vitrification processes contains various stages such as excavation, mixing, pretreatment and melting feeding. The main limitation of in situ vitrification is the potential of the soil to melt so that current can pass through it. Therefore, in situ vitrification can be performed only under wet soil with low alkali content. This technique can be applied for small scale remediation of heavy metals polluted sites. Under field conditions or at large scale, this technique can be highly expensive.

Rosestolato et al. (2015) remediated approximately 400 kg of soil using electrokinetic remediation and removed about 60% of total Mercury (Hg) from the contaminated soil in three months. Electrokinetic remediation method was also used in combination with other techniques/processes such as electrokinetic microbe joint remediation (Yu et al., 2009), electrokinetic-chemical joint remediation, electrokinetic-oxidation/reduction joint remediation (Yang et al., 2015), coupled electrokinetic phytoremediation (Mao et al., 2016), electrokinetics coupled with electrospun polyacrylonitrile nanofiber membrane (Peng et al., 2015), and electrokinetic remediation conjugated with permeable reactive barrier (Rosestolato et al., 2015).

Lee et al. (2016) used Potassium dihydrogenphosphate (KH2PO4) as an anolyte and showed that removal efficiencies increased by greater than 50% for Arsenic and greater than 20% for Copper. However, removal of the Pb and Zn was relatively inefficient (below 20%).

Rozas and Castellote (2012) studied the effectiveness of different experimental variables (type of electrolyte, constants of precipitation (pKs) and chelation equilibriums, initial and final pH of the catholyte and anolyte, and zeta potential) for different electrolyte solutions (distilled water, acetic, citric acid and ethylene diamine tetraacetic acid (EDTA)). They reported that the zeta potential, pH of the cathodic solution influenced the efficiency of electrokinetic remediation. Mao et al. (2016) used coupled electrokinetic phytoremediation and showed that electro-kinetic field lowered soil pH to around 1.5 and enhanced dissolution of Pb, As and Cs, thus increasing their overall solubility.

Electrokinetic remediation method operates well for soils having low permeability; this is according to Hanson et al., (1992). It is economically effective because it is easy to install and operate (Virkutyte et al., 2002), moreover, electrokinetic remediation does not abolish the original nature of the soil (Page and Page, 2002).

However, even though electrokinetic methods have proven to be more effective than most traditional techniques used in remediating low permeability soils contaminated with mixed heavy metal contaminants, there are still challenges, the application of optimal voltage gradient and the effective remediation of contaminated soil (Mohammed et al., 2013). Most of the works reviewed on electrokinetic remediation failed to state the optimal voltage for electrokinetic remediation process, let alone monitor the effect of its variation on soil properties. The usual voltage gradient of 1V/cm for bench-scale studies when applied to soils could lead to excessive soil heating, reduction in the soil moisture content, high energy and process fluid consumption (Mohammed et al., 2013). Hence, the focus of this research work is to vary the voltage applied to an electrokinetic remediation process and evaluate the impact on the geotechnical properties of the soil.

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A. The Objectives of the Research

The objectives of the research are:-

- 1. Determination of geotechnical and chemical properties of the lead contaminated soil.
- 2. To electrokinetically remediate the contaminated soil by varying the voltage at 15V, 30V, 45V and 60V.
- 3. Determination of the geotechnical and chemical properties of the remediated soil accordingly and evaluation of its suitability for use as road base material.

II. MATERIALS AND METHODS

A. Electrokinetic Remediation

The Lead contaminated soil was collected from Birnin Gwari Local Government Area mining site, Kaduna State, Nigeria.

The electrokinetic remediation setup in this study shown in Fig.1 below was based on the model adopted by Bimastyaji et al., (2018). The setup consists of a cell (like a cuboid) made from plexi glass material that is divided into three compartments, the middle partition (30cm x 30cm x 25cm) is the largest containing the lead contaminated soil, while the other two (30cm x 5cm x 25cm) are situated at either end containing the chelating agents. The cell is designed in such a way that it allows fluid flow in all the compartments from one end to the other, to achieve this, a well perforated plexi glass was used as the divider between the three compartments which permits injection of the processing fluid and allow electroosmotic flow between the anode and cathode. Electrodes used for this study was graphite (8mm thick and 30cm long) placed vertically with a face to face configuration at the anolyte and catholyte compartments for passing Direct Current from the DC Supply using connecting cables attached to clips through the set-up. Solar Power set-up was used to ensure continuous power supply to the electrokintic set-up. 0.1mole of EDTA (Ethylenediaminetetraacetic Acid) and 0.01mole of citric acid were used as the chelating agents at cathode and anode compartments respectively. By connecting the setup to a direct current source, remediation process was automatically enabled depending on the voltage.

The remediation was complete when there were no more effluents produced at the cathode and no more change in the current. The remediated soils were collected from the cells and sliced into three (3) parts (cathode region, center, anode region) from which samples were collected to determine the concentration of lead.

B. Schematic Setup of Electrokinetics System

The schematic laboratory set-up of the electrokinetic remediation is shown below.

Fig.1: Laboratory set-up for the Electrokinematic Remediation Process

The Electrokinetic remediation experiment was conducted at the Civil Engineering Laboratory, Nigerian Defense Academy, Kaduna. The remediation was in four (4) phases by varying the voltage at (15Volts) 0.5V/cm, (30Volts) 1V/cm, (45Volts) 1.5V/cm and (60Volts) 2V/cm on four (4) lead contaminated samples

accordingly. The geotechnical properties of the remediated soils were conducted at the Geotechnical Laboratory, Civil Engineering Laboratory Department, Nigerian Defense Academy, Kaduna and Baze University, Abuja. Determination of concentration of Lead (Pb) was carried out using the Oxford Instrument Analyzer (XRS-FP analysis).

The engineering properties which include moisture content, specific gravity, particle size distribution, atterberg limit (liquid limit, plastic limit), British Standard Light (BSL) compaction, British Standard Heavy (BSH) compaction, West Africa Standard (WAS) compaction tests were determined accordingly.

C. Unconfined Compression Strength (UCS)

The remoulded specimen for the test had a minimum diameter of 38mm with a height of 76mm. The disturbed sample was compacted using the applicable compactive effort under consideration and the appropriate optimum moisture content; the sampling tubes were then driven into the compacted soil to obtain a cylindrical specimen. The specimen after extrusion was trimmed appropriately to conform to the dimensions above. The specimens were then wrapped in a thin rubber membrane. Enough specimen was prepared to allow curing in open air for 7, 14, 21 and 28days. On attaining the curing ages above, the specimen was unwrapped and positioned in between the bottom and upper plates of the UCS testing machine. The dial gauge was adjusted to 0 and the loading stearing was properly seated. Then, compression load was applied to induce deformation on the specimen so that axial strain can be recorded at 0, 20, 40, 60, 80, 100, 120, 140, 160, 180, 200, 250, 300, 350, 400, 450, 500, 550, 600, 650, 700, 750, 800, 850, 900, 950, 1000, the maximum compressive load that caused failure of the specimen was obtained and used in the calculation for the Unconfined Compressive Strength (UCS). Mathematical,

$$
\text{UCS} = \frac{\text{Dial gauge reading } X \text{ PRF } X \left(1 - \frac{\Delta L}{L_0}\right)}{\text{Cross sectional Area of the Specimen} \text{ kN/m}^2} \tag{1}
$$

Where, PRF = Proving Ring Factor, Lo = Original Length, ΔL = Change in Length. However, the durability test was conducted by curing (soaking) the cylindrical specimen obtained from each of the compactive efforts (BSH, BSL and WASC) in water for 7days. After which the specimen were tested and the strength calculated.

D. Experimental determination of the Durability of soil (resistance to loss in strength)

Remoulded samples of the remediated soil were compacted into split moulds of 38mm internal diameter and 76mm length to their respective optimum moisture content using all the three energy levels (BSH, BSL and WASC). The cylindrical compacted samples were placed into water proof polythene nylon to cure. Three samples were removed from each energy level and tapped. The tapped samples were left standing in a bowl of potable water to allow for absorption of water for 7days. The samples were retrieved after 7days curing and tested for strength. Hence the resistance to loss in strength is determined from the equation.

$$
UCS(7 days curved + 7 days soaked) \n= UCS(14 days curved) \n\times 100 \n(2)
$$

E. California Bearing Ratio, CBR

The California Bearing Ratio (CBR) test is an indirect measurement of resistance of soil material to penetration of standard plunger under controlled density and moisture conditions. The sample was initially sieved through the 20mm sieve from which 6kg of the sample of soil specimen was weighed. Water was added to the soil in the quantity such that optimum moisture content was reached. Then the soil and water are mixed thoroughly. Spacer disc was placed over the base plate at the bottom of mould and a coarse filter paper is placed over the disc. The prepared soil was divided into five. The mould was cleaned and lubricated. Then one part of the mould was filled with one part of the prepared soil. That layer was compacted by giving 62 evenly distributed blows using the 4.5kg rammer; this was repeated for the remaining four parts of the soil. Then the excess soil was strucked off. The base plate was then removed and the mould inverted. Then it was clamped to the baseplate. Surcharge

weights of 2.5kg were placed on top surface of the soil. Mould containing specimen was placed in position on the testing machine. The penetration plunger was brought in contact with the soil. Then dial reading was adjusted to zero. Load was applied such that penetration rate was 1.25mm per minute. Load at penetration of 0.5, 1, 1.5, 2, 2.5, 3, 4, 5, 7.5, 10 and 12.5mm were noted. Hence, California Bearing Ratio was calculated at 2.5mm and 5.0mm from which the average is evaluated. The top and bottom of the compacted soil was tested. Then it was completely soaked in water for 24hrs. After soaking, the sample was tested again for CBR.

The method was repeated for both BSL (with 62 blows, 3layers using 2.5kg rammer) and WAS (with 10 blows 5 layers using 4.5kg rammer).

III. RESULTS AND DISCUSSION

The index properties and compaction characteristics of the electrokinetic remediated soil are shown in Tables 1, 2 and 3 below.

Table 1: Sieve analysis and Liquid limit results of remediated and unremediated soil

Table 2: Specific gravity and moisture results of remediated and unremediated soil

Table 3: Compaction test results of remediated and unremediated soil

Fig.2 : Sieve analysis and liquid limit test results

The Nigerian General Specifications for Roads and Bridges (2016) requires that liquid limit, plastic limit, gradation, density/moisture content relation and CBR be carried out on materials for use for the classification of compacted base and sub-base (Clause 6201). In Fig.2 above, the results showed that the soil is generally silty fine sand with not less than 40% passing the sieve no.200. 45% passed through sieve 200 for unremediated soil and slightly reduced to 40%, 40.4% and 40.2% at 30V, 45% and 60V respectively. According to AASHTO (American Association for State and Highway Transportation Officials) soil classification system, all these samples (unremediated and remediated) fall into the A-4 group of soil (Silty soils).

Other index properties conducted on the soil samples are Specific Gravity and Moisture Content as presented in Fig.3 below.

Fig.3: Specific gravity and moisture content test results

An average Moisture of 24% (Fig.4) was required during the remediation processes. Specific Gravity of the soil particles lie between 2.65-2.85. Soil containing organic matter and porous particles may have Specific Gravity below 2.0 and soils having heavy particles may be as high as 3.0 or more. As indicated in Fig.3 above, the specific gravity ranges between 1.9 and 2.2 which shows that the soils are generally light weight.

Fig.4: Maximum dry density with voltage at different compactive efforts

Fig.5: Optimum moisture content with voltage at different compactive efforts

Figs. 4 and 5 shows the maximum dry density and optimum moisture content respectively, the test was used to determine the relationship between moisture content and dry density of soil for a specified compaction energy level. The results indicate an improvement in maximum dry density (MDD) of the remediated soil at 30V, 45V and 60V across all the three compaction energy levels. Comparing the MDD at 30V $(1.920mg/cm³)$ to that of the unremediated soil (1.905mg/cm³), there is a slight increment of 0.8% at an average optimum moisture content of 10% when BSH compaction energy was used. This percentage increase is similar in all the compaction energy levels that were used across all the voltage variations. These trends of results are consistent with the findings of Haruna et al., (2023).

Fig. 6: Unsoaked CBR with Voltage at different compactive efforts

Fig. 7: Soaked CBR with Voltage at different compactive efforts

The California Bearing Ratio (CBR) is an indirect test method performed to evaluate the strength of the Lead contaminated soil before and after remediation, the test was to determine the suitability of the soil for use as base course materials. Figs.6 and 7 showed the CBR values of unsoaked and 24hrs soaked compacted samples using all the three energy levels of BSH, BSL and WAS.

Material for compacted sub-base and base courses specified by clause 6200 of the Nigeria General Specifications for Roads and Bridges (2016) requires that the minimum strength of such base course material shall not be less than 80% CBR value being determine at maximum dry density and optimum moisture content unsoaked.

The specifications stipulate that for a material to be suitable for sub-base, the CBR (24hrs soaked) must not be

less than 30% (Clause 6201). None of the soil samples is suitable for use as base course material but used as sub-base material since the unsoaked CBR values obtained from the BSH compaction exceeds the minimum requirement of 30%. Details of soaked and unsoaked CBR results are presented in table 4 below

	UNSOAKED			SOAKED		
	BSH	WAS	BSL	BSH	WAS	BSL
0V	41	24	21	16	8	
15V	42	24	20	16	8	
30V	51	28	22	15	8	8
45V	52	27	22	16	8	9
60V	58	30	23	16	10	8

Table 4: Summary of CBR test results

Fig.8: UCS with Voltage at different compactive efforts

Presented in Fig. 8 above was the UCS test results obtained at 21days of curing (details in Table 5). The Unconfined Compression Strength (UCS) test results were carried out to determine the strength development of the soil before and after remediation. The test samples were tested at 7, 14, 21 and 28days of curing in an openair. At BSH compaction effort, more than $700kN/m²$ were recorded at 21days of curing, there was gradual fall in this value with BSL and WAS compaction efforts as shown. All the UCS values failed to satisfy the minimum values of 1700kN/m² and 3000kN/m² recommended by Ingles et al., (1973) for base course and sub-base materials respectively.

	BSH	WAS	BSL
0V	71	77	62
15V	77	83	76
30V	89	82	75
45V	90	83	77
60V	76	85	77

Table 5: Resistance to loss in strength, (%)

However, the resistance to loss in strength presented in table above indicate that the highest durability with resistances of 89% and 90% at 30V and 45V respectively using the British Standard Heavy (BSH) compaction.

This, when compared to the resistance to loss in strength of 71% in unremediated soil, has respectively 25.3% and 26.8% durability advantages. Also, with WAS compaction effort, the material remediated at 15V, 30V, 45V and 60V meet the 80% resistance to loss in strength required for a durable soil material recommended by Ola (1974) making the soil suitable for use as base course material.

IV. CONCLUSION

- All soil materials (remediated and unremediated) tested belongs to the A-4 group of soils, a silty sand material.
- None of the materials is suitable for use as road base course material, the unsoaked CBR values does not satisfy the specified minimum value of 80%.
- Similarly, none of the materials is suitable for use as road base course material as the UCS result does not satisfy the recommended 1700kN/m²
- The soil is suitable for use as sub-base material; the unsoaked CBR values obtained from the BSH compaction exceeds the minimum requirement of 30%.
- With WAS compaction effort, the material remediated at 15V, 30V, 45V and 60V meet the 80% requirement for resistance to loss in strength making the soil suitable for use as base course material.

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