

Research Article

Test on the Stabilization of Oil-Contaminated Wenzhou Clay by Cement

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Oil-contaminated soils have been paid much attention due to the reclamation of industrial lands in coastal cities of China. As known, oil-contaminated soils are inapplicable for construction due to their weak engineering properties, thus leading to the requirement of remediation and reclamation for oil-contaminated sites. This study presents an experimental investigation on the stabilization of contaminated soils with Portland cement. Investigations including the Atterberg limits, unconfined compressive strength, direct shear strength, and microstructure of cement-stabilized soils have been carried out, verifying the suitability of applying cement to improve engineering properties. Experimental results show that the geotechnical properties of contaminated soil are very poor. With the application of cement, the liquid limit and plasticity index of contaminated soil samples decrease dramatically, and the strength of treated soils has been improved. Experimental results from scanning electron microscope (SEM) indicate that cement-stabilized oil-contaminated soil is featured with a stable supporting microstructure, owing to the cementation between soil particles. This also confirms the applicability of cement to be served as an additive to treat oil-contaminated soils.

1. Introduction

Remediation and reclamation of oil-contaminated sites has been paid increasing attentions in recent decades. This is mainly due to the fast development of industry all over the world and its impact on global environment [1, 2]. As known, oil spills during transportation, leakage from storage tanks, discharge of industrial waste liquid with oil, and so on would result in serious contamination to soils. For example, leather industry is popular and advanced in Wenzhou, China, which is regarded as one of the most important economic pillars for the local government. However, due to the loose regulation and lagging industry in the past few decades, liquid industrial waste was dumped without proper treatment in this region [2]. Contaminated soils normally contain toxic heavy metals and chloride salt, which have been suggested to be stabilized using cement [3–6]. However, the stabilization for contaminated soils induced by oil pollution becomes much more complicated and difficult.

Investigations on engineering properties of oil-contaminated geomaterials have been carried out since 1990s. For instance,

Alsanad et al. [7, 8] carried out a laboratory test to investigate the effects of oil contamination on strength parameters, compressibility, permeability, and compaction characteristics of the Kuwaiti sand. Later on, Khamchayan et al. [9] carried out series of tests to study the effects of oil contamination on geotechnical properties of clays, and they found that increasing oil content for soil samples would decrease its strength, permeability, maximum dry density, optimal water content, and Atterberg limits. However, they did not provide specific measures to solve this problem. With the booming population in coastal cities around the world, oil-terminated soils have been employed for construction after proper stabilization. Thus, a few measures have been reported in literature for oil-contaminated soils, including the replacement of oily soil with uncontaminated soil, incineration, absorption, biodegradation, and so on, which are time-consuming and expensive [2, 10]. Therefore, some scholars recently commented that soil improvement with cement is attractive for its mature technology and economy [1, 10, 11]. As known, cement reacts with water and binds loose particle materials in a short time, which



FIGURE 1: The location of borrow pit.

would produce strength and durability for soils [12]. For example, Akinwumi [11] applied Portland cement to treat oil-contaminated soil in his/her study. Other study on the stabilization/solidification of contaminated soils [1, 3, 13–17, 26–29] also suggested that cement-stabilized soils have been improved sufficiently and are capable to support a structural foundation or road pavements.

Despite this, the solidifying effect of cement on oil-contaminated soils remains to be studied. First and most importantly, the extent of contamination depends on the chemical composition of the contaminant and the properties of soils [22]. Also, in connection with repair works, geotechnical properties and mechanical behaviors should be investigated for any potential applications of oil-contaminated soils. In addition, mechanism of cement-stabilized oil-contaminated soil is also required through microscale investigation. However, there are limited studies on geotechnical properties of contaminated soils in literature.

Based on the discussion aforementioned, a laboratory testing program was carried out to investigate the effects of improvement on Wenzhou clay with different doses of oil and various contents of cement. The testing included basic soil properties, Atterberg limits, direct shear strength, unconfined compressive strength, and microstructure characteristics of cement solidified natural and contaminated soils.

2. Material and Methods

2.1. Materials. Soil samples used in this study were obtained from a borrow pit at Wenzhou (latitude $27^{\circ}51'N$ and longitude $121^{\circ}08'E$; Figure 1 shows the location), China. The marine clay sludge in coastal area is also named as Wenzhou Clay, with typical characteristics being high moisture content, high compressibility, and low strength [10, 23]. Samples were collected from the sidewall of the soil profile at around 10 m depth. They were stored in labeled and sealed sacks and transferred to Geotechnical Laboratory (Wenzhou University), field subsamples were also taken to determine their natural moisture contents.

Plant oil (peanut and soybean reconciled oil) was applied in this study, for the consideration that its physic-chemical characteristics are similar to that of the petroleum oil (i.e., nontoxic, nonvolatile, etc.). The main components of plant oil (obtained through the product description) are unsaturated fatty acid (around 80%, including 41.2% oleic

acid and 37.6% linoleic acid) and saturated fatty acid (around 20%, including palmitic acid, stearic acid, and arachidic acid). While, the main components of petroleum oil are low levels of heavy metals and total petroleum hydrocarbons (TPH), as reported in literature [13, 14]. Both plant oil and petroleum oil would impose lubrication effects on soil particles, thus leading to lower geotechnical properties of contaminated sites. As known, the main mechanisms involved in the stabilization/solidification of oil-contaminated soils are chemical fixation and physical encapsulation or adsorption [13, 24]. Based on the concept of process of envelopes [13–17], contaminants generated by similar process are characterized by similar physical properties and composition; thus, the treatment method applied to plant oil-contaminated soil also can be applied to other waste of the same type. This explanation may justify the adoption of plant oil to replace petroleum oil in this study. Another consideration is that it is difficult to obtain crude oil nearby; commercial petro oil has been purified and is featured with volatile organic compounds, imposing difficulties on the control of waste dosage with room temperature.

2.2. Experimental Program. Soil samples were oven-dried at $105^{\circ}C$ for 24 hours, then the soil particles were crushed and passed through $75\ \mu m$ sieve. Hereafter, both cement and dry soil sample were mixed by a mixing drum. The dry cement-soil mixture was then sprayed by oil-water mixture, the volume of which was calculated according to the optimal compaction moisture content (OMC) and maximum dry density (OMD) [25, 26]. Finally, the contaminated soil was stored in an airtight plastic container for a period of time and allowed to equilibrate prior to commencement of laboratory tests [27].

Specific gravity, Atterberg limits, compaction, unconfined compressive strength (UCS), and scanning electron microscopy (SEM) tests were conducted on both natural soils, contaminated soils (0%, 0.1%, 1%, and 3% oil contents, by dry weight of soil), and cement-stabilized contaminated soil samples with various proportions of Portland cement (4%, 8%, and 12%, by weight of the soil). These tests were performed for the purpose of studying the effects of cement on the improvement of geotechnical properties of oil-contaminated soils.

The soil plasticity index was determined from liquid and plastic limits tests, with the help of a combined liquid plastic apparatus. The UCS tests were conducted based on the method suggested by ASTM D2166 [28]. Specimens (30 mm diameter \times 80 mm height) were prepared at the optimal moisture content and tested after 0, 7, 14, and 28 days of curing coated with preservative film under controlled temperature ($20 \pm 3^{\circ}C$) and relative humidity (95%). The direct shear test was conducted following a standard approach [29]. Samples were prepared at their optimal moisture contents and tested after 0, 7, 14, and 28 days of curing.

3. Results and Discussion

3.1. Materials Characterization. The chemical compositions of Wenzhou clay were expressed in terms of oxides and

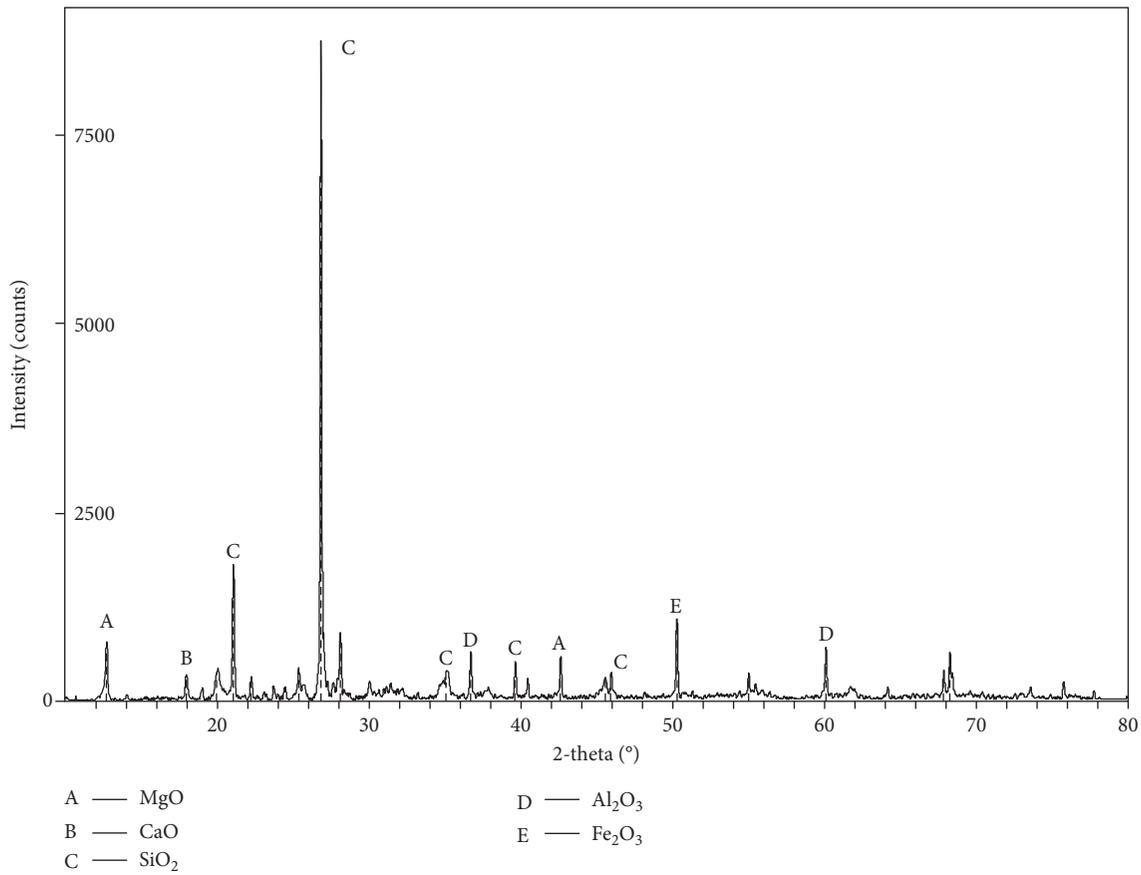


FIGURE 2: X-ray diffractograms (XRD) of uncontaminated clay.

TABLE 1: Chemical compositions of uncontaminated Wenzhou clay (XRD results).

Compounds	Percentage (%)
Silicon dioxide (SiO ₂)	48.6
Magnesium oxide (MgO)	10.9
Aluminum oxide (Al ₂ O ₃)	7.8
Ferric oxide (Fe ₂ O ₃)	5.2
Calcium oxide (CaO)	2.5
Others	24.7

calculated from elemental analysis determined by XRD, as shown in Figure 2, and the main components are listed in Table 1. And the components of cement used in the test are shown in Table 2, which can be obtained directly from the “Production Description” printed on the cement package. It is apparent that the major components of natural Wenzhou clay is silicon dioxide (SiO₂), taking up around 50%, then other oxides; the main content of Portland cement is calcium oxide (CaO), taking up about 60%, followed by silicon dioxide (SiO₂), with a portion of around 24%.

3.2. Geotechnical Properties of the Uncontaminated Soil and the Plant Oil-Contaminated Soil. Basic soil properties, including liquid limit, plastic limit, OMC, specific gravity, maximum dry density (MDD), moisture content, and density of the uncontaminated clay, are presented in Table 3.

TABLE 2: Chemical composition of cement (product description).

Compounds	Percentage (%)
Silicon dioxide (SiO ₂)	23.67
Calcium oxide (CaO)	59.98
Ferric oxide (Fe ₂ O ₃)	3.07
Aluminum oxide (Al ₂ O ₃)	7.21
Magnesium oxide (MgO)	2.07
Potassium oxide (K ₂ O)	0.68
Sodium oxide (Na ₂ O)	0.17
Sulfur trioxide (SO ₃)	2.14
Loss	1.01

This further confirms that Wenzhou clay is characterized by higher water content and high liquid limit.

Figure 3 shows the plots of liquid limit, plastic limit, and plasticity index of oil-contaminated clay samples with different doses of oil. As can be seen, the liquid limit and plasticity index of the contaminated soil samples decrease with the increasing oil content, while the change in plastic limit is not apparent. This can be explained by noting that clay minerals exhibit surface charge imbalance and the negative charged surface turns to be balanced by hydrated cations, thereby forming a thin layer of water (diffuse double layer) bonded to its surface [5, 10, 22]. The attractable hydrated cations provided by the clay minerals have been increased due to the oil contamination, and this consequently increases the thickness of the

TABLE 3: Basic soil properties for uncontaminated Wenzhou clay.

Natural moisture content (%)	Density (g/cm ³)	Specific gravity	Liquid limit (%)	Plastic limit (%)	OMC	MDD
66.0	1.58	2.68	58.0	26.1	30.0	1.45

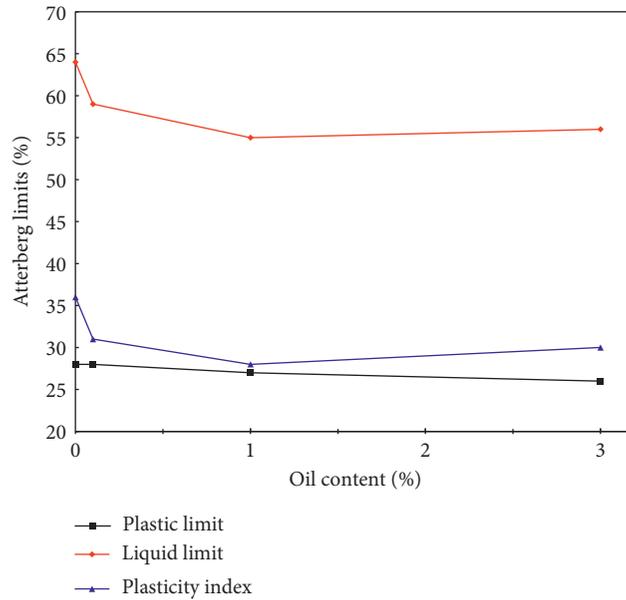


FIGURE 3: Influence of oil content on Atterberg limits of contaminated clay.

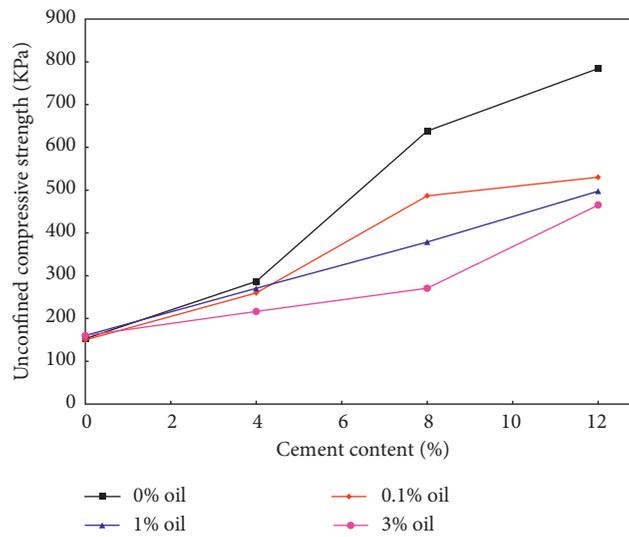


FIGURE 4: UCS of the cement-stabilized oil-contaminated soils with different oil contents.

diffuse double layer. In addition, lubricating characteristics of oil itself also seem to be another reason to explain the decrement of both liquid limit and plasticity limit in oil-contaminated soils.

3.3. *Unconfined Compressive Strength.* Figure 4 plots the variation of UCS of cement-stabilized oil-contaminated soils. It shows the strength of oil-contaminated soils (contaminated by different oil contents) stabilized by an addition

of 12% cement after curing 28 days. UCS of all oil-contaminated soil samples increases with the increasing cement, which indicates that cement can be served as an effective additive to solidify oil-contaminated soils. While, the stabilization effect in soils with lower oil contents is better than that in their counterparts with higher oil contents. The stabilization induced by cement lies on the exchange between monovalent (hydrated) cations in contaminated soils and divalent cations (such as Ca²⁺ and Mg²⁺) provided by cement [30, 31]. Inspection of Figure 4 also shows that, the strength of soil would

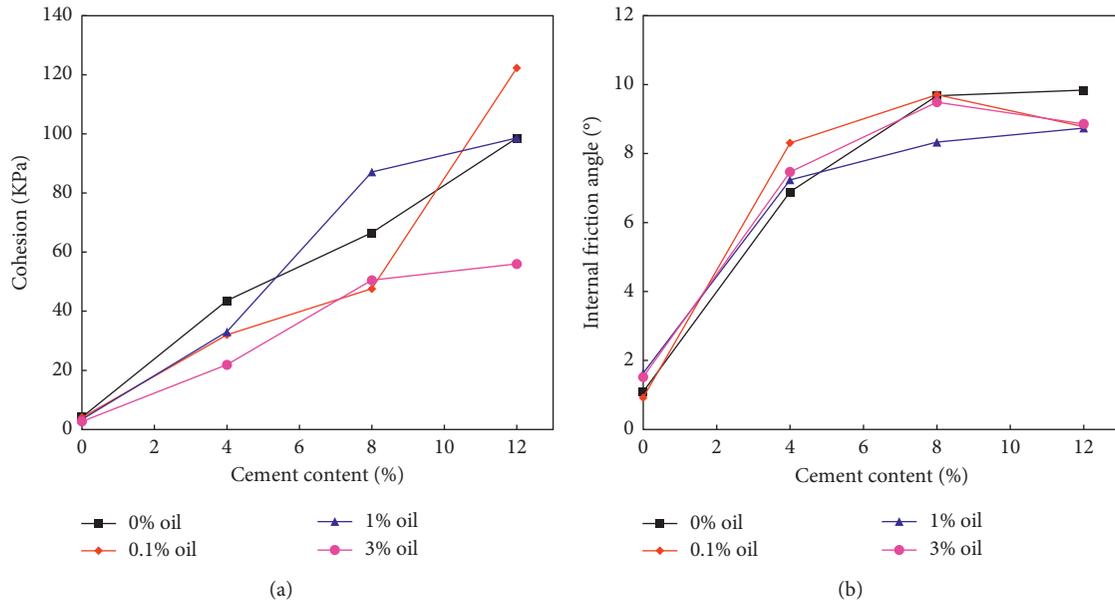


FIGURE 5: Influence of cement contents on shear strength parameters of contaminated soils.

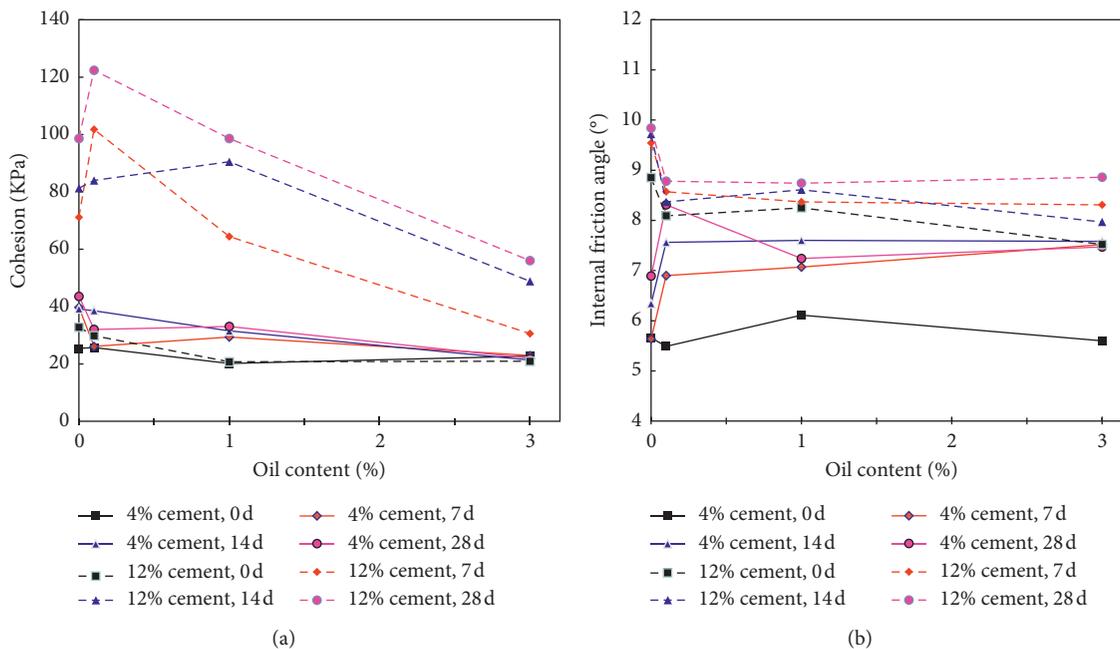


FIGURE 6: Influence of oil content on shear strength parameters of soil samples.

decrease significantly along with increment of oil contents. The presence of oil appears to act as a hydration retarder and decreases the strength of soils. As stated in Section 3.2, the size of diffuse double layer on the surface of clay minerals decreases with increasing cement contents, thus in turn reducing the barriers between soil particles and getting soil clumps together.

3.4. Direct Shear Test. A graphical illustration of changes in cohesion strength (c) and friction angle (ϕ) for the oil-contaminated soil with different doses of cement is

presented in Figure 5. These samples were cured for 28 days before carrying out direct shear test. Obviously, with the increasing cement being added in contaminated soil samples, both cohesion strength and friction angle increase dramatically. This tendency is similar to that of UCS discussed in Section 3.3, particularly for the case of cohesion (Figure 5(a)). While, the friction angle turns to be stable when the dose of cement approaches a certain level (Figure 5(b)). This is because the friction angle mainly depends on the size of soil particles, which may change slowly for cases with higher cement contents [5, 22].

Effects of oil content on the improvement of contaminated soils are demonstrated in Figure 6. It presents a negative correlation between oil content and shear strength parameters (cohesion (c), internal friction angle (ϕ)). There exists a remarkable decrement in cohesion (c) along with increasing oil contents, as shown in Figure 6(a). However, from the plots of soil specimen with 12% of cement (cured for 28 days), one can find that the cohesion value of uncontaminated soil specimen is equal to that of contaminated soils with 1% of oil. This attributes to two main reasons: on one hand, cohesion and internal friction angle can be improved by cement; on the other hand, they get deteriorated by the presence of oil. The competitive results of two contradict mechanisms also can be seen in Figure 6(b) for internal friction angle, which depends on relative amounts of both cement and oil.

4. Scanning Electron Microscopy Analysis

SEM was applied to capture the morphological changes in both natural soils and cement-stabilized oil-contaminated soils. The standard procedure for SEM is as follows:

Step 1: *Specimen preparation*: Soil samples were oven-dried firstly, then they were cut into small thin pieces; hereafter, these thin soil samples were mounted onto a sample holder, the surface of which was covered by fulmargin (Figure 7). Later on, thin soil samples were loaded into an airlock chamber; after the chamber being vacuumed, metal spraying was employed to treat them.

Step 2: *SEM scanning*: Soil samples were mounted into the sample chamber, and then scan parameters were adjusted, and an optimal point for observation was figured out. Thereafter, the magnification was increased to zoom in on details of soil samples, and photos were finally taken (Figure 8).

Figure 9 compares the SEM photomicrograph of cement-stabilized contaminated soils (0% and 3% oil content) with various doses of cement (4%, 8%, and 12%). The curing age of cement solidification soil was 28 days, and the scanning electron microscope (SEM) image magnification was 10000 times.

From the comparative analysis of Figures 9(a), 9(c), and 9(e), it is known that natural soils stabilized by cement are characterized by flocculations, which become denser with the increasing cement. Obviously, the existence of flocculations produced by cement would result in a more stable soil structure with dense particles and thus enhancing its strength. Figures 9(b), 9(d), and 9(f) show the microscopic diagram of cement-stabilized soil contaminated by 3% oil. Similarly, with the increasing dose of cement, the amount of flocculations increased dramatically, indicating an enhanced strength for contaminated soils. This can be explained by noting that flocculations are the product of cement hydration during the curing period, and a larger amount of cement would result in stronger hydration, products of which in the soil particles are the cementation of loose soil particles in the form of flocculations, thereby forming solid aggregates and improving the soil strength [1, 5, 10, 22].

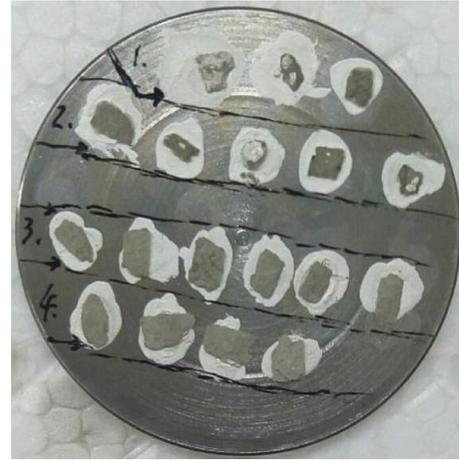


FIGURE 7: Soil samples placed on a sample holder.



FIGURE 8: SEM scanning.

Comparison between Figures 9(a) and 9(b) indicates that with the incorporation of 3% oil, the cement-stabilized contaminated soils were characterized by thin and loose structure. This difference becomes more apparent comparing Figures 9(d) and 9(f) with Figures 9(c) and 9(e), suggesting that flocculations of cement-stabilized contaminated soil formed loose and sliding structures and the cementation effect was weaker in oil-contaminated soil samples. This may be explained as that the addition of additive agents (cement in this study) would produce noncrystalline chemical compounds, which coats over soil particles and bridges them. Coating over solid grains thus in turn enhances the bridging action between soil particles and further results in the development of a strong cellular and nodular mass [32].

5. Conclusions

In this paper, laboratory tests on cement-stabilized oil-contaminated soils were carried out, and the improvement on Atterberg limit, unconfined compressive strength, shear strength, and microstructure were studied. The following conclusions can be obtained based on this study:

- (1) The liquid limit and plasticity limit of contaminated soil decreased significantly compared with that of natural soils, with slight influence on plastic index being observed.
- (2) With increasing oil content, the unconfined compressive strength of cement-stabilized soil decreased

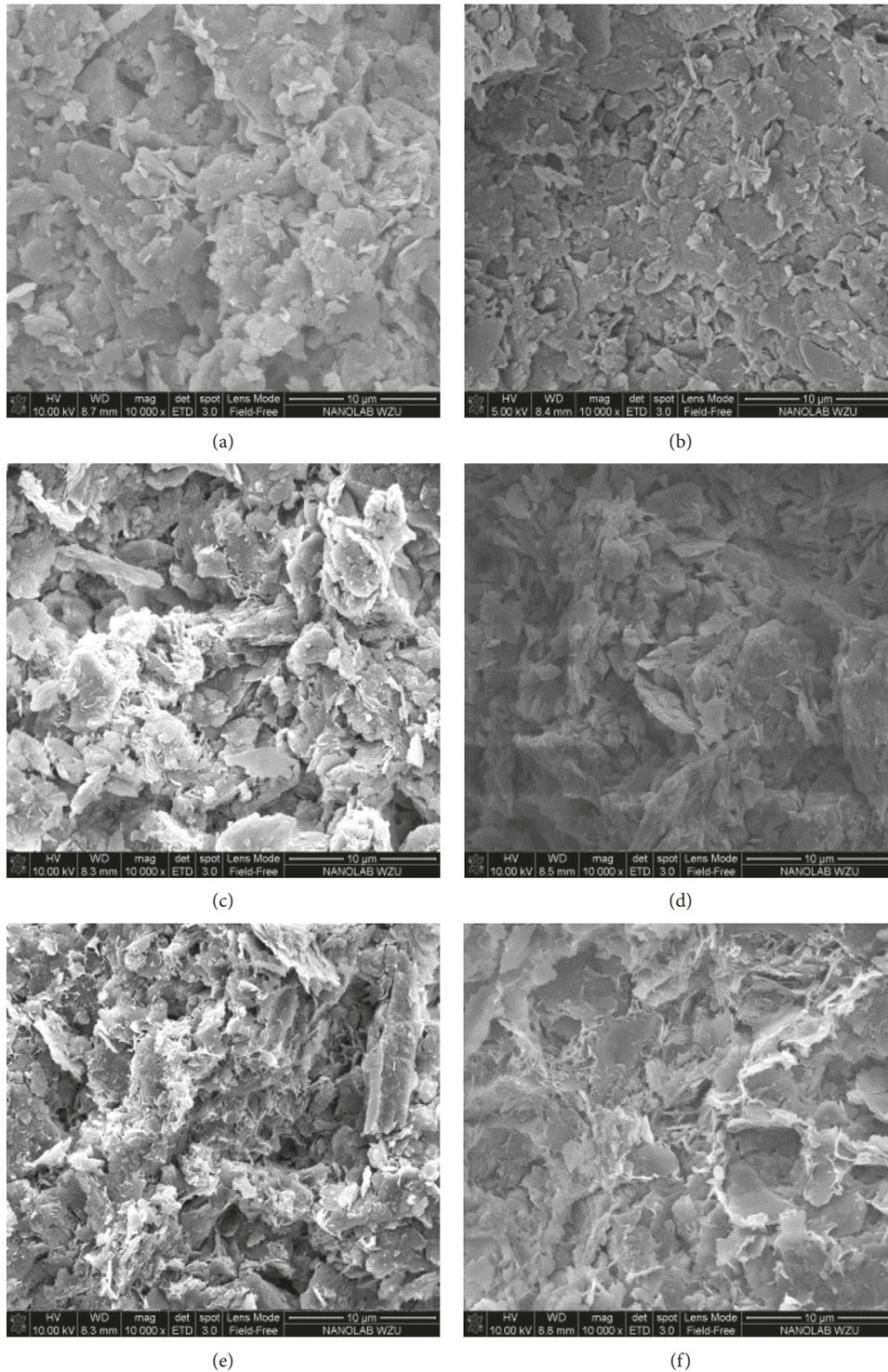


FIGURE 9: SEM photomicrograph of oil-contaminated soils stabilized by cement. (a) 0% oil; 4% cement, (b) 3% oil; 4% cement, (c) 0% oil; 8% cement, (d) 3% oil; 8% cement, (e) 0% oil; 12% cement, (f) 3% oil; 12% cement.

dramatically; by contrast, with the increase in cement content and curing age, the unconfined compressive strength had been enhanced remarkably; this confirms that cement can be served as an additive agent to solidify oil-contaminated soils.

(3) Direct shear strength tests show that with the increase of oil content, both cohesion and internal friction angle of solidified soil turned to decrease, while, with increasing the curing age and cement content, both properties had been improved gradually.

- (4) The results of SEM show that the hydration products of cement stabilized oil-contaminated soil decreased with the increment in oil content, and its microstructure got deteriorated. However, with the increasing dose of cement, the hydration products increased and the soil structure had been enhanced significantly.

Data Availability

The experimental data presented in this study can be referred to the corresponding author (Jianjun Ma), who would share these data once required.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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