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Nanoclay Stabilization of Crude Oil Contaminated Soils

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ABSTRACT: Human activities can pollute not only air and water but also soil. One of the most important types of soil pollution is the contamination with oil and its products. Such contaminations are known to affect the geotechnical characteristics of soils as well as their physicochemical properties. These effects are required to be considered and remedied to ensure the safety of structures built on the contaminated soils. Stabilization of contaminated soils with cost-effective stabilizers seems to be a simple and quick approach to limit the impact of contamination. With the development of nanotechnology and its widespread use in all fields of engineering, the possible benefits of this technology for geotechnical engineering have received increasing attention. Nano montmorillonite is a nano-product obtained from clay minerals and regarded as a natural and environment-friendly stabilization material. In this research, the effects of crude oil contamination on the strength parameters of soil derived from Isfahan oil refinery site were first considered using direct shear tests. The efficiency of nanoclay stabilizer to treat the contaminated soil was then evaluated considering the nanoclay content and treatment time. The tests results showed that the stabilized samples with 2.25% nanoclay additive (as optimum content) had a significant strength recovery. Thus, the treated soil cohesion and friction angle enhanced respectively to 1.7 times and 1.2 times those of untreated soil.

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1. INTRODUCTION

Soil stabilization has been commonly used in several studies to improve soil physical properties such as shear strength, bearing capacity, and frost heave susceptibility [1–6]. Nowadays, soil and groundwater contamination with toxic pollutants is a worldwide environmental problem [7]. Oil contamination, mainly occurs during transportation, and leakage from storage tanks or pipelines. Subsequently, soils and groundwater may be contaminated [8]. So far, various methods have been used to eliminate soil contamination. The easiest way to reach a result is "Solidification/Stabilization" [9]. Solidification/Stabilization refers to a group of waste remediation techniques in which contaminated waste is mixed with certain materials in order to reduce, physically or chemically, the amount of contamination that washes off into the environment or make the contaminated waste clean enough to be landfilled or to be used as a constructional material. In waste solidification and stabilization research, the goal is often to obtain a slightly altered waste that that can be safely disposed of in landfills without much environmental concern [10, 11, 12]. The choice of solidification/stabilization solution should not only be cost effective but also environmentally responsible in the sense of not causing secondary environmental pollution.

With resent rapid developments in the field of nanotechnology [13], the effect of nanoparticles like nanoclay on solidification/stabilization has received increasing attention. Clay minerals are divided into four categories: kaolinite, illite, montmorillonite, and chlorite. Montmorillonite refers to a group of clay minerals that consists of a sheet of gibbsite sandwiched between two sheets of silica, which have van der Waals bonds formed in the presence of water and cations. The main source of montmorillonite in nature is bentonite soil [14]. Montmorillonite nanoclay can be described as 2:1 layered smectite with a plate structure [15] and the chemical formula (NaCa)0.33(AlMg)2(Si4O10)(OH)2.nH2O [16]. These nanoparticles are produced by the treatment of montmorillonite to remove naturally occurring impurities such as quartz, kaolinite, illite, hematite, calcite, and feldspar [17]. So far, only a few studies have investigated the effect of nanoclay on different properties of contaminated soils. Zhang studied the soil nanoparticles and their effects on geotechnical properties [18]. You et al. reported that nanoclay could be used to improve the mechanical properties of asphalt mixtures and showed that nanoclay improves the dynamic shear modulus and viscosity of bitumen [19]. Majeed and Taha studied the effect of three different types of nanomaterials (nano CuO, nano MgO, and nanoclay) on the geotechnical properties of a soft soil from Penang region. This

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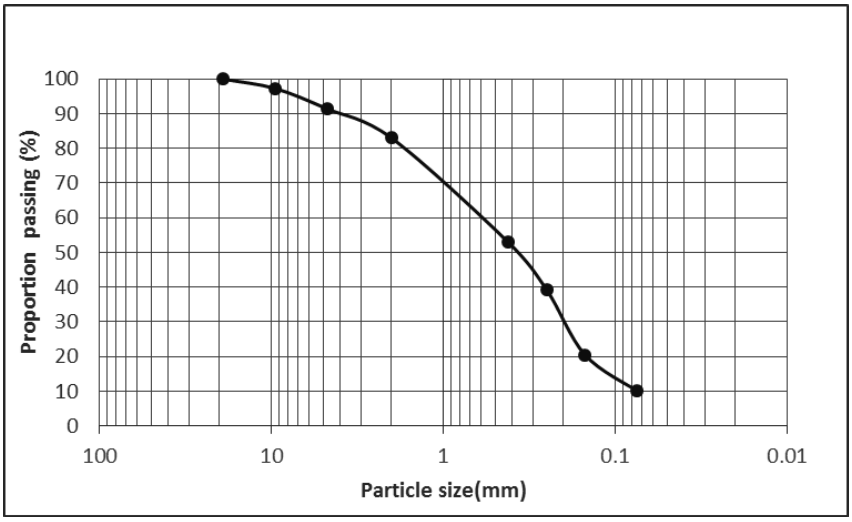

Fig. 1. Particle Size Distribution of the Testing Soil

study found that the addition of nanoparticles increased the maximum dry weight, optimum moisture, and unconfined compressive strength of the soil and reduced its plastic limit, liquid limit, plasticity index, and shrinkage limit [20]. Neethu and Remya investigated the behavior of several soils mixed with nanoclay. The results obtained for two types of sedimentary soil and a kaolinite clay showed that nanoclay additive increased the plastic and liquid limits, permeability coefficient, and unconfined compressive strength. With increasing the amount of nanoclay, the coefficient of consolidation of both types of soil first decreased but began to increase once nanoclay quantity went beyond an optimal value [21]. Nikookar et al. studied the properties of nanoclay-stabilized silty soils. The confined compressive strength tests conducted in this study showed the soil strength enhancement due to adding up to 1.5% nanoclay for high-plastic silty soils and up to 1% for low-plastic silty soils. It was found that adding more nanoclay would not be cost-effective as its effect on soil strength would be insignificant [22]. Taha and Taha conducted some experiments to investigate the effect of nanomaterials on soft soil stabilization. This research found a generally significant improvement in maximum dry density, plastic index, and uniaxial compressive strength, but clarified that the results also depend on the type of nanomaterial [23].

The effects of hydrocarbon pollutants on the geotechnical properties of soils have been previously reported by Lekmine. In this study, several experiments were carried out to investigate the mechanical properties of various soils contaminated with hydrocarbon oils. The pollutants used in this study were fuel oil and kerosene. The results showed that oil contamination reduced the soil strength, which decreased with time elapse [24]. Al Sand and Ismael conducted a research program to measure the amount of oil pollution from the second Gulf War in the sand surrounding the lakes created to accumulate floating oil patches. To determine the effect of oil pollution on the engineering characteristics of Kuwaiti beach sands contaminated with oil. The results of these experiments showed that the oil pollution up to 4 percent enhanced the soil compressibility while at higher contamination the compressibility decreased [25].

Khamehchiyan et al. have made an extensive research on the oil polluted soils from the Persian Gulf coasts. It was found that, in general, oil contamination reduces the permeability and strength in all specimens [26]. Abousnina et al. suggested the use of oil-contaminated sand as a road construction material which seems to be a cost-effective solution to minimize adverse environmental impacts. In this context, the effect of oil pollution on mechanical properties of sand was investigated. The results showed that with an increase in contamination up to 1%, the soil shear strength increases while it experiences some reduction at greater percentages of contamination [27].

The available literature on soils contaminated with crude oil or other petroleum derivatives have been mostly focused on geotechnical engineering after the contamination. The choice of nanoclay is also environmentally responsible in the sense that it does not cause any secondary environmental pollution. Moreover, due to the previous studies, the geotechnical properties of the soil have been improved and have been considered as stabilizing. Also, none of the previous studies considered the effect of nanoclay on the geotechnical properties of the contaminated soil.

This paper reports the results of a laboratory study in which the strength parameters of soil specimens exposed to different amounts of crude oil contamination were investigated at various conditions. The effects of pollution time elapse, various nanoclay additives, and treatment time were among several variables considered in this research. The results show a significant improvement in the strength properties of the soil specimens following the nanoclay stabilization.

2. MATERIALS AND METHODS

2.1. Materials

The soil samples used in this research were collected from the Isfahan refinery site from a depth of 0-3 meters. In order to identify the soil, the size distribution of soil particles was

Table 1. Soil Specifications

Gradation Coefficients	Cu=8, & Cc=0.89
Liquid Limit	31
Plastic limit	20
Plastic Index	11
Specific Gravity (Gs)	2.8
Soil Classification (USCS)	SP-SC

Table 3. Results of chemical analysis of nanoclay

%	Symbol	Number
0.98	Na2O	1
3.29	MgO	2
19.6	Al2O3	3
50.95	SiO2	4
0.86	K2O	5
1.97	CaO	6
0.62	TiO2	7
5.62	Fe2O3	8
15.45	LOI	9

first determined according to ASTM D 422-63. As seen from Fig.1, the fine fraction of the soil is about 10%. Thus both plastic properties and gradation coefficients are required to proceed the soil classification. The soil specifications are presented in Table 1. Also, the specifications of crude oil used in Isfahan refinery, as the source of soil pollution, are summarized in Table 2.

2.2. Nanoclay

The nanoclay used in this study is 99% pure montmorillonite purchased from Sigma-Aldrich Inc. (Germany). The physical and mechanical properties of this montmorillonite are provided in Table 2 and its chemical analysis are presented in Table 3.

2.3. Methods

According to the field reports, the range of crude oil pollution at the Isfahan Refinery site is estimated to be between 4% and 8%. To better control the soil conditions, clean soil samples derived from the refinery protected site were contaminated with crude oil at 4 and 8% of the soil dry weight. The required amount of soil for direct shear tests was also mixed with desired amount of crude oil. It should be noted that at least 2 days should elapse to allow soil particles to absorb the added oil (transmitted from free-flow phase to absorbed phase). To achieve the desired state, the polluted samples were stored within sealed two layer plastic bags [28].

Table 2. Physical and mechanical characteristics of clay montmorillonite

Mineral type	montmorillonite
Particle size	1.18 nm
PH	7.3-7.6
electrical conductivity	25 MV
Ion exchange coefficient	48 (meq/100 gr)
Empty gap between particles	60 A°
Color	yellow
Moisture	1-2 %

As nanomaterials consist of ultrafine particles (10-9mm), the method of mixing them with soil is very important for the end results. Naturally, the mixing procedure should be done such that the nanoparticles are uniformly dispersed across the soil and not clumped together. Research has shown that using greater amounts of nanoparticles may be associated with a higher chance of clumping, which can reduce the soil strength. In this research, the contaminated soil was first mixed with desired amount nanomaterials, the adopted amount of water was then added to the mixture. The contaminated soil and nanomaterials were mixed in two steps. First, the materials were mixed manually by dividing the soil into several fractions and mixing each fraction with a certain amount of nanomaterial. The soil fractions were then placed in a container and combined with a mixer. Finally, the appropriate amount of water was added to the mixture [29].

2.4. Testing Procedures

To determine the soil strength parameters of both polluted and nanoclay stabilized soil samples, direct shear tests were conducted on them based on the ASTM D3080-90 standard. The shear box used here has a circular cross section of 6.3cm diameter in which 2 cm height soil sample is placed and compacted to desired conditions [30]. After placing the desired vertical load on the sample, shear force is applied through a shear displacement-control system at a speed of 1 mm/min. For each set of direct shear tests, three similar samples were sheared separately under three normal stress levels of 0.5, 1, and 1.5 kg/cm^2. As seen, the shearing rate is sufficiently quick and the tests may be regarded as undrained tests if the samples water content approaches to a saturated state.

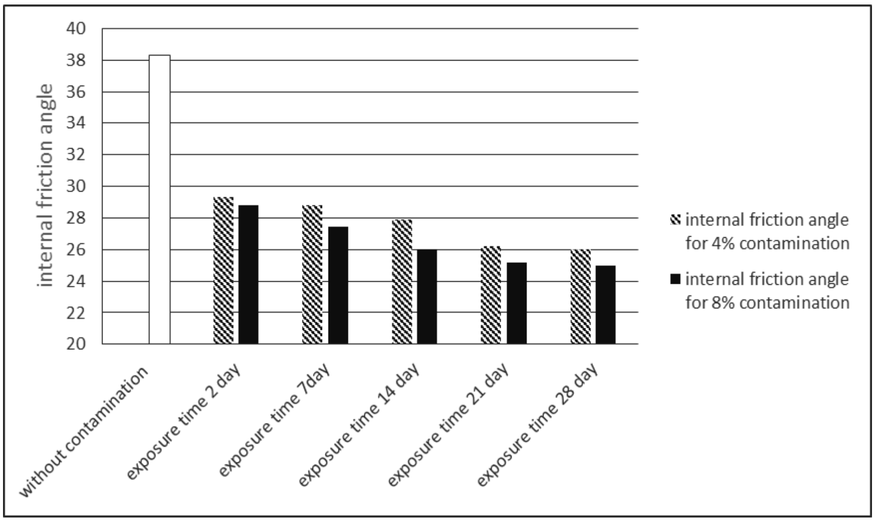

Fig. 2. Changes of soil friction angle versus exposure time

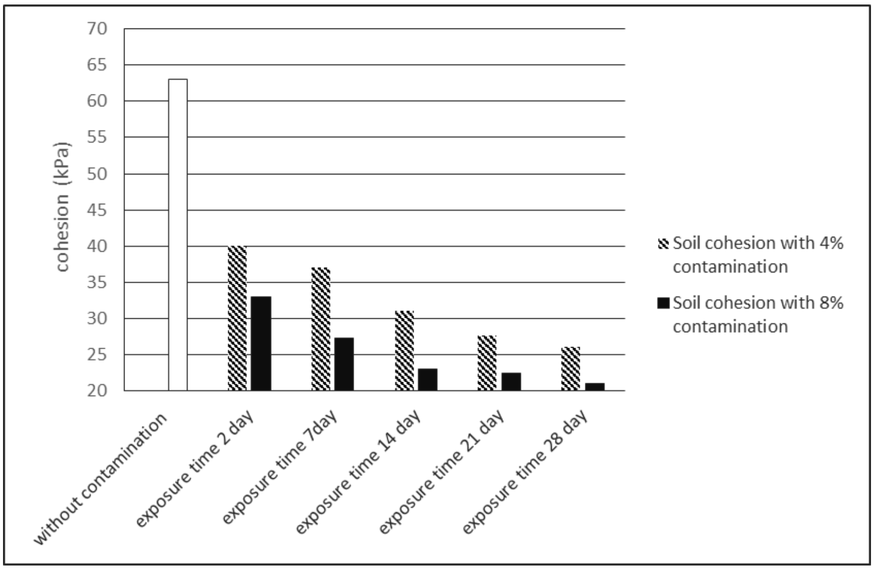

Fig. 3. Changes of soil cohesion versus exposure time

3. RESULTS AND DISCUSSION

3.1.Effect of crude oil on geotechnical properties

Figs. 2 and 3 show respectively the changes of soil cohesion and friction angle of the soil samples with 4% and 8% oil contamination at various exposure times. In these figures the clean soil strength parameters are also included for comparison.

The decreasing trend of the friction angle in Fig. 2 may be attributed to the reduction of effective friction between soil grains due to the lubricating effect of oil. The results show that with the prolongation of exposure to oil contaminant, the friction angle decreased at a slower pace. Eventually, oil absorption by the soil seems to become almost complete after about 21 days with no significant change in the friction angle at 28 days elapse from initial pollution.

As shown in Fig. 3, the test results indicate that the soil cohesion experiences more reduction at longer oil exposure time. This is because the presence of oil facilitates the agglomeration of fine soil particles, which leads to a decrease in the specific surface of the soil, and in turn, results in less bonding between soil particles and thus reduction in the cohesion. With time elapse of oil pollution and the prolongation of soil-oil interaction, further reduction occurs in the soil cohesion. As can be seen, this cohesion reduction is greater in the samples with 8% contamination in comparison to that of 4% polluted samples.

The changes in the soil cohesion and friction angle with the amount of contamination are illustrated in Figs. 4 and 5. It can be seen that with 2% contamination, the soil actually shows a slightly increased cohesion, which may be due to oil lubricant effect between soil particles, while the presence

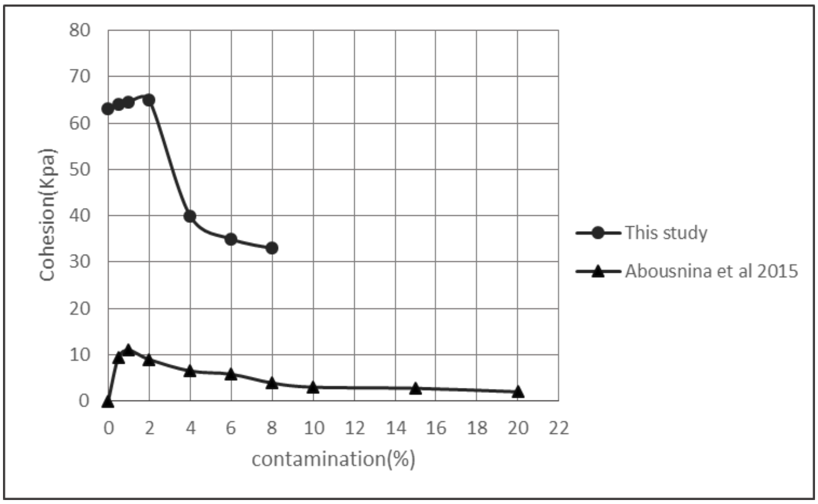

Fig. 4. Changes of soil cohesion versus contamination content

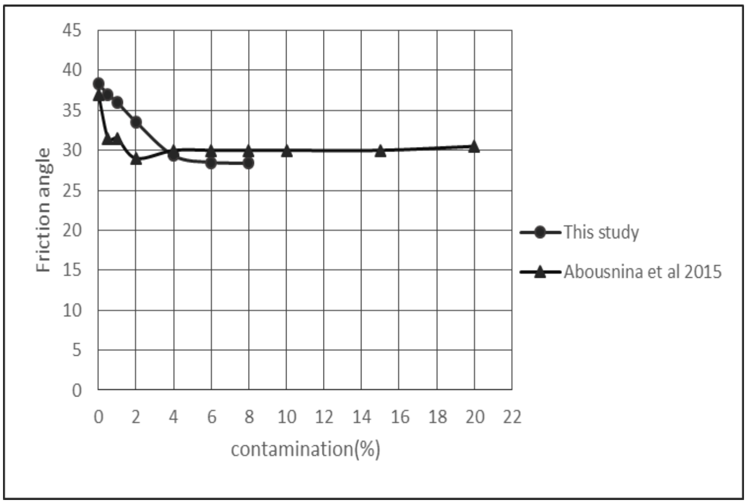

Fig. 5. Changes of soil friction angle versus contamination content

of oil contamination at any amount reduces the soil friction angle. Nevertheless, the rate of friction angle reduction decreases as the amount of contamination increases. This is because of the oil effect on the interlocking of soil grains and the same slide potential at the presence of higher amounts of oil contamination i.e. higher amounts of oil does not change this role significantly. These results are consistent with the findings of Abousnina et al. [27].

Such results may also be explained by the viscosity and inherent cohesion of the crude oil products. In fact, the reduction in the cohesion of contaminated soils at higher contamination contents may be attributed to the reality that the soil particles seem to be fully coated with crude oil leading to a reduction in the soil grains interlocking resistance. This observation is also in agreement with Seed et al. (1961) who state the reduction in cohesion is due to forming thicker films of oil around the particles of soil, and by increasing the content of crude oil, the chance of inter particle slippage would also increase which in turn results in a decrease in the shear strength. This process may be seen in Fig. 4 and 5 as the soil particles are coated with oil at higher levels of oil contamination. Hence, after a slight increase in the soil cohesion at 2% contamination, the cohesion as well as friction angle followed a reduction trend at higher contamination contents. Similar trend has been reported in previous studies as those presented in Figs. 4 and 5 [31].

3.2. Results of direct shear tests on nanoclay-treated contaminated soils

The test specimens were prepared at optimum compaction and water content. The optimum nanoclay content was determined for two-day treatment of the soil with two days of exposure to 8% crude oil contamination. In Figs. 6, 7, and 8, the results of shear tests on the specimens with different

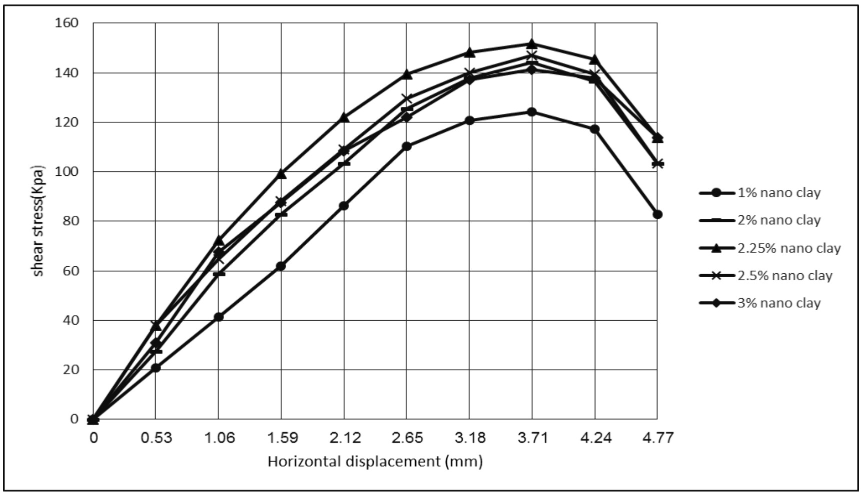

Fig. 6. Shear stress versus shear displacement at the vertical stress of 50kPa

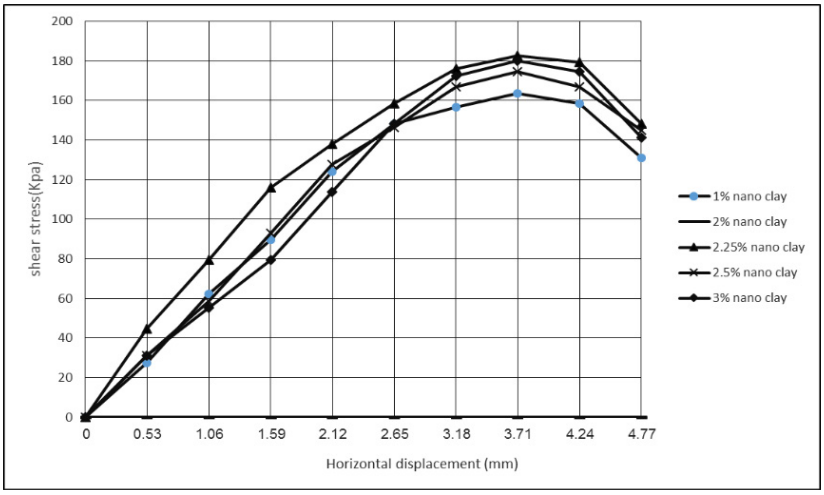

Fig. 7. Shear stress versus shear displacement at the vertical stress of 100kPa

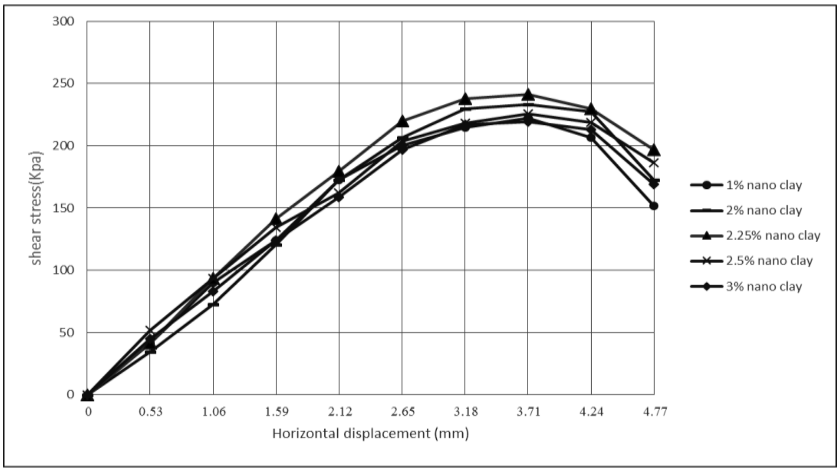

Fig. 8. Shear stress versus shear displacement at the vertical stress of 150kPa

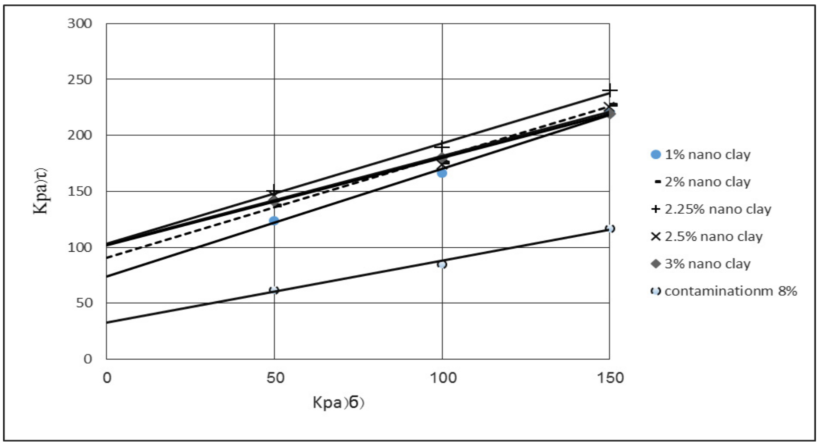

Fig. 9. Mohr-Coulomb diagram of the specimens with different nanoclay contents (contamination content=8%, exposure time=2 days, treatment time=2 days)

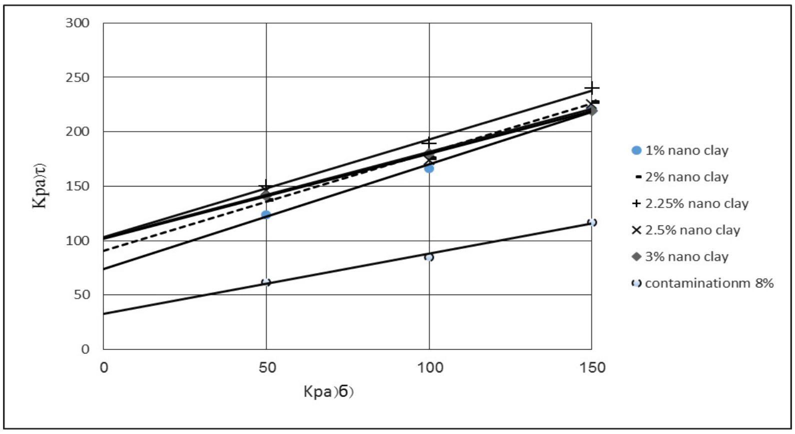

Fig. 10. Cohesion versus nanoclay content for contaminated soil specimens

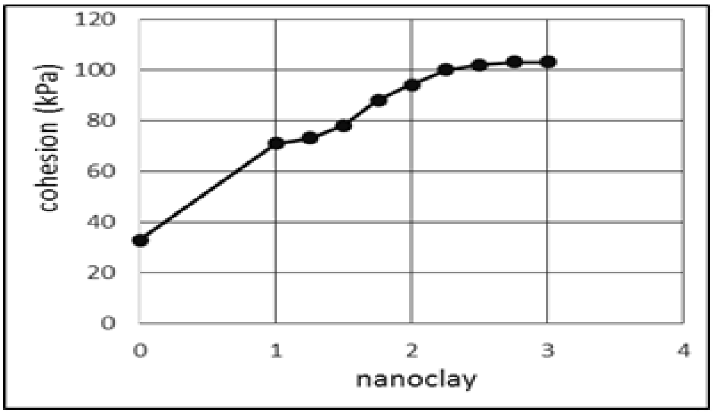

Fig. 11. Friction angle versus nanoclay content for contaminated soil specimens

nanoclay contents are presented as shear stress-displacement diagrams for three normal stress levels. Fig. 9 shows the shear strength of the soil exposed to 8% contamination for two days following two days of nanoclay treatment.

Based on the test results presented in Figs. 6-9 for samples with 8% contamination, adding nanoclay significantly improves the strength of the contaminated soil. The highest strength enhancement has been achieved for specimens treated with 2.25% nanoclay. Thus, the 2.25% nanoclay additive may be considered as an optimum nanoclay content regarding the soil strength improvement.

Figs. 10 and 11 show the trends of change in the cohesion, friction angle, and shear strength of the soil versus the amount of nanoclay additive. As seen, the cohesion reaches

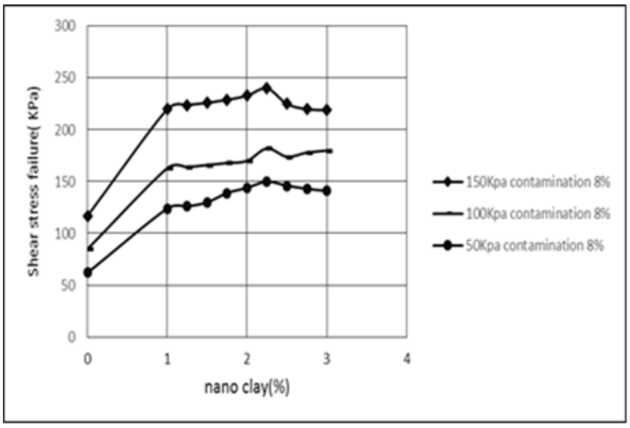

Fig. 12. Shear strength versus nanoclay contents for contaminated soil specimens

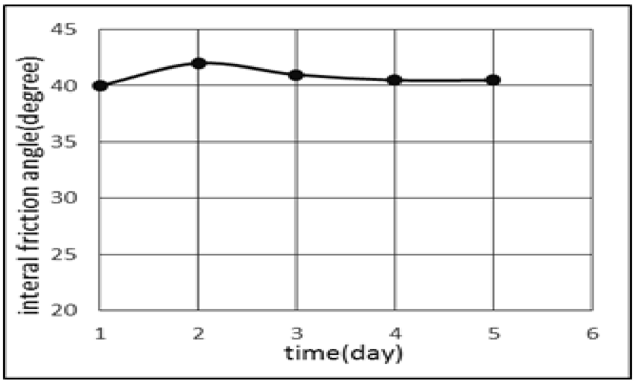

Fig. 13. Friction angle of the soil specimens treated with the optimal nanoclay content with different treatment times

its maximum value at 3% nanoclay content, but the amount of cohesion gain at nanoclay contents higher than 2.25% nanoclay is relatively small and negligible. According to Fig. 11, the highest friction angle is achieved by adding 1% nanoclay, and adding more nanoclay decreases this angle. To explain these results, it may be concluded that the presence of high nanoclay content reduces the role of interlocking effect between soil particles. In Fig. 12, it can be seen that the soil with 2.25% nanoclay exhibits the highest shear strength at all three levels of vertical stress i.e. 50, 100 and 150kPa.

Based on the aforementioned results, the following may be concluded:

- The addition of nanoclay increases the cohesion of the crude oil-contaminated soils, but this effect becomes insignificant at nanoclay content greater than 2.25%.
- The addition of nanoclay generally increases the friction angle of the crude oil-contaminated soils, but with nanoclay content greater than 1% a slight decrease is observed in the friction. The rate of decrease in the friction angles becomes significant at nanoclay content greater than 2.25%, apparently due to undermining the interlocking of the soil grains.

3.3. Effect of treatment time on the strength of contaminated soils

A series of direct shear tests were conducted to determine the preferable treatment time for nanoclay-stabilization of the soil contaminated with crude oil for two days and also to evaluate the behavior of this soil after different treatment times. For this purpose, several specimens with 2.25% nanoclay content, which gives the optimum results in terms of shear strength, were constructed and subjected to direct shear test after 1, 2, 3, 4 and 5 days of treatment. During this time, the specimens were placed inside thick multilayered bags to maintain their moisture.

As shown in Figs. 13 and 14, the highest strength for the soil with 8% contamination was obtained at the second day of treatment. It can be seen that there are very small changes in friction angle and cohesion after the second day of treatment. Such a result may be attributed to measurement errors as well as slight changes in the samples moisture content.

3.4. Effect of exposure time on the duration of chemical reactions

Figs. 15 and 16 show the effect of exposure time on the duration of chemical reactions between soil, water, and nanoclay at 8% contamination.

The results presented in Figs. 15 and 16 show that as the exposure time increases, the treatment time remains unchanged. Such results may be considered as an indication that chemical reactions between soil, water and nanoclay do not change with the prolongation of treatment time.

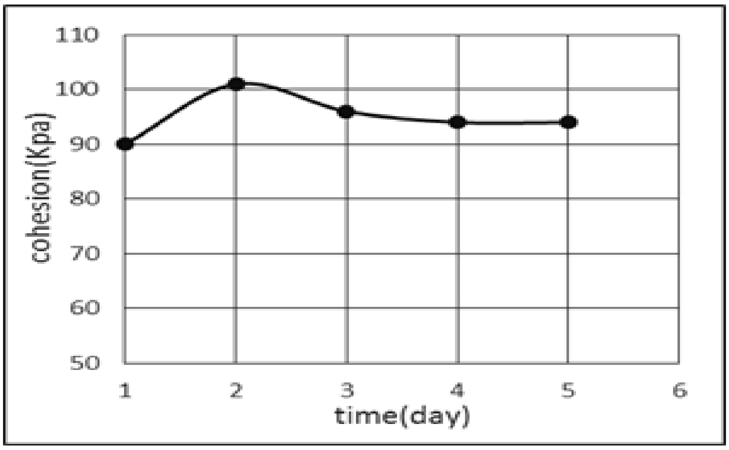

Fig. 14. Cohesion of the soil specimens treated with the optimal nanoclay content with different treatment times

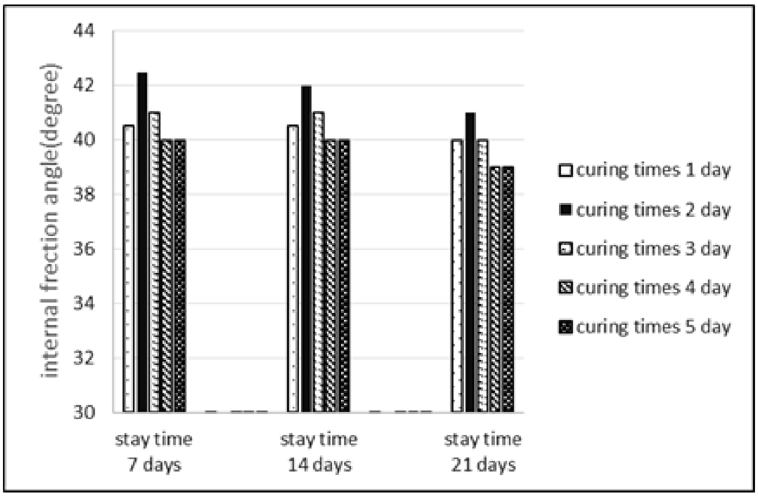

Fig. 15. Friction angle of the soil specimens at different exposure times and treatment times

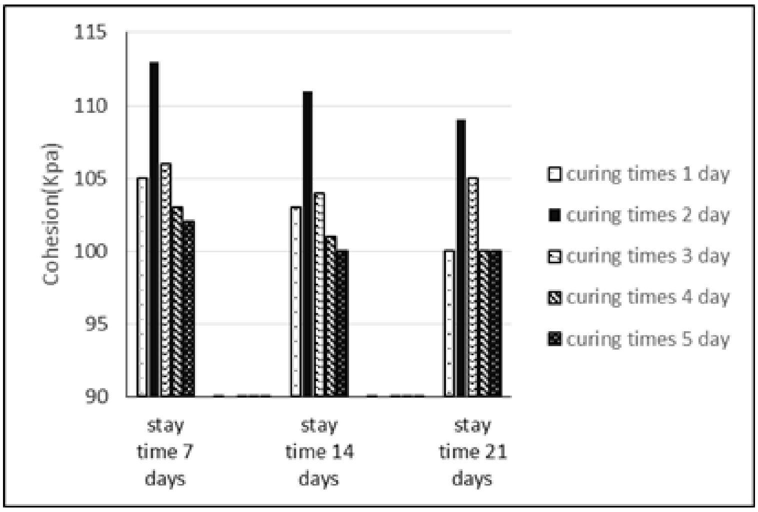

Fig. 16. Cohesion the soil specimens at different exposure times and treatment times

4- CONCLUSION

This study investigated the effect of nanoclay additive at 1, 1.25, 1.5, 1.75, 2, 2.5, 2.5, 2.75 and 3% by dry weight of soil on the shear strength parameters of oil-contaminated soil specimens after 2 days as treatment time. The shear strength parameters of both untreated and treated soil samples were determined through standard direct shear tests. The results obtained from this investigation can be summarized as follows:

1. The addition of 2.25% nanoclay (the optimum amount) to the oil-contaminated soil results in 1.2 times improvement in the friction angle compared to the untreated soil.
2. The addition of 2.25% nanoclay (the optimum amount) to the oil-contaminated soil results in 1.7 times improvement in the cohesion compared to the untreated soil.
3. Nanoclay-treated soils present shear strength 2-2.5 times (depending normal stress level) of untreated samples, but increasing the nanoclay content beyond the optimum value leads to reduced shear strength.
4. The shear strength of the stabilized soil decreases slightly with the prolongation of contamination exposure time.

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