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Improvement of petroleum contaminated clayey soil by carbon fiber polymeric composite and diatomic soil

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Abstract

The overarching goal of this research is to use polymeric composites enriched with diatomic soil to improve petroleum-contaminated clayey soil. The effect of petroleum on the geotechnical properties of clayey soil was investigated in the laboratory in the first stage using uniaxial, direct shear, and permeability tests. The polymeric composite material was then created and mixed with the diatomic soil. The geotechnical properties of petroleum-contaminated clayey soil were studied using a polymeric composite material mixed with diatomic soil (PCD). Petroleum reduced the shear resistance, internal friction angle, and uniaxial resistance of the clay for contamination percentages ranging from 0% to 12%. Whereas 12 percent petroleum content causes the greatest changes in soil mechanical resistance. According to the results of the direct shear test, adding 5.5 percent PCD increases the shear resistance of the base material and contaminated base material to average values of 32 and 48 percent, respectively. Furthermore, the results of the petroleum permeability test show that adding 5.5 percent PCD reduces soil permeability. The results show that the improved clay by PCD can be used as a liner for the base of petroleum reservoirs.

key words

In-Situ Synthesis

Nanofluid

Cerium Oxide

Enhance Oil Recovery

Micromodel

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1.Introduction

Petroleum is one of most important fuel sources all over the world. The soil contaminated by the petroleum not only pollutes the underground water, but also affects the bearing capacity of the oil facilities and poses potential causes of superstructures instabilities. Several investigations have been accomplished on the influence of petroleum and its derivations on the mechanical [1-5] and electrochemical [6] behavior of the soils. Some of the researchers reported a significant reduction in the bearing capacity of shallow foundations and friction angle of the contaminated soils by oil. Al-Sanad et al. [2] showed a significant reduction in the internal friction angle and an increase in the compressibility of the soil. They also indicated that increasing the amount of the oil causes the decrease in shear strength and permeability of poorly graded sandy soil. A series of the tests in the south of Iran [7] were conducted and it was found that oil contamination causes a decrease in strength as well as the reduction in the consistency limits and permeability of sandy and clayey soils because of non-polarity of oil and its products. Some of the studies [8] showed that contaminated clay behaves like cohesionless soil and this is attributed to the formation of flocs due to the spherical agglomeration of oil coated clay particles. Safihian et al. [9] also observed that adding diesel reduced internal friction angle and unconfined compressive strength of the illite clay.

Many sites contaminated with organic materials have been successfully stabilized nowadays [10]. Although the application of cement stabilization of contaminated soil is very popular, it has an adverse effect on the environment since the manufacture of cement causes 5–7% of global carbon dioxide emissions [11]. Therefore, different investigations have been carried out on the application of nano materials for soil stabilization [12-15]. It has been proved that adding carbon nanotube to kaolinite caused a lower soil strength and hydraulic conductivity and a higher compressibility [16]. The effect of three types of nanomaterials (i.e., nano calcium, nano magnesium and nano clay) on geotechnical characteristics of soft soils was also reviewed [13]. The results expressed that Atterberg limits decreased by adding nano materials to the samples while their compression strength increased. Taha and Taha [17] accomplished numerous tests to determine the behavior of the clay treated with nano-Al_2O_3. They concluded that both values of expansive and shrinkage strains were decreased. In another investigation [18] it was concluded that mixture of the cement/ash and nano-Al_2O_3 reduces the maximum dry unit weight and increases the optimum moisture content. Adding nano-clay [19] to the soil increases the liquid and plastic limits as well as the shear strength parameters of treated specimens. A series of the tests on the clayey soils by adding nano-SiO_2 [20] were carried out and it was found that the addition of nano-SiO_2 leads to a reduction in the swelling index of clay. The effect of adding nano-clay and glass fiber on clayey soil with low liquid limit was accomplished by Changizi and Hadad [21]. The results showed that angle of internal friction and cohesion of clay increased. In addition, the fundamental properties of soil mixed with nanoparticles was reviewed by Krishnan and Shukla [22].

Therefore, it is necessary to stabilize the soil where the strength is reduced because of the presence of the petroleum. Clay soils stabilized with conventional additives have been widely used as infrastructure materials for roads, embankments, retaining walls, pavements, and other constructions in recent years [23]. The combination effect of diatomic soil and polymeric composite materials on the mechanical properties of the contaminated clay is not clear. It is worth noting that diatomite is a fine sedimentary rock of biogenetic origin and mainly consists of a mix of sand, fines (silt and/or clay), fossilized diatoms [24], amorphous hydrous silica ($SiO_2 \cdot nH_2O$) and widely apply as sorbents of petroleum compounds [25,26]. The aim of this study is the improvement of geotechnical properties of petroleum contaminated clay by a mixture of the diatomic soil and polymeric composite produced using epoxy resin and carbon nanofiber. Using this method, in addition to improve the petroleum contaminated soil, a compacted clay liner can be prepared which is usable under oily material facilities. The property of these liners is that their strength does not decrease during contamination and petroleum contamination may be kept by the liner. According to Falamaki et al. [27] in order to decrease the costs, this compacted clay liner can be multilayer, i.e., during the construction a thin layer of the liner is mixed with the additive materials.

2.Materials and Methods

2.1 Soil, petroleum and polymeric composite materials

A clayey soil sample was obtained from Shiraz

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Table 1 Index and physical properties of the used soil

Physical Property	Value
Unified classification	CL
Liquid Limit (LL) (%)	32
Plastic limit (%)	22
Plasticity index (%)	10
Specific gravity	2.66
Max. dry density (kN/m3))	16.68
Optimum moisture content (%)	14

City, located in the southwest of Iran. According to the unified soil classification system, the used soil is classified as CL (clay with low plasticity). The soil index and physical properties were determined experimentally and are shown in Table 1.

Petroleum for this study was provided by Sarvestan Petroleum Refinery Company, which is located in the southwest of Fars province, Iran. Thermal-chemical processes were used to create the composite in this study. The composite was then supplemented with diatomic soil as an additive. The main components of this PCD material are epoxy resin and carbon nanofiber.

The resin used was manufactured by Mandegar Baspar Co. in Iran. This material is RL510 with hardener code H513 and is made with epoxy-based phenol A and polyamine concrete hardener. In the structure of this epoxy resin, a reactive diluent type is used, which improves both electrical isolation properties as well as mechanical characteristics and strike protection abilities. This product is resistant to thermal shocks and does not crack or fracture. It is also suitable for use in harsh environmental conditions due to its high chemical strength. This polymeric is used in liquid form and is widely available and reasonably priced. Table 2 shows the physical, facial, and mechanical properties of this type of polymeric.

Carbon nanofiber is created artificially using chemical vapor deposition (CVD), and its proper-

Table 2 Physical, facial and mechanical properties of epoxy resin

Properties	Result
Visual Appearance	Clear Liquid
ratio of resin/hardener (by weight)	100:13
Dielectric Constant ASTM D150	3.91
Density @ 25°C (77°F), g/ml	1.1
Tensile Strength (MPa)	85
Tensile Elongation (%)	4.9
Compressive Strength (MPa)	289
Flexural Strength (MPa)	134
Flexural Strength (MPa)	3309
Tensile Modulus (MPa)	3240

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Table 3 Chemical properties of carbon nanofiber

Property	Value
Purity	99%
Diameter (nm)	150-300
Length (micrometer)	>10
Standard deviation	1.15±0.2
Percentage of OH group factor	3.51
Percentage of COOH Factor Group	2.82

ties are shown in Table 3.

The diatomic soil was a white powder prepared by the Vaezzade Chemical Company in Iran, and it is related to salty water in northern Iran. It is commercially and economically available. As an additive, this material with varying weight percentages will be added to the composite.

2.1.1 Polymeric composite production process

The thermal method was used in this investigation for PCD production. To make 1 g of this composite, 1 g of the polymeric (epoxy resin) was poured into a 50 ml beaker with 10 ml dimethyl formamide solvent and left for about 2 hours to cause the predicted swelling. The mixture was then supplemented with nanoparticles (carbon nanofiber) (1 percent by weight of epoxy resin). The suspension is then poured into a 250 ml flask, which contains a magnet for permanent solution mixing. It was heated and mixed simultaneously for about 5 hours at 80° by the thermal reflex system shown in Figs. 1a and 1b to achieve a simple solution. The reflex thermal system consists of a heater, an oil bath (paraffin), and a reflex pipe, and it is set to 110°. The flask containing the suspension was then placed in an oil bath and reflexed. After 20 minutes, the hardener with a weight percent related to resin based on table2 specifications was added to the suspension and the reflex system was restarted. The achieved solution was then added to 20-30 ml of deionized water, which was magnetically mixed, to precipitate, and finally the sediment was gathered and dried in an oven at 70°. Whereas the required composite for all tests is 200 g, the material weight ratios increase to speed up the test based on the weight ratios mentioned for producing 1 g of composite, and the required material was produced in a few steps. Figure 1d depicts a PCD sample made with carbon fiber and epoxy resin. Fig. 1f depicts a sample of PCD made from carbon fiber and epoxy resin that was powdered and passed through sieve No. 200 with 5.5 percent dry base material, 2.5 percent composite, and 3 percent diatomic soil used for adding to soil samples. The 5.5 percent is an approximate optimized amount obtained experimentally through trial and error. The results of an experimental investigation into the optimal amount determination of composite material revealed that adding more than 3% diatomic soil causes cohesion reduction, collapsing, and disintegrating of the sample; and adding more than 2.5 percent produced composite from fiber and resin causes an increase in strength. However, in order to reduce project costs and economic issues, a PCD amount of 5.5 percent is considered optimal, consisting of 3 percent diatomic soil and 2.5 percent composite produced from resin and nanofiber.

2.2 Sample Preparation and Test Procedures

The soil was mixed with 0, 4, 8, and 12 percent petroleum by weight of dry soil for sample preparation. The samples were then isolated and cured for 24 hours for chemical and moisture equilibrium using the optimum moisture (14 percent). The effect of petroleum on the geotechnical properties of the contaminated soil was investigated using uniaxial compression tests, direct shear tests, and consolidation tests. To examine the effect of PCD, 5.5 percent PCD was added at the end of sample preparation to some of the non-contaminated and contaminated samples. These samples were also subjected to a falling head permeability test. According to Table 4, all tests were performed in accordance with ASTM standards.

Table 5 shows the experimental plan for this study›s goal. A strain controlled direct shear test (ASTM-D3080) was performed to determine shear

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Fig. 1 PCD production process (a) carbon nanofiber (b) diatomic soil (c) Epoxy resin and hardener (d) thermal reflex system for polymeric composite (e) adding achieved suspension to deionized water (f) produced PCD sample.

strength, cohesion, and internal friction angle. The direct shear test mold was 1010 square centimeters in size. The loading speed was one millimeter per minute, and the normal stresses were 50, 100, and 150 kPa. These tests were performed to investigate the effect of petroleum percent on shear strength, as shown in the related rows 1–4. (table3-4). Furthermore, tests 5 and 6 investigated the effect of PCD on these parameters in two conditions: without petroleum contamination and with approximately 12% petroleum contamination. The effect of petroleum amount on clayey soil was investigated by adding 0%, 4%, 8%, and 12% weight percent of base material to the soil (related rows number 1 to 4, Table 4). The unconfined compressive test with controlled strain was carried out (ASTM-D2166). The samples were prepared in cylindrical molds 4 cm in diameter and 8 cm in height, and the tests were carried out under normal load with controlled strain. The falling head permeability test (ASTM-D5856) was performed to determine the hydraulic conductivity of the base material without additives and with 5.5 percent composite in two statues of petroleum or water. The soil samples for the permeability test were prepared for each composition based on the optimal moisture percent and maximum dry density. To avoid sample disturbance and reduce permeability equipment error, the composition was directly compressed in the permeability falling head cell

Table 4 Evaluated parameters of each test

Test	Standard	Evaluated parameters
Shear test	D3080 – ASTM	Internal friction angle (φ); cohesion (c), shear strength (τf)
UCS test	D2166 –ASTM	Unconfined shear strength (Cu)
Falling head test	ASTM – D5856	Permeability coefficient (k)

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in three layers with similar masses and tested using the standard proctor compaction method. Following sample preparation, a falling head permeability test was performed (ASTM-D5856). The test began with optimized moisture (non-saturated) and progressed to fully saturation and constant permeability ratio. Samples of soil were saturated from the bottom up. Following the completion of the test, the cell, piezometer, and tube were evacuated, and petroleum was added to the cell with no change in the compacted sample. After bleeding the piezometer and tube connected to the cell, the test was restarted.

er, it increases cohesion. This reduction appears to have occurred as a result of petroleum as a lubricant, allowing the particles to slip and slide against each other, but petroleum products reduce friction between the particles better, reducing the space between them and also the frictional angle. Ur-Rehman et al. [8] expressed that the reason of decreasing in shear strength and internal friction angle is due to petroleum existence adjacent to clay particles, consequently, the cationic exchange capacity and cohesion between the particles decreases. Kererat [29] showed that bearing capacity for the gasoline-contaminated silty sand decreased by 30-52% depending on the concentration of the oil. Moreover, the internal friction angle measured by direct shear test decreased with the gasoline content and the cohesion increased with increasing oil content.

Table 5 Experimental program

No.	Test composition	Type of test			
		Falling head test		UCS test	Direct Shear test
		Petroleum	Water		
1	Soil	+	+	+	+
2	%4Pet. + Soil			+	+
3	%8Pet. + Soil			+	+
4	%12Pet. + Soil			+	+
5	5/5% PCD + Soil	+	+	+	+
6	12%Pet. + 5/5% PCD + Soil			+	+

3 Results and Discussion

3.1 Effect of petroleum contamination on shear strength of base material

Fig. 2 depicts the shear stress-horizontal displacement curves of contaminated soil. These findings suggest that adding petroleum to the soil reduces shear strength. In addition, Fig. 3 depicts the Mohr-Coulomb failure envelope for base material and contaminated soil with varying percentages of contamination. According to this figure, the shear strength of contaminated samples under each stress is less than the base material›s strength. These changes are more pronounced under higher-than-normal stresses.

Abdunaser [28] concluded that the oil present between the soil particles allows the particles to move easily in relation to each other, resulting in a reduction in the angle of internal friction and, in some cases, soil adhesion. Fig. 4 shows the effect of petroleum contamination on internal friction angle and cohesion of the base soil. Increasing in petroleum concentration in the clay decreases the internal friction angle from 21^{o} to 5^{o}, howev-

3.2 Simultaneous effect of petroleum and PCD on shear strength

The results showed that adding 12% petroleum content results in the greatest shear strength changes. As a result, the simultaneous effect of petroleum and PCD on shear strength of base material was investigated at 0% and 12% petroleum concentration. In Fig. 5, the shear stress-displacement behavior of a base material with and without PCD under similar normal stress is compared. Adding 5.5 percent PCD to the base material increases shear strength by about 21 to 53 percent under normal stress of 50 to 150 kPa. In fact, Azzam [12] reported similar findings regarding the effect of nanomaterials on the shear strength of clayey soils. The filling of soil pores by nano particles, as well as the effect of PCD, increase soil shear strength.

Furthermore, Fig. 6 depicts the simultaneous

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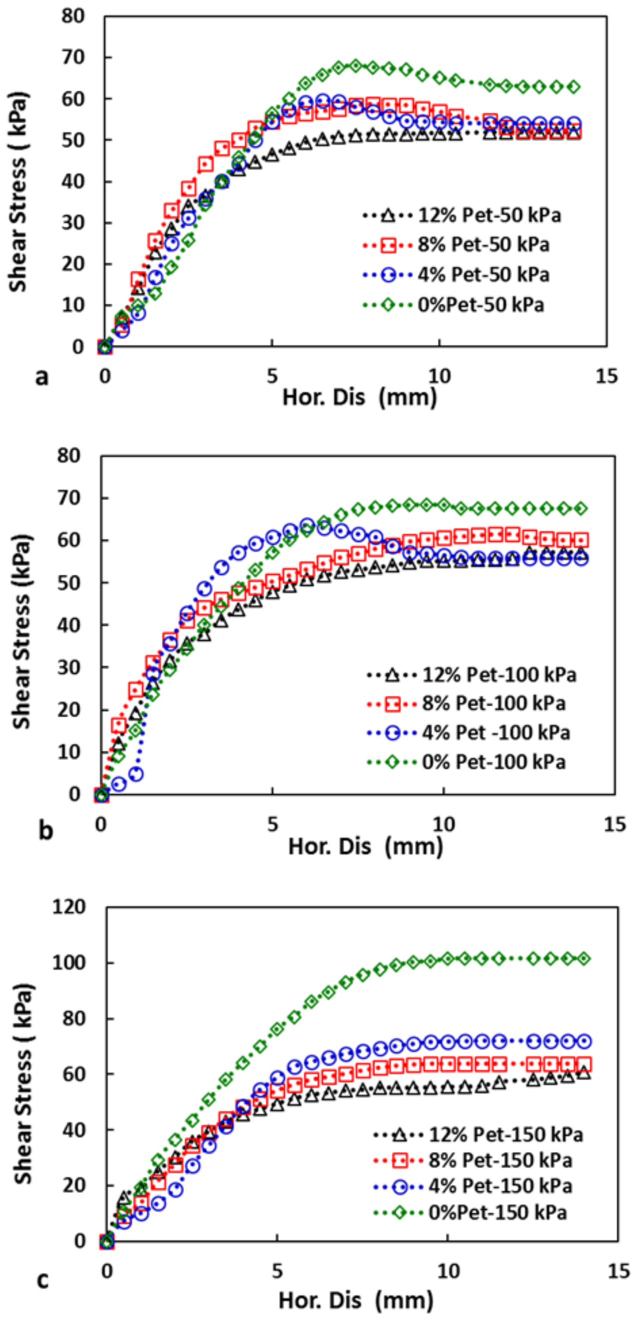

Fig. 2 Shear stress versus horizontal displacement from direct shear test

effect of petroleum and PCD on direct shear test results. The results show that adding PCD to samples containing 12% petroleum increases shear strength by about 11, 61, and 74% for normal stresses of 50, 100, and 150 kPa, respectively.

When the results of Figs. 5 and 6 are compared, it is discovered that adding 5.5 percent PCD is more effective in increasing the strength of the sample with 12 percent contamination. The reason for this is the presence of a diatomic soil function in the PCD structure. Because of their porous structure, diatomic soils can accept nonpolar petroleum fluid. In other words, because this material absorbs petroleum, its effect on increasing shear strength is more pronounced in the sample with 12% contamination.

Fig. 7 presents the effect of PCD on Mohr-Coulomb failure envelope for base material and petroleum contaminated soil. Particle friction and internal friction angle decreases due to petroleum adding to the base material. By adding PCD to the soil, the shear strength increases (Fig. 7a), and internal friction angle increases about 4 degrees (21 to 25 degree). On the other hand, the cohesion significantly increases (38.8 kPa to 53 kPa). Based on Fig. 7b, PCD adding causes that decreasing

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$$\tau_{f(0\,\%\,pet)} = \sigma \tan(21^{o}) + 38.8$$
$$\tau_{f(4\,\%\,pet)} = \sigma \tan(10^{o}) + 42.7$$
$$\tau_{f(8\,\%\,petl)} = \sigma \tan(6.5^{o}) + 47.3$$
$$\tau_{f(12\,\%\,pet)} = \sigma \tan(5^{o}) + 47.6$$

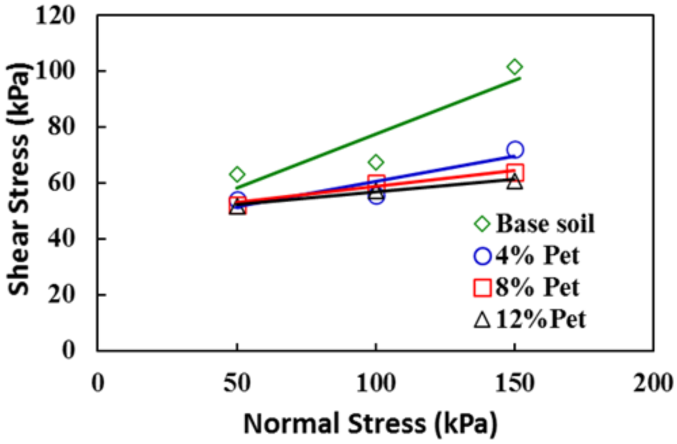

Fig. 3 Mohr-Coulomb failure criterion of different samples

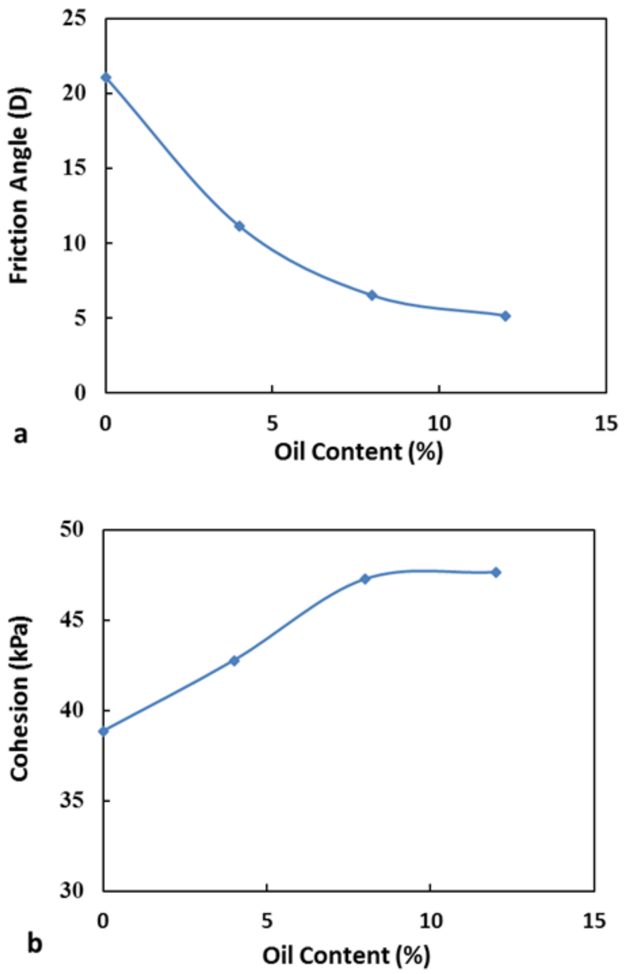

Fig. 4 Internal friction angle and cohesion changes with petroleum percentage

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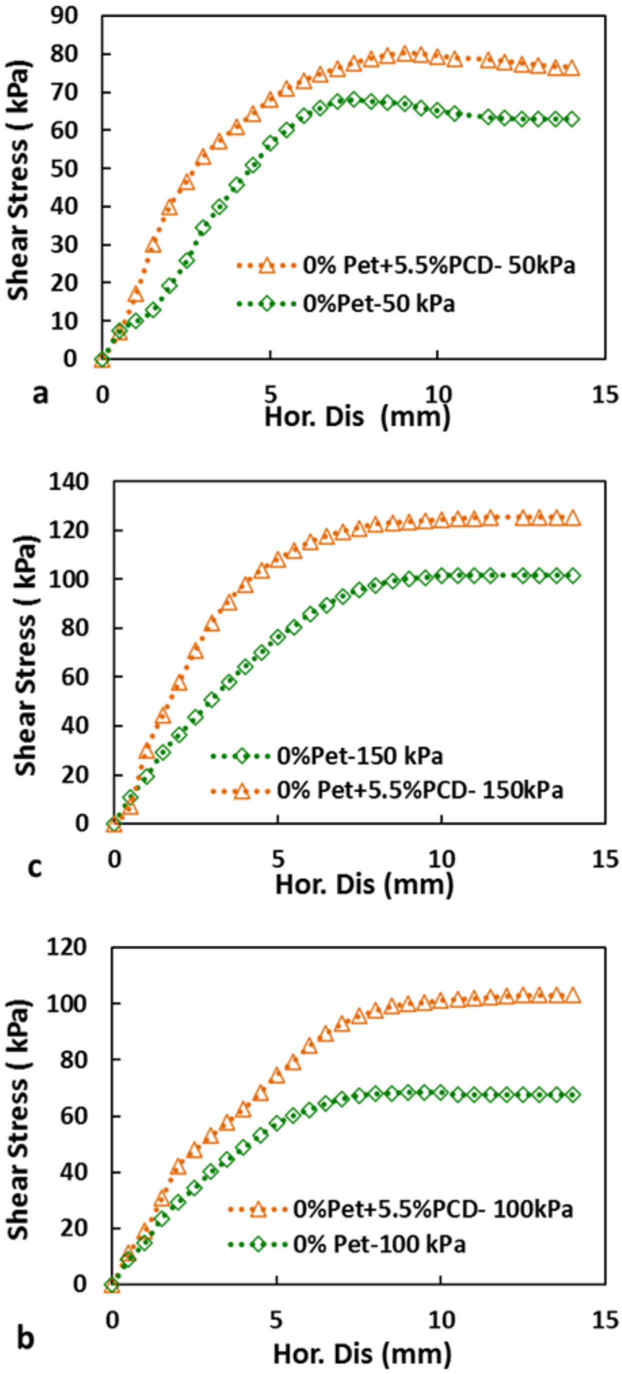

Fig. 5 Effect of PCD on shear strength of the clay in direct shear test results

the friction due to petroleum to be compensated and internal friction angle increases from 5 to 32 degree. PCD existence between soil particles as filler causes that petroleum has low effect on shear strength. In addition, due to petroleum absorption by diatomic material, only soil particles are lubricated and cohesion between soil particle and composite material increases. Sadighi and Rowshanzamir [30] reported that the addition of 2.25 percent nanoclay to oil-contaminated soil improves the friction angle by 1.2 times when compared to untreated soil. They also stated the addition of 2.25 percent nanoclay to oil-contaminated soil improves cohesion by 1.7 times when compared to untreated soil.

Figure 8 depicts the unconfined shear strength curves for petroleum-contaminated samples without PCD additive and samples contaminated with 12 percent petroleum and 5.5 percent PCD. The results show that increasing the petroleum content

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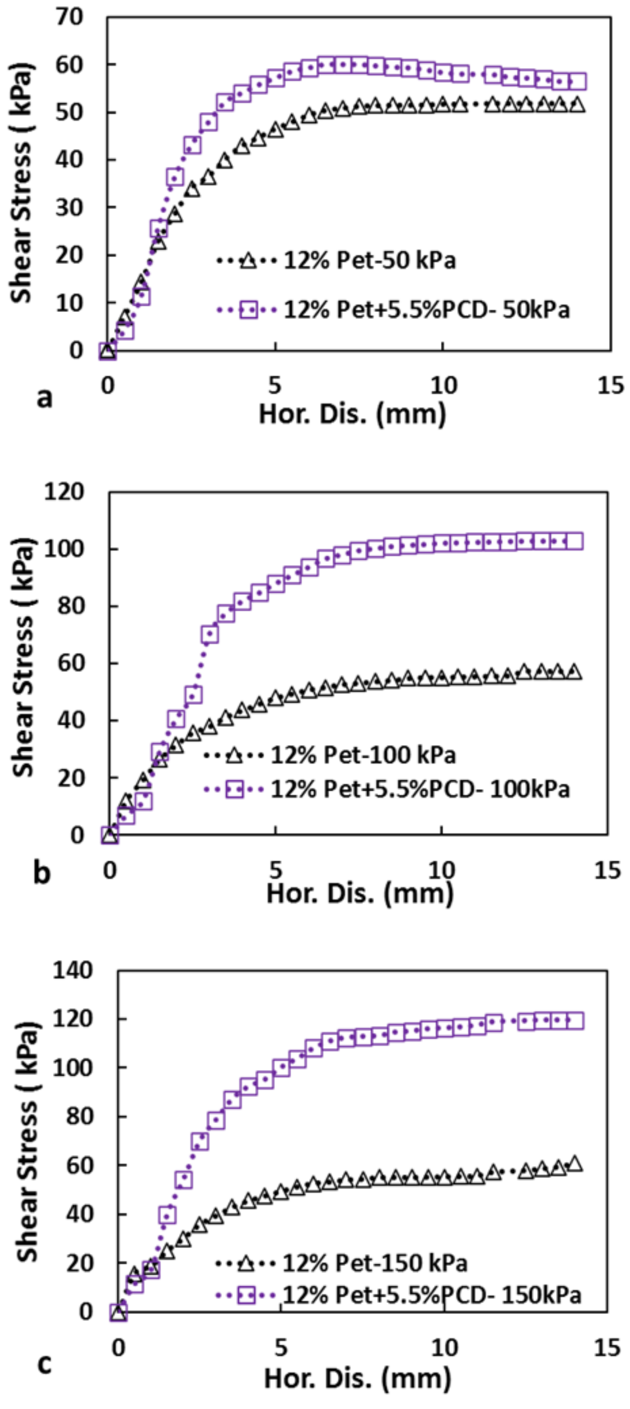

Fig. 6 Simultaneous effect of petroleum and PCD on direct shear test results

from 0% to 12% reduces the shear strength of the sample from 64.3 to 39.9. Furthermore, the unconfined shear strength of the sample, which contains 12 percent contamination and 5.5 percent PCD, increases by approximately 56.3 percent. This is consistent with the shear strength results of the direct shear test. According to Kalhor et al. [31], clay stabilized with nano-SiO_2 increases unconfined shear strength values by 14 - 29 percent.

3.3 Effect of petroleum and PCD on soil permeability

An essential element in designing of compacted soil liners is the permeability [31]. Gerashi et al. [32] concluded from a review of the research presented in this field that the presence of chemicals in the soil slightly increases the permeability of the clay. The results vary with time, measurement and testing, soil properties, and the type of chemicals used, which in some cases reduced permeability

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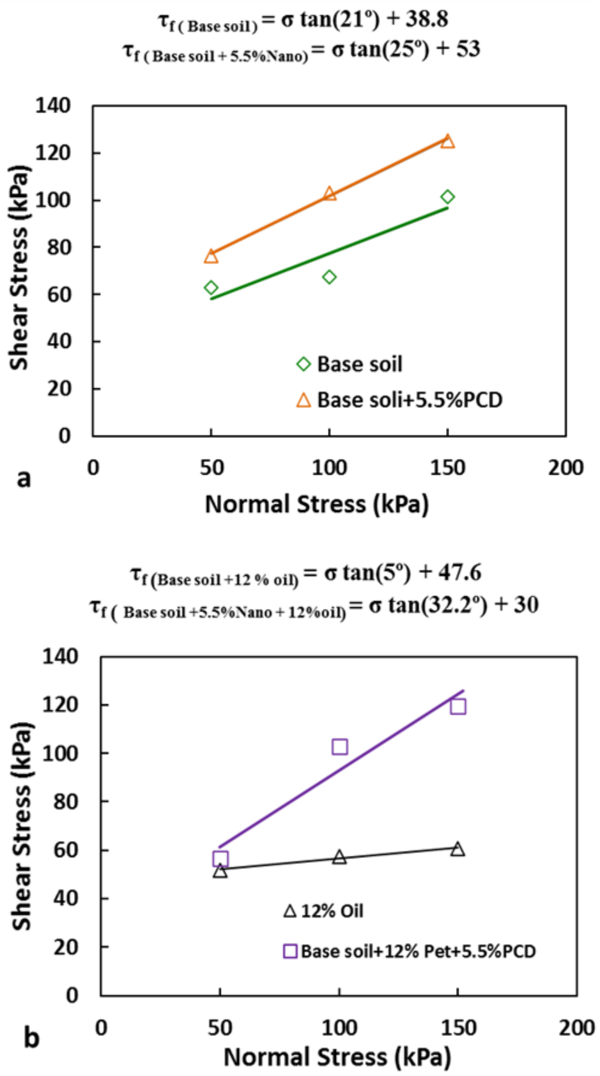

Fig. 7 Mohr-Coulomb failure criteria (a)base material and base material with 5.5% of PCD (b) base material and base material with 12% petroleum and 5.5% PCD

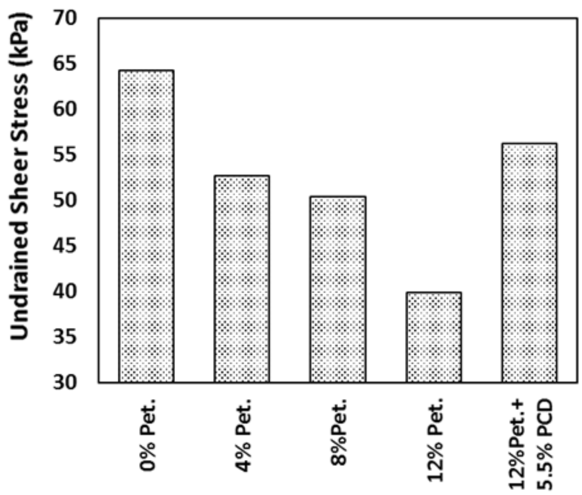

Fig. 8 Unconfined shear strength of petroleum contaminated clay without and with 5.5% PCD

in the presence of chemicals. Permeability test for base material without additive and including 5.5% of PCD for the two passing fluid such as water and petroleum was carried out and the results are presented in Table 6. The results show that petroleum causes decrease in permeability for water. The reason can be due to more viscosity of petroleum relative to water. Adding PCD petroleum contaminated soil sample decreases the permeability. In fact, PCD fills the soil pores, consequently, permeability decreases significantly. A typical clayey soil has a fabric made up of small clusters, each of which contains a slew of small particles. The void ratio of clayey soils is caused by voids within and between individual clusters. When flow occurs, the majority of the fluid flows in the pore spaces between the clusters rather than the space inside the clusters and between the clay particles [31].

Table 6 Summary results of permeability tests

Test No.	Material	Passing fluid	Permeability (cm/sec)
1	Base material	water	4.63×10-9
2	Base material	petroleum	1.65×10-9
3	Base material with 5.5% PCD	petroleum	5.68×10-10

4 Conclusions

A polymeric composite material was created in this study by thermally combining nanocarbon fiber and epoxy resin and mixing it with diatomic soil. This substance (PCD) was used to stabilize petroleum-contaminated clay. The effects of petroleum on the shear strength of the used clay were also investigated. The shear strength is reduced by up to 25% as the petroleum content increases from 0% to 12%. This decrease in shear strength is proportional to the soil›s petroleum content. Moreover, Increasing the percentage of petroleum reduced the internal friction angle and increased the cohesion of the base material. Adding 5.5 percent PCD to the base material and base material mixed with 12 percent petroleum contamination increased shear strength to average values of 32% and 48% for normal stress between 50 and 150 kPa, respectively.

The Mohr-Coulomb failure envelope is studied in the two states of base material and petroleum contaminated soil, and it is found that adding only the petroleum to the base soil reduces the internal friction of soil particles. Adding PCD to the base material raises the internal friction angle by approximately 4 degrees. Adding PCD to petroleum-contaminated soil, on the other hand, compensates for the petroleum effect by increasing the internal friction angle from 5 to 32 degrees. According to unconfined uniaxial compression test results, petroleum reduces the uniaxial shear strength of clay by about 37%. However, when 5.5 percent PCD was added to the sample along with 12 percent contamination, the strength increased significantly.

Permeability test results show that the permeability measured with petroleum as influent is less than water for the base material without additive, due to the higher viscosity and density of petroleum relative to water. The addition of PCD to the soil reduced its permeability. In fact, PCD significantly reduced permeability due to its fine structure, filling soil porosity, and formation of cohesion between soil particles, which prevents fluid passage. According to the findings, the improved clay by PCD may be used as a liner for the base of petroleum reservoirs.

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