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Effect of MTBE Contamination on the Geotechnical Properties of Clayey and Sandy Soils

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ABSTRACT

Nowadays, contamination of soil and groundwater by hydrophobic and hydrophilic organic compounds has become a major environmental issue. One of the most dangerous contaminants that have been recognized to be carcinogenic in most areas of the world is Methyl tert-butyl ether (MTBE), which unfortunately still used in some countries such as Iran as an octane booster in reformulated gasoline. The leakage of underground storage tanks contaminates the soil layer and changes its geotechnical properties. In this study, extensive laboratory experiments including compaction, direct shear and Atterberg limits were performed on clean and contaminated specimens. These tests are done in three types of soil, namely CL, SC and SP with different amounts of contamination. According the results, with increasing the MTBE content, maximum dry density and plastic limits of fine-grained soil increase while compaction characteristic of coarse-grained soil decreases. Moreover, cohesion in fine-grained soils considerably reduces while there is no noticeable change in the internal friction angle. Therefore, the shear strength of fine-grained soils decreases, while almost remaining constant for coarse-grained soils.

1. INTRODUCTION

In the late 1970s, fuel oxygenates were added to gasoline as an alternative to tetra-ethyl lead to control carbon monoxide emissions in urban regions. Historically, the most common oxygenate in the United States was MTBE which was used to reformulate gasoline at a concentration from 11 to 15% by volume (Det al. 2003). Because of the low cost of feedstocks, easily blend with gasoline and the high-octane number annual consumption of MTBE exceeded four billion gallons per year in the United States (Squillace et al. 1996). However, MTBE has been declared to be a highly carcinogenic compound as a result of inhalation cancer tests (USEPA 2000).

Pouring of gasoline at distribution centers and leakage from corroded storage tanks or pipelines contaminate the soil environment. In these cases, pollution of soils despite affording consequences on health causes undeniable changes in the physical and mechanical properties of soils (Khosravi et al. 2013).

Several methods such as soil washing methods, vacuum extraction, incineration, biological methods, and MTBE dissociation by catalysts are utilized for remediation and reclamation of the contaminated soils (Flores et al. 2000, Ossai et al. 2020, Riser-Roberts 2020). Implementation of most of the mentioned methods is limited and uneconomical in extended contaminated areas. In such cases, considering environmental issues, using contaminated soil for engineering purposes could be an option. Moreover, aforesaid contamination can affect the engineering properties of soil layers and the serviceability of the existing structure may encounter a great risk during its lifetime. Hence, an investigation of the geotechnical characteristics of contaminated soil seems necessary. Very few studies have already been carried out on the geotechnical properties of contaminated soils.

Contamination of two types of fine-grained soils, which were contaminated by Glycerol and Propanol, has been investigated in (Meegoda and Ratnaweera 1994). The study reported that contamination increases the compressibility of low and high plasticity clay. Furthermore, the authors in (Al-Sanad et al. 1995) examined the influence of oil contamination on the geotechnical properties of Kuwaiti sand. Their results revealed that although oil contamination increases compressibility, it decreases the strength and permeability of the sand. Afterward, the aging effect on the contaminated sand was evaluated in (Al-Sanad and Ismael 1997) which demonstrated that the strength and stiffness of the oil-contaminated Kuwaiti sand increased due to aging and oil content reduction. The influence of temperature on the permeability, compressibility, and strength of the oil-contaminated sands has been studied in (Aiban 1998). The authors found that when the ambient temperature is above room temperature, the shear strength parameters don’t change while the compressibility and deformation increase. A drastic reduction in friction angle and a considerable increment in volumetric strains were observed in (Evgin and Das 1992) during triaxial tests on both loose and dense quartz sand saturated with motor oil.

In recent years, several studies were performed on the behavior of contaminated fine-grained soil (Askarbioki et al. 2019, Yazdi and Sharifi Teshnizi 2021, Zhu et al. 2023, Khodary et al. 2023, Falamaki et al. 2023). It has been concluded that crude oil decreases the strength, permeability, maximum dry density, optimum water content and Atterberg limits of clayey and sandy soils (Khamehchiyan et al. 2007). However, their results were inconsistent with (Kermani and Ebadi 2012) which found an increment in the friction angle, maximum dry density, compression index, and Atterberg limits. The consolidation test has been carried out on clayey soils contaminated by petroleum hydrocarbons and it observed that settlements increase 35 to 50 percent (Singh et al. 2008). The geotechnical characteristics of kaolinite contaminated by gas oil have been studied and reported an increase in cohesion and a decrease in friction angle and compressibility (Khosravi et al. 2013).

Mahshahr refinery is one of the most important MTBE production centers in Iran. The operation of this refinery in the South of Iran has affected the soil in the area due to contamination leakage of damaged transmission pipes and storage tanks. Thus, the soil and water are highly prone to be contaminated with oil and oil-based products, especially MTBE. The fine-grained nature of soils in Southern Iran (Khamehchiyan et al. 2007), inconsistency among the authors who studied on, and lack of investigation on the geotechnical characteristics of contaminated soils with specific fractions of petroleum such as MTBE, are three key reasons for the selection of this subject.

The main purpose of this study is to investigate the influence of MTBE on the geotechnical properties of three soil types as a representative of Mahshahr polluted soils. The laboratory program included Atterberg limits, compaction and direct shear tests. Results indicated a different behavior in coarse-grained and fine-grained soils under the influence of contamination.

2. Material and Methods

In this study, three types of artificial soil have been selected to evaluate the variation of geotechnical parameters for both fine-grain and coarse-grain soils. Firoozkooh sand, which is classified as SP (poorly graded sand) in the Unified Soil Classification System (USC), was chosen as a representative type for coarse-grain soils. Soil samples with SC (clayey sand) and CL (low plasticity clay) classification were prepared by adding 20% and 50% kaolinite to the aforementioned sand by dry weight, respectively. Fig. 1 shows the grain size distribution of all the samples, specified by sieve analysis and hydrometer test (ASTM D422-63). Table 1 represent X-ray Diffraction (XRD) analysis results used to distinguish minerals of Firoozkooh sand and used kaolinite in this study. Some of the basic properties of the soil samples such as specific gravity (ASTM D854), plastic and liquid limits (ASTM D4318) and maximum dry density (ASTM-D698) have been summarized in Table 2. MTBE which is used in the present paper has been provided from the Mahshahr refinery. Table 3 indicates its physical properties of it.

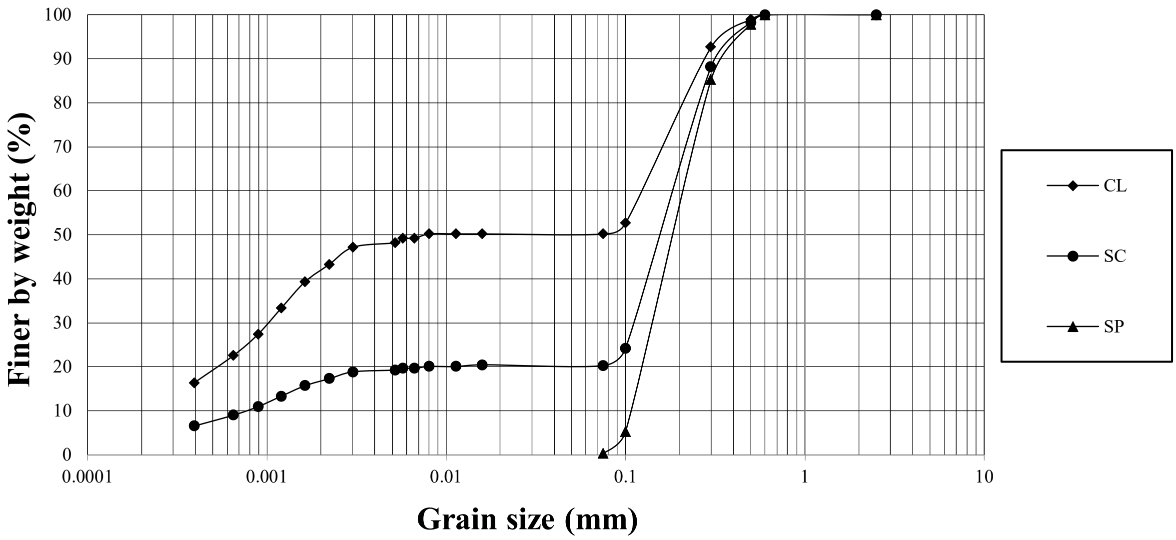

Fig. 1 Grain size distribution curve for soil samples

Table 1. XRD analysis of soils

Chemical component	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	K_2O	LOI	Sum
Percentage in kaolinite	57.12	30.47	0.84	0.45	1.13	0.67	8.57	99.25
Percentage in Firoozkooh sand	97.88	1.15	0.25	---	0.34	---	---	99.62

Table 2. Basic soil properties

soil	Specific gravity (Gs)	Liquid limit (%)	Plastic limit (%)	Maximum dry density (gr/cm3)	Percent finer than 2 µm
SP	2.65	NP	NP	1.69	0
SC	2.62	NP	NP	1.64	15.72
CL	2.59	48.5	25.6	1.91	39.32

NP: not possible.

Table 3. The MTBE properties (from the supplier, Mahshahr refinery)

Molecular Formula	molecular weight (g/mol)	Specific gravity at 68 °F	Dielectric constant at 20 °C	Surface tension at 32 °C (N/m)	Kinematic viscosity at 20 °C (mm^2/s)
$C_5H_{12}O$	88.15	0.74	2.6	1.83	0.47

For each test, the required amount of kaolinite and Firoozkooh sand were first passed through Sieve #4 to remove rocks, remnants of plants or large pieces of trash and then dried by oven at 105°C for 24 hours. Afterwards, purified soils were mixed at chosen proportion as explain in previous section. The prepared specimen was blended with MTBE in the amount of 0, 4, 8 and 12% by weight of dry soil samples and placed in an insulated container. The soil and MTBE were stirred together until a uniform mixture was obtained. The containers maintained in an oven in the temperature of 27°C for one week to allow all possible interactions to take place.

There were some limitations in doing experiments with the specified amount of contamination due to the volatile nature of MTBE. Hence, in the Atterberg limits and compaction tests, MTBE losses were determined by measuring the exact amount of soil, water and MTBE at the beginning and at the end of each test and assuming that evaporation of water is negligible compared to MTBE. The aforesaid procedure was carried out three times and the average amount of MTBE losses was added to the specimen at the beginning of each experiment to compensate for its loss. For example, to modify the results of the compaction test in the sample with 8% contamination, the authors initially calculate MTBE losses during the proctor test with 8% contamination three times and the average amount of losses are added to the soil prior to the main test. Thus, the final test has been run with for example 9% MBTE but reported it to be 8% considering the added 1% as what would be lost from the soil during the test. However, the indirect shear test, this procedure was not followed due to the experiment and its time-consuming nature of it. In this case, the average value of MTBE in the beginning and at the end of the application of shear stress was measured and this value was reported as contamination percent.

3. Results and Discussions

As mentioned earlier, the geotechnical properties of the contaminated soil samples have been investigated in three different laboratory tests. The laboratory program included Atterberg limits, compaction and direct shear tests. Details of the tests and their results are described as follows.

Atterberg limits or consistency limits are based on the concept that fine-grained soils can have different states as water content changes and are specified by plastic limit (PL), liquid limit (LL), and plasticity index (PI). In this paper, Atterberg limits have been evaluated for low plasticity clay (CL) contaminated with different amounts of MTBE. Results in Fig. 2 indicate that the PL increases considerably as the MTBE content increases to 12% while the LL decreases with a very slight slope and consequently the PI decreases.

An increase in the PL can be explained by a diffuse double-layer theory which describes the combination of two opposite layers of charged ions surrounding the negatively charged surface of clay particles. Water that is attracted to clay particles is known as double-layer water or adsorbed water while water in the pore space of soil that moves easily is called free water (Das 2021). MTBE is a highly polar compound that dissolves in water due to the presence of negatively charged oxygen atoms (Moyer and Kostecki 2004). As mentioned before, dry soil and pollution were mixed and kept for one week to allow all potential interactions to happen. However, contaminant does not attract the soil particles because of repelling forces between them. Application of modified clays with monovalent or polyvalent cations to decontaminate the water from MTBE can prove the impossibility of reaction between natural clay minerals and MTBE (Gitipour et al. 2008). Solubility of MTBE in water causes a significant reduction in water potential to establish polar covalent bonds with cohesive soil. Consequently, at a low amount of water, the clay particle behaves more like granular soil. As the moisture content increases, double-layer develop, and therefore, more water is required to plasticize the clay. Results are in agreement with the investigation of (Khamehchiyan et al. 2007, Khosravi et al. 2013) who believed that crude oil and gas oil with nonpolar molecules cause the PL of fine-grained soils to decrease. Since liquidity characteristics of soil depend on free water, and because MTBE is unlikely to adsorb to the soil particles, some part of the water added to the sample will appear as free water due to the aforementioned reasons. This fact could justify little decreases in the liquid limit. Moreover, the viscosity of organic fluid has been considered the effective factor in liquid limit in the study (Meegoda and Ratnaweera 1994). According to Table 3, MTBE has a lower viscosity than water, thus decreasing the liquid limit. Finally, the difference between PL and LL results in the reduction of PI.

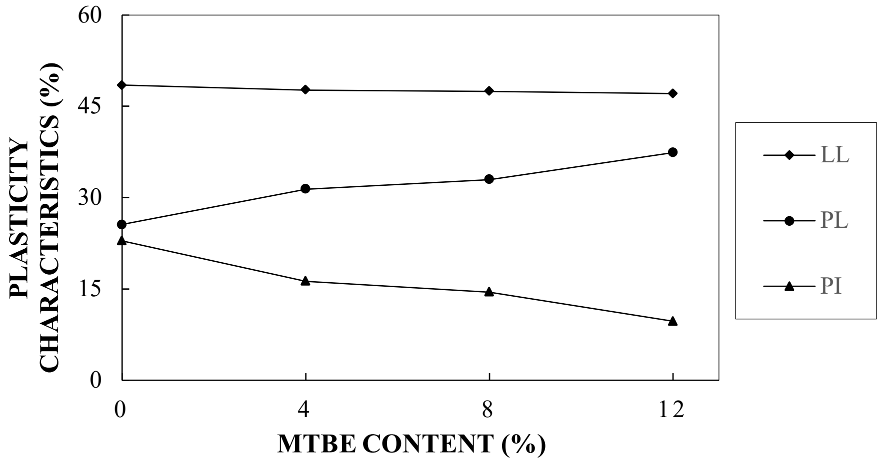

Fig. 2 The influence of MTBE on Atterberg limits of CL samples.

The results of Standard Proctor compaction tests (ASTM-D698, Method A) on artificially contaminated soil samples are shown in Figs. 3-5 in the form of dry density versus water content. In all of the samples, the presence of MTBE causes the optimum water content to reduce, while maximum dry density shows different variations. Improvement in the maximum dry density of CL samples can be attributed to structural changes of clay minerals in the proximity of MTBE. As explained above, in a high amount of contamination or low amount of water, repelling forces increase due to the existence of the same charged ions in the molecules of MTBE and clay. This will result in a lower degree of flocculation and a higher dry unit weight (Das 2021). The same results have been obtained in experiments on oil-contaminated clay by (Rehman et al. 2007). As it can be seen from Fig. 4, with an increase in the amount of MBTE, the maximum dry density has first increased from 1.64 gr/cm^3 to 1.69 gr/cm^3 and then it has decreased to 1.62 gr/cm^3. It should be noted that two factors affect the compaction of SC samples. The first factor is the presence of kaolinite which causes a slight increase in dry unit weight at MTBE content up to 4% due to the same reason cited earlier. The second one, observed at a higher amount of contamination, is the viscosity of a porous liquid. In this case, MTBE moves in the pores of the granular soil like water and because of its lower viscosity value and lubricating effect of it, the compaction characteristic curve shows a decreasing trend. Supplementary experiments have been carried out on SP samples to verify the results of proctor tests on clayey sand. The results shown in Fig. 5 indicate that contamination reduces the lubrication effect of liquid in pores space, thus making compaction difficult to perform. This observation is in agreement with the results presented by (Al-Sanad et al. 1995, Meegoda et al. 1998). These authors considered the presence of high viscosity crude oil to be the cause of increased compaction. Thus, a reduction in maximum dry density considering the reduced viscosity seems to be reasonable.

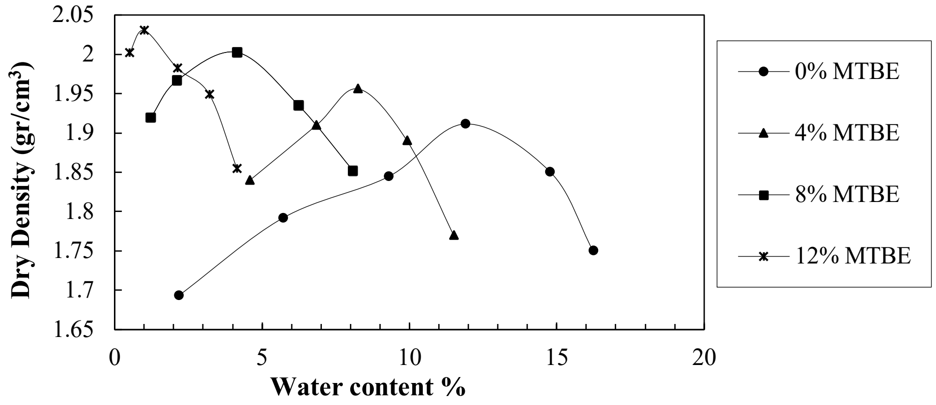

Fig. 3 Compaction characteristics of CL samples contaminated with different MTBE contents.

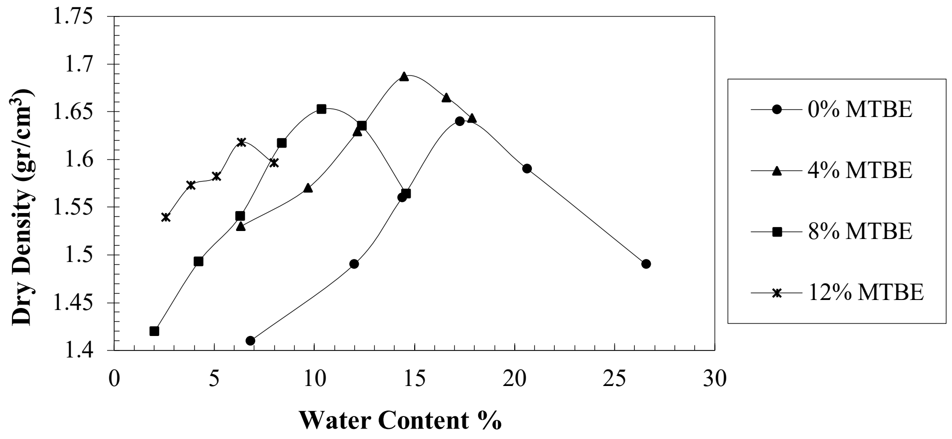

Fig. 4 Compaction characteristics of SC samples contaminated with different MTBE contents.

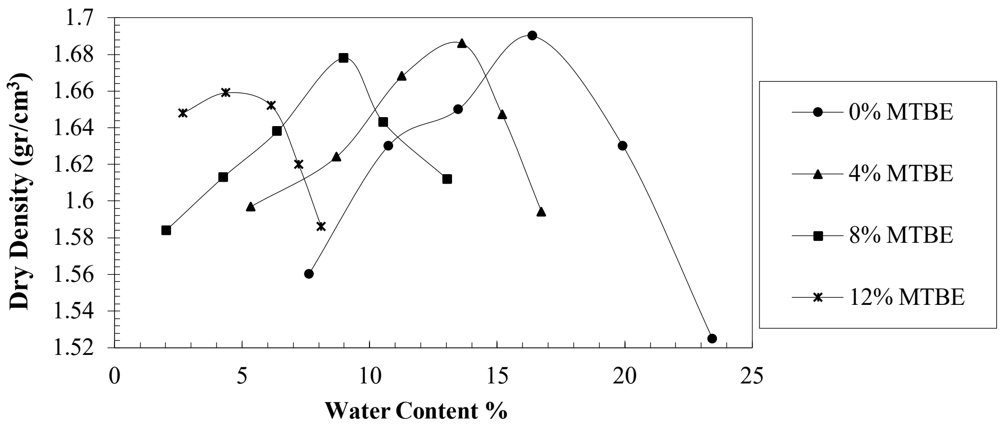

Fig. 5 Compaction characteristics of SP samples contaminated with different MTBE contents.

The shear resistance of soil is an important parameter as it controls load-bearing capacity, as well as slope stability and foundation design of a civil engineering structure. Hence, a comprehensive set of direct shear tests (ASTM D3080-72) were conducted with a standard 100*100*30 mm shear box at a constant shear displacement rate of 1 mm/min. The tests were performed at the vertical normal stresses of 1, 2 and 3 kg/cm^2. In this research, 63 direct shear experiments have been carried out and the effect of MTBE has been studied in a wide range of contamination.

Figs. 6 and 7 represent the influence of MTBE on the cohesion of CL and SC soils. The reported results indicate a significant reduction (22%) and a slight increase (6%) in the cohesion of contaminated CL and SC samples, respectively. The increment in the cohesion of SC samples might be attributed to the intrinsic viscosity of contamination. Khamehchiyan et al. (2007) have contemplated the surface tension force of the water as the reason for the apparent cohesion of wet sand (Khamehchiyan et al. 2007). As shown in Table 3, the surface tension of MTBE is less than water, and therefore, the rate of increase in cohesion of contaminated samples is lower than specimens with water as pore fluid.

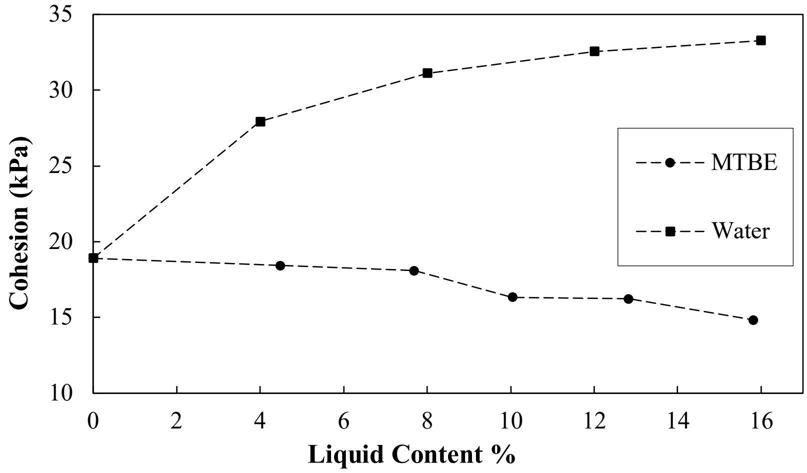

Fig. 6 The influence of MTBE and water content on the cohesion of CL samples.

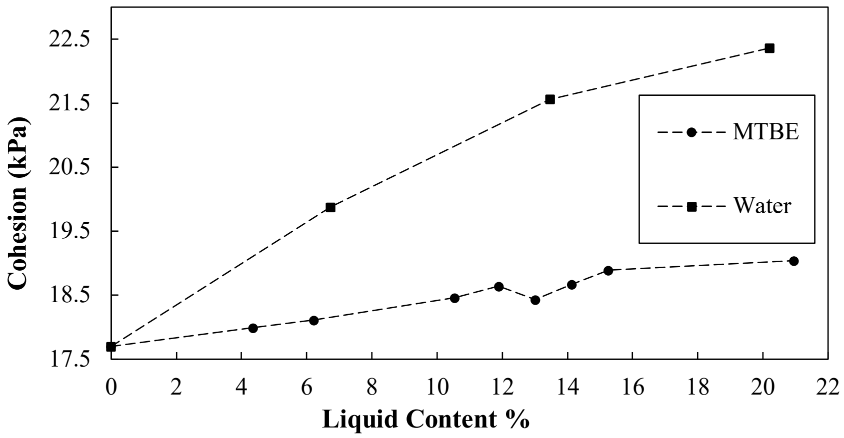

Fig. 7 The influence of MTBE and water content on the cohesion of SC samples.

In this study, specimens have been prepared by premixing soils with the contaminant. According to (Meegoda and Rajapakse 1993), in this method, long-term exposure to chemicals is simulated and contamination changes the soil structure. Ratnaweera and Meegoda (2006) found that a reduction in pore fluid dielectric constant causes a reduction in shear strength of long-term contaminated soils (Ratnaweera and Meegoda 2006). The low value of the dielectric constant of MTBE compared to water could justify the reduction in the cohesion of soil type CL. Also, studies have shown that the cation exchange capacity (CEC) of contaminated soil is lower than that of uncontaminated soil (Rehman et al. 2007). Moreover, a reduction in flocculation of contaminated clay results in a reduction in cohesion. Khamehchiyan et al. (2007) and Ur-Rehman et al. (2007) have both reported a decrease in soil cohesion due to oil contamination of soil (Khamehchiyan et al. 2007, Rehman et al. 2007). Considering the close dielectric constant of oil and MTBE, observation of similar effects is not unexpected.

The effect of MTBE and water on the friction angle of CL and SC soils are graphically illustrated in Figs. 8 and 9, respectively. One can see that although contamination does not considerably change the friction angle of soil type CL, a small increase in water content significantly reduces this parameter. Al-Shayea (2001) related a small reduction in the friction angle of gas oil-contaminated soil to the lubrication effect of gas oil on the surface of the particles (Al-Shayea 2001). The MTBE has a lower viscosity than water, which cause the soil to show higher friction angle values. Moreover, as explained before, in the vicinity of MTBE, clayey soil behaves like granular soil. This can also be deduced here from the lower reduction in the friction angle. Khosravi et al. (2013) have reported similar results on kaolinite contaminated with gas oil (Khosravi et al. 2013). Ghaly (2001) and Shin et al. (2002) performed direct shear tests on oil-contaminated sand and found that as contamination increased, the internal friction angle of soil type SC reduces (Ghaly 2001, Shin et al. 2002).

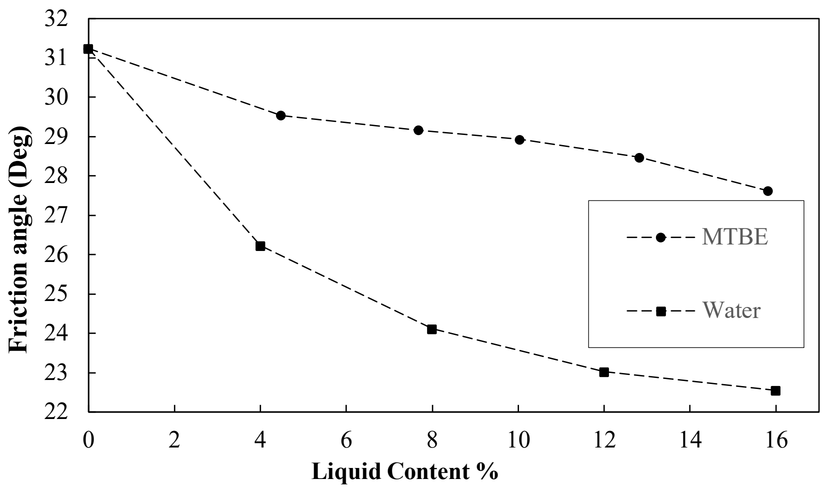

Fig. 8 The influence of MTBE and water content on the friction angle of CL samples.

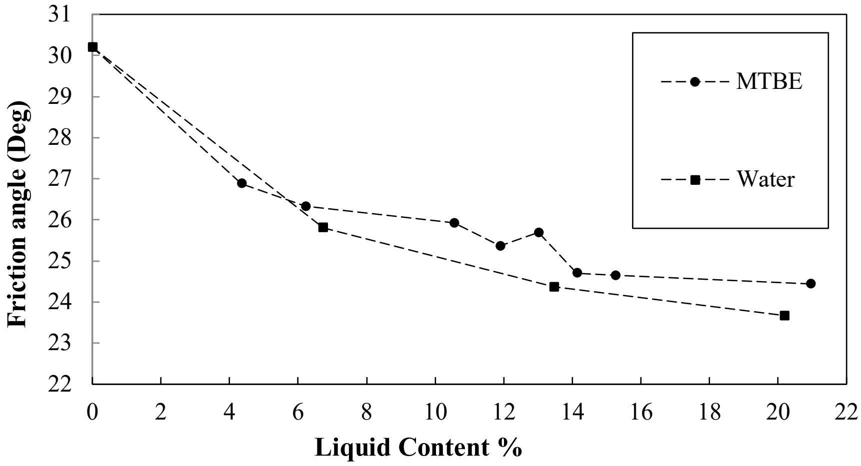

Fig. 9 The influence of MTBE and water content on the friction angle of SC samples.

Figs. 10 and 11 show the variations of shear strength of CL and SC samples, based on Mohr-Coulomb failure criterion, at 100 KPa vertical stress. It can be seen that the effect of contamination on the shear strength of soil type SC is negligible while it greatly reduces the shear strength of soil type CL.

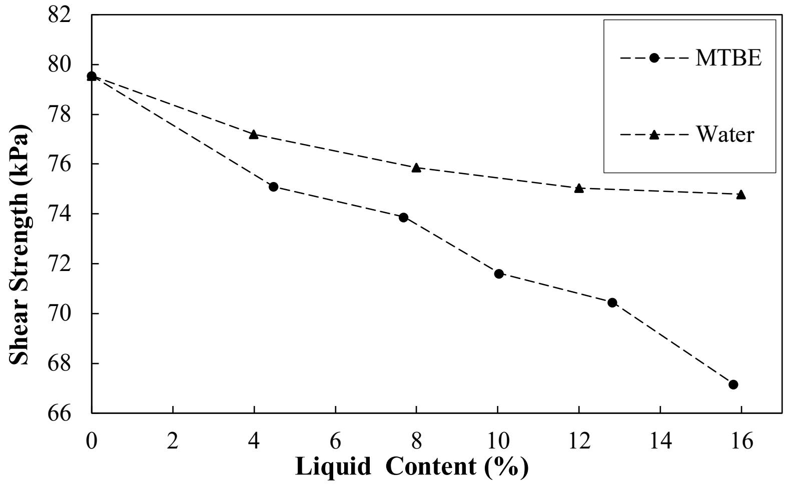

Fig. 10 The influence of MTBE and water content on the shear strength of CL samples.

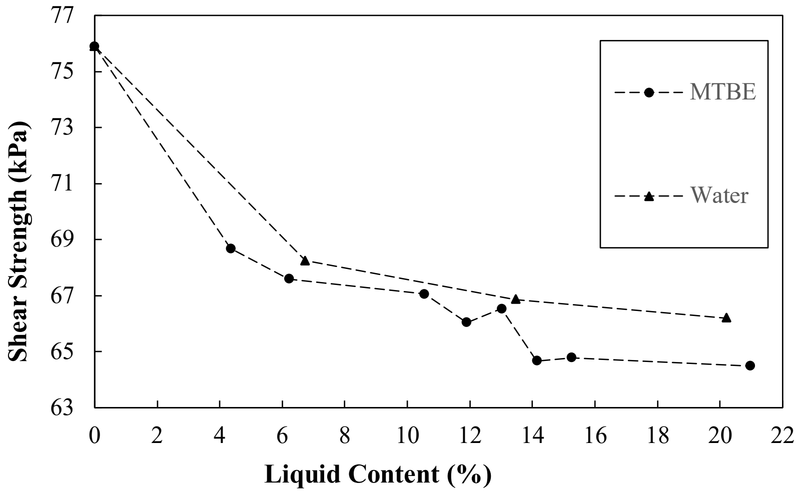

Fig. 11 The influence of MTBE and water content on the shear strength of SC samples.

4. Conclusion

A comprehensive set of laboratory testing programs was carried out to evaluate the basic geotechnical properties of soils contaminated with MTBE. For this purpose, three types of artificial soil are classified as SP, SC and CL in the Unified Soil Classification System were selected as a representative of Mahshahr refinery polluted soil in the south of Iran. The laboratory tests included Atterberg limits, compaction and direct shear have been performed on clean and contaminated specimens. The results indicated a considerable increase in the plastic limit and a slight decrease in the liquid limit of CL samples due to contamination. This may be associated with the polar characteristics of the contaminant in question. The optimum water content of all three soils decreases with an increase in contamination, while the maximum dry density of fine-grained and coarse-grained soil shows different behavior. Structural changes in clay minerals in the presence of MTBE cause the compaction characteristic curve of CL soil to increase while because of the lower viscosity value and lubricating effect of MTBE, the dry unit weight of SP samples tends to decrease. The results of direct shear tests indicate a significant reduction and a slight increase in the cohesion of CL and SC samples, respectively. In general, the shear strength of fine-grained soils reduces while there is no noticeable change in the shear strength of coarse-grained soils.

Results of this paper could be used, with considering environmental concerns, as an alternative to the remediation methods which are limited and uneconomical in lightly or moderately contaminated areas. Also, further investigation on serviceability assessment of structures supported on soil contaminated with different amounts of MTBE can be based on the findings of this study.

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