

# Journal of Testing and Evaluation

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### DOI: 10.1520/JTE20210477

Impacts of Pre-contamination Moisture Content on Mechanical Properties of High-Plasticity Clay Contaminated with Used Engine Oil doi:10.1520/JTE20210477



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#### Reference

K. R. Omar, B. Fatahi, and L. D. Nguyen, "Impacts of Pre-contamination Moisture Content on Mechanical Properties of High-Plasticity Clay Contaminated with Used Engine Oil," *Journal of Testing and Evaluation* https://doi.org/10.1520/JTE20210477

#### ABSTRACT

The oil contamination of soils and the remediation techniques to enhance the engineering properties of the ground have been an emerging challenge in the geoenvironmental field. While several studies were conducted to examine the behavior of the contaminated granular soils, little is known about the mechanical properties of the oil-contaminated clays. This paper investigates the impacts of the in situ pre-contamination moisture content (PMC) on the behavior of fine-grained soil contaminated with various levels of used engine oil. Extensive laboratory experiments were performed on sandy clay with different initial moisture conditions and various amounts of used engine oil varying from 0 to 16 %. The experimental results, including the Atterberg limits, linear shrinkage (LS), unconfined compressive strength, shear strength, and small-strain shear modulus in conjunction with microstructural image analysis, were reported and discussed. It is observed that when oil content was increased, both LS and plastic limit (PL) increased while the liquid limit decreased in the contaminated soil. Moreover, the inclusion of engine oil contributed to the reduction in the plasticity index, which was also impacted by the PMC of the soil. An increment in the PL was correlated with a significant decrease in shear strength, shear modulus, and other associated parameters such as friction angle and cohesion. In agreement with the results, a broader range of elasticity and improved stability at the microstructure level was associated with a lower pre-contamination water content (PMC). Overall, this paper shows that knowledge of site moisture levels before contamination is essential to evaluate the implications of contamination by used engine oil.

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Manuscript received July 11, 2021;

30, 2022; published online May 26,

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accepted for publication March

2022.

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#### Keywords

oil contamination, small-strain shear modulus, Atterberg limits, unconfined compression strength, shear modulus, microstructural analysis

### Introduction

In recent years, the development of green infrastructures has gained significant attention. For example, in stormwater management, the natural properties of soil having a high infiltration capacity and permeability are exploited to prevent flooding and recharging water supplies rather than relying fully on pumps and drains. Moreover, soil can be utilized for filtration purposes to avoid contamination of stormwaters with contaminants such as hydrocarbons.<sup>1</sup> Rapid advances in information technology have led to the concept of smart structures with continuous and real-time monitoring of infrastructures and underlying soil via sensors and data analytics to allow an enhanced delivery of services and improve long-term performance.<sup>2</sup>

Oil and petroleum products provide major industrial applications worldwide, particularly for heavy industries and transportation. More specifically, engine oils are utilized in various engines for lubrication purposes. After a period of usage, the structure of the oil changes by the absorption of heavy metals, and therefore, disposal or replacement is needed. An inappropriate disposal process of the used engine oils or leakage from machinery can contaminate the underlying subsurface soil and ground and underground water.<sup>3</sup> The occasional contamination happens accidentally during exploration, the production operation processes, and transportation. However, most crude oil spills have been caused because of ageing facilities, lack of maintenance, and human error.<sup>4</sup> It is essential to appreciate the impact of oil contamination in soils that underline crucial infrastructure, such as roads and railways, or within heavy machinery foundations. Depending on the site and environmental conditions, the in situ moisture content of soil can range from dry to wet and it can affect compressive behavior, shear strength, unconfined compressive strength (*UCS*),<sup>5</sup> and permeability coefficient of soils.<sup>6</sup> Hence, it is important to conduct experimental studies on soft clay contaminated with oil to understand the properties of contaminated soil and assist geotechnical practitioners in reducing uncertainties in evaluating foundations and earthworks and assessing the stability of ground for construction activities.

The presence of oil has significant impacts on the soil characteristics as it degrades the physical and mechanical properties of the soil.<sup>7</sup> Such degradations may result in unbalanced settlement, causing cracks and leakage in the overlying structures (e.g., oil tanks and pipelines) and the surrounding. Moreover, as reported by Khamehchiyan, Charkhabi, and Tajik,<sup>5</sup> the chemical composition of the hydrocarbon and the soil properties can impact how the contamination affects the ground response. Currently, there is no worldwide standard legislation for evaluating and remedying polluted land.<sup>8</sup> The Australian and New Zealand Environment and Conservation Council have been working closely with the National Health and Medical Research Council to establish technical guidelines to inform and educate governments, industries, labor unions, and the general public about the issues associated with evaluating and controlling contaminated land in Australia. Although these guidelines were implemented in 1992, they have since been revoked and replaced by new guidelines under the National Environment Protection Council Act 1994 by the National Environment Protection (Assessment of Site Contamination) in 1999.<sup>8</sup> In the Australian and New Zealand Environment and Conservation Council guidelines and the National Environment Protection (Assessment of Site Contamination), a recommended order of options for site cleanup and management is outlined as (I) treating oil on site to eliminate contaminants and reduce hazards to an acceptable level and (II) treating excavated soil off site and subsequently returning it to the site.<sup>8</sup> This enables waste to be disposed properly at a disposal facility. Alternatively, it may be used as a landfill, depending on residual contamination levels. According to Rajabi and Sharifipour,<sup>9</sup> several studies were conducted to investigate the effects of various types of oil contamination, including used engine and motor oils, crude oil,<sup>10</sup> and diesel oil<sup>11</sup> on various geotechnical properties such as Atterberg limits, strength, permeability, and optimum water content. A study conducted by Rajabi and Sharifipour<sup>9</sup> showed that short-term weathering effects reduce the shear modulus of oil-contaminated sand.

Similarly, Khodary, Negm, and Tawfik<sup>6</sup> reported that compared to water, crude oil had a diminished surface tension, which was the primary reason why laterite soil became less cohesive because of the addition of oil. According to Khamehchiyan, Charkhabi, and Tajik,<sup>5</sup> the addition of crude oil decreases the attraction forces of cohesion between soil grains (i.e., soil cohesion) resulting from interparticle slippage. Specifically, the reduced cohesion is caused by the intermolecular cohesive forces experiencing a diminution. Furthermore, Al-Adly, Fadhil, and Fattah<sup>12</sup> showed that soil compaction characteristics were slightly dependent on the oil contamination extent, while the bearing capacity of the soil was significantly reduced because of contamination. Safehian, Rajabi, and Ghasemzadeh<sup>13</sup> observed that the soil strength parameters, including cohesion, internal friction angle, and *UCS*, decreased at a microscopic scale, whereas the compression coefficient increased because of oil contamination. Moreover, an increase in the Atterberg limits with increasing oil content was reported by Ratnaweera and Meegoda.<sup>14</sup> In contrast, Jia et al.<sup>15</sup> observed a significant decrease in Atterberg limits with the oil content. Furthermore, contradicting results were reported on the unconfined compression strength of oil-contaminated soil in studies conducted by Khamehchiyan, Charkhabi, and Tajik<sup>5</sup> and Khosravi et al.,<sup>7</sup> where the former reported degradation of the strength and the latter observed an enhancement in the strength because of the addition of oil.

The amount of water within clay particles strongly influences properties of soils such as shear strength and permeability.<sup>16</sup> Water molecules function as dipoles and can be absorbed onto the surface of clay particles that are negatively charged. Moreover, exchangeable cations tend to be hydrated on the soil surface and may leave the surface. Oil is a pure hydrocarbon nonpolar substance that displaces water molecules from around clay particles.<sup>17</sup> Consequently, there is an impact on the charge content. Natural soils contain some pore water, which usually comes into contact with the clay. After the oil contaminates the clay, the oil infiltrates the clay-water system. The moisture content of the soil after oil contaminated soil. For instance, a contractor adds an additional amount of water during construction to perform compaction; however, they may be unaware that a high amount of free water is present in the pore spaces because of oil contamination. This means that the soil might already be located on the wet side of the compaction curve.<sup>18</sup> Engineers can alter the moisture content for compaction once this effect is considered. According to Kermani and Ebadi,<sup>19</sup> when water in excess of a specified quantity is added, an excessive amount of free water is created. This enlarges the interparticle space, which ultimately lowers cohesiveness. Moreover, increasing the water quantity to the saturation point completely wipes out capillary suction, further reducing cohesion.<sup>20</sup>

The impacts of the initial moisture content have been assessed by several researchers on compressive behavior,<sup>21</sup> shear strength, UCS,<sup>5</sup> and permeability coefficient of soils.<sup>22</sup> Listyawan, Wiqoyah, and Sukmawati<sup>23</sup> reported that soft soil properties such as cohesion, suction, and swelling depend on the soil's original conditions, including water content and dry unit weight. Furthermore, it is observed that the mechanical behavior of the oil-contaminated clay depends on the soil characteristics, particularly the initial moisture content, soil type, type, and amount of oil within the soil matrix. Indeed, the initial moisture condition of the ground plays a vital role in the interaction between the soil matrix and the oil particles, impacting the behavior of oil-contaminated soil significantly. However, to the best of the authors' knowledge, the studies in the literature are limited to a single initial moisture content of soils (e.g., initial moisture content of 0 %<sup>24</sup> and 10 %<sup>11</sup>). Also, the effect of the precontamination moisture content (*PMC*) of the soil can likely explain some of the contradicting observations made in the literature. Hence, the main objective of this paper is to investigate the effect of pre-contamination moisture conditions on the mechanical properties of soil contaminated with various oil contents.

An extensive experimental program was performed on a reconstituted soil sample composed of a mixture of kaolin, bentonite, and sand, with varying initial moisture content or *PMC*. Furthermore, used engine oil with a varying content ranging from 0 to 16 % was used as a contaminant. The adopted oil content was capped at 16 % because further addition of oil beyond this limit resulted in highly saturated samples with oil being squeezed out

during sample preparation, in line with the observation made by other researchers.<sup>5,19</sup> Based on the results of the Atterberg limits, further experimental tests, including the *UCS*, bender element (BE), direct shear, and amplitude sweep tests (ASTs), as well as scanning electron microscopic (SEM) imaging, were conducted on contaminated samples.

### Mechanisms of Oil-Clay Interaction

Molecules are distinguished into two major types: polar and nonpolar. The interaction between clay and oil particles plays a significant role in understanding the behavior of contaminated soils. Nonpolar molecules, such as oil, are characterized by equal distribution of electrical charges throughout, without the formation of poles with positive or negative charges. On the other hand, polar molecules, such as water, are characterized by the lack of even electron sharing between two atoms in a covalent bond, which gives rise to a dipole, whereby a portion of a molecule is slightly positively charged and the other portion is slightly negatively charged. A molecule and a charged surface can interact based on the Van de Waals force and the force stemming from the diffuse double layer (DDL) that develops at the water-soil particle interface. As Kermani and Ebadi<sup>19</sup> explained, dipolar water molecules are attracted to the clay particle surface with a negative charge in the DDL, and the cations within the DDL refer to the water retained by the particles of clay because of attraction forces. Furthermore, hydrogen bonds are formed because of the attraction of hydrogen atoms in the water molecules to the oxygen atoms in the clay layer. Hence, clay shows a particularly strong retention of adsorbed water. The liquid state of soil depends on the free water, representing the water within the pore space that clay particles do not assimilate and can travel freely in the soil.<sup>9</sup> An illustration of the interaction between oil and water on the clay surfaces is depicted in figure 1. In figure 1.4, clay particles absorb dipolar water molecules because of their negative surface charges. The addition of oil, a nonpolar substance (neither electropositive nor negative charges), stops water molecules from bonding to clay particles, preventing water from reaching the DDL of clay particles. In addition, oil replaces the adsorbed water over time and interacts with clay particles, as shown in figure 1B. Subsequently, higher quantities of free water can enter the conductive channels, which in turn can improve soil hydraulic conductivity.<sup>17</sup>



FIG. 1 Mechanisms of (A) clay and water interaction and (B) oil-clay water interaction.

The interaction of soil particles and pore fluid can be clustered into two groups: mechanical interaction and physicochemical interaction. Mechanical interaction is the dominant mechanism in granular soils where solid particles contact each other in translation and rotation. For instance, the contaminating fluid's viscosity, density, and surface tension properties are dominant parameters, particularly in granular soils. On the other hand, physicochemical interaction controls the behavior of fine/cohesive soils in which the DDL is formed around the particles.<sup>25</sup> In addition, fine-grained soils are mainly prone to physicochemical interactions because of the formation of a DDL surrounding the soil particles during the pore fluid-soil mineral interaction.<sup>6</sup> According to Mitchell and Soga,<sup>26</sup> various soil structures are produced because of changes in DDL thickness. The soil structure is relatively dispersed for larger DDL thickness, while with the decrease of the DDL, the clay structure tends to flocculate. The DDL thickness relies on numerous components, including temperature, anion absorption, pH, the concentration of pore fluid (e.g., oil/water), dielectric constants, density, viscosity, ion type, and surface tension.

Oil viscosity plays a vital role in soil-oil interaction. The presence of contaminants in the soil is an issue for environmentalists and a source of deterioration to the geotechnical characteristics of soil. Because of its electrochemical properties, clay soil is mostly affected by fluid substances (such as oil) that contaminate the environment.<sup>4,27</sup> When contaminants travel through soil or groundwater, they are influenced by diverse variables, such as soil permeability and the adsorptive capacity of soil solid.<sup>7</sup> The soil characteristics and the chemical properties of the contaminant together shape the extent of any soil contamination.<sup>5</sup> In addition, gravity causes the downward movement of hydrocarbon contaminants into the soil, causing soil in the pathway to become contaminated. In addition, water can transport hydrocarbon contaminants, whereby capillary action causes them to reach regions in the soil deposit.<sup>9</sup>

When crude oil is spilled, the liquid hydrocarbon migrates down to the groundwater causing partial saturation of the soil along the pathway of the hydrocarbons.<sup>4</sup> Once crude oil reaches the ground, depending on the permeability of contaminated soil and the level of the water table, the light hydrocarbons start to spread horizontally by migrating within the capillary zone, which causes further saturation of the soil. The extent of spreading is much greater with light crude oil than with medium and heavy crude oil because light crude oil contains a high percentage of light hydrocarbons that can easily penetrate and migrate through the soil structure.<sup>4</sup> Sharma and Reddy<sup>28</sup> indicated that the permeability of contaminated soil is based on the viscosity of the fluid filling the voids, where an increase in intrinsic permeability (k) is because of an increase in density and a decrease in viscosity, which indicates the significant effect that density and viscosity have on the permeability of the soil. Moreover, various kinds of clay minerals that are chemically active can interact differently with pore fluid. Consequently, the permeability response of the soil is affected as a result of changes in the soil structure.

Rajabi and Sharifipour<sup>9</sup> suggested that the spread of crude oil within the vadose/unsaturated zone is influenced by the interaction between the three immiscible fluids (air, water, and oil). However, the dominant infiltration process is comprised of advective dissemination wherein presumed as inactive air phase. According to Darcy's law, it is possible to define the rate at which oil penetrates the soil while viscosity and density remains constant. Most importantly, oil and its byproducts comprise the contaminants in the ground that can infiltrate the soil at different depths depending on soil permeability and contaminant viscosity. According to Brown et al.,<sup>29</sup> as the oil infiltrates, it is influenced by soil properties (pore size and porosity) as well as physical properties (viscosity, density, and surface tension) and the volume of oil spilled.

Moreover, viscosity denotes the extent to which a fluid is resistant to deformation because of the internal friction that permits the fluid to be resistant to flow. Fresh crude oil with volatile qualities becomes more viscous as evaporation transpires. Thus, in the aftermath of a spill, fluids with low viscosity can infiltrate soil at a higher speed than those with high-viscous fluids.

The viscosity of used engine oil in the soil changes with time because of several factors such as the location and seasonal variation of the water table and the presence of fissures in the soil.<sup>30</sup> Furthermore, chemical reactions within the soil matrix for an extended period of time affect the viscous properties of oil. For example, oxidation of soil increases the viscosity, while biodegradation of soil results in reduced viscosity. However, this effect is beyond the scope of this current study. Extended future studies, including long-term experimental studies, should be

conducted to investigate the change in viscosity of used engine oil with time, which can have significant effects on the geotechnical properties of oil-contaminated clay.

Another factor affecting the changes in viscosity is temperature. The viscosity index (VI) can gauge viscosity, which is one of the principal characteristics of the oil. Viscosity is highly influenced by temperature changes, increasing liquidity as temperature rises.<sup>31</sup> The VI employs empirical numbers to signify the temperature dependence of the oil viscosity. While a low VI is indicative of a comparatively significant change in viscosity with temperature, a higher viscosity in oil implies a slower flow and a thicker fluid.

The intrinsic permeability (k) of contaminated soil increases when there is an increase in density and a decrease in the fluid viscosity filling the voids. When the permeability of the soil increases because of the decrease in viscosity of the crude oil with increasing temperature, the crude oil spreads faster, and therefore, a larger area is affected.<sup>31</sup> Furthermore, the possibility of crude oil contamination reaching the underground water is higher. Therefore, the viscosity of the spilled oil, to some extent, influences the rate of spreading and, as viscosity is temperature dependent, the viscosity of oil decreases exponentially with increasing temperature.<sup>30,31</sup>

The surface attachment, which is essentially the adsorption of contaminants to the clay particle, can be categorized into chemical and physical adsorption streams. The physical adsorption mainly results from the van der Waals forces between the clay minerals and the contaminants. On the other hand, in chemical adsorption (or chemisorption), a stronger bond of a chemical substance is formed (i.e., covalent bond).<sup>25</sup>

Rajabi and Sharifipour<sup>9</sup> explained that the DDL expands at a reduced ionic strength, conducive to organic molecule desorption. In addition, at the contamination stage, the guest molecules of organic contaminants pass through the interlayer spaces of clay minerals, disrupting the stability of the intermediate water-clay interface.<sup>26</sup> Zheng et al.<sup>17</sup> reported an interfacial binding between the oil and clay mineral following the displacement of water molecules. As discussed by Ugochukwu,<sup>32</sup> the covalent bond is formed because of the binding of multivalent cations at the clay surface to the polar compounds in the oil. Concomitantly, there is direct adsorption of certain organic polar compounds onto the mineral surface, making the clay surface more oil-wet by supplanting the most labile cations occurring at the surface of the clay. The organic molecules' surface availability and the displacement of water molecules are the determinants of organic compound adsorption on clay surfaces within aqueous systems.<sup>26</sup> Although exchanging cationic organics with inorganic adsorbed cations is possible, the exchangeable cations' displacement is impossible if the cation size is smaller than the organic cation. Furthermore, the overall quantity of retained organics may be supplemented by van der Waals forces attracting large organic molecules to clay surfaces.

Important factors affecting physicochemical interactions are the dielectric constant of the pore fluid (i.e., oil and water), the type of clay mineral, and the concentration and capacity of electrolytes. Moreover, the geometrical properties of the particles (e.g., size, position, and shape) can remarkably influence the interaction forces. Among these, the dielectric constant of the nonpolar contaminant plays a significant role in altering the thickness of the DDL of the kaolinite.<sup>6</sup> Oil as a contaminant contains a variety of nonpolar hydrocarbons with a low dielectric constant (e.g.,  $\varepsilon_{oil} = 2.1$ ) compared to a higher value for water ( $\varepsilon_{water} = 80$ ). Accordingly, the thickness of DDL is reduced because of the addition of oil.<sup>26</sup> The changes in the electric potential and DDL thickness of the water-clay and oil-clay are schematically depicted in **figure 2**. From this figure, it can be inferred that the electric potential on the surface of the clay particles and, more importantly, the interparticle repulsion forces drop proportionately with the reduction of the dielectric constant ( $\varepsilon$ ).<sup>25,33</sup> Thus, the addition of oil can weaken the bonds between the soil particles and result in the detachment of the clayey films from the particle surface.<sup>9</sup>

### Test Materials and Sample Preparation Method

In this study, multiple trials were performed with different compositions to generate high-plasticity soil (classified as CH) prior to finalizing the mixture. A mixture of kaolinite (70 %), bentonite (15 %), and sand (15 %) was adopted to represent fine-grained soil such that it can absorb and hold contaminants for an extended period of time because of sensitivity to the contaminants.<sup>34</sup> Moreover, there is considerable empirical evidence from several



researchers, such as Khamehchiyan, Charkhabi and Tajik,<sup>5</sup> Jia et al.,<sup>15</sup> and Kermani and Ebadi,<sup>19</sup> wherein finegrained soils are employed to explore the significance of contaminated soils. Ghadyani, Hamidi, and Hatambeigi<sup>27</sup> used kaolinite and bentonite as fine-grained soils to investigate the behavior of oil-contaminated clay subjected to induced shearing. In this study, the adopted composition was used to simulate high-plasticity clay found on site, similar to other existing research studies conducted by Jia et al.<sup>15</sup> and Meegoda and Ratnaweera.<sup>35</sup> K-10 kaolin clay is commercially available in large quantities and classified as low-plasticity clay, while the addition of Active Bond 23 bentonite to the mixture is to increase the mixture plasticity, as bentonite contains large portions of montmorillonite, which can hold a high amount of absorbed water inside its microscopic structure.<sup>26</sup> The properties of K10 kaolin clay and Active Bond 23 bentonite are given in **Table 1**.

In this study, used engine oil Castrol GTX 20W-50, frequently used in vehicle engines, was adopted. The automobile industry produces various waste oils, including spent engine oil, lubricating oil, and grease from mechanical workshops. This engine oil is in a liquid state and insoluble in water. Such oils include various hydro-carbons (*n*-alkanes and polycyclic aromatic hydrocarbons) and engine additives (e.g., amines and organometals), which cause damages to the surrounding environment.<sup>7,25,27</sup> Indeed, the chemical composition of the adopted used engine oil consists of up to 90 % base oil, which is mainly composed of hydrocarbons. The used engine oil in this study is black compared to the new engine oil, which is brown. According to Achuba and Peretiemo-Clarke,<sup>36</sup> exposure of new mineral-based crankcase oil to elevated temperatures and significant mechanical strain results in the production of a brown-to-black liquid called spent engine oil or used mineral-based crankcase oil. In addition,

#### TABLE 1

Properties of K10	kaolin clay	and bentonite a	dopted in this	study
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Soil Property	K10 Kaolin Clay	Active Bond 23 Bentonite
Liquid limit, <i>LL</i>	48.0 %	340 %
Plastic limit, PL	19.2 %	50 %
Plasticity index, PI	28.8 %	290 %
Specific gravity, Gs	2.66	2.30
Linear shrinkage, <i>LS</i>	7.0 %	35 %
Color	White	Dark gray
Soil classification, USCS	CL	СН

Weight density, kg/m <sup>3</sup>	926
Viscosity at 40°C (mm <sup>2</sup> /s)	156
Viscosity index	153
Particles > 6 $\mu$ m, part/mL	1,480
Fuel dilution, (% w/w)	<4.0

## TABLE 2 Properties of used engine oil in this study

at an elevated temperature (i.e.,  $40^{\circ}$ C), the viscosity of the used engine oil is measured at 156 mm<sup>2</sup>/s, which is less viscous than the new engine oil with the viscosity of about 181 mm<sup>2</sup>/s. The properties of the used engine oil adopted in this study as a contaminant are presented in Table 2.

A range of approaches for preparing contaminated samples for laboratory testing has been suggested in the literature, such as the method of adding the contaminant-water solution to dry clay,<sup>5,14</sup> adding the water to contaminated dry soil,<sup>24</sup> or adding contaminant to moist clay.<sup>37</sup> Ratnaweera and Meegoda<sup>14</sup> explained that adding contaminants to dry soil prevented the formation of a metastable soil structure by promoting a homogeneous pore fluid concentration in the sample. This study prepared the contaminated samples by adding the used engine oil to the soil with different initial (pre-contamination) moisture contents. Different amounts of water, namely 0 %, 5 %, 10 %, and 20 % by dry weight of mix, were added to the dry soil to simulate various ground *PMCs*. For the Atterberg limit and linear shrinkage tests, the mixture was kept in an air-tight plastic bag to ensure homogeneity and moisture equilibrium, particularly within the clay particles. After 24 h of curing, different percentages of used engine oil ranging from 0 to 16 % were added to the soil mixture. The selected oil contents ranged from 0 to 16 %, as per the recommendations of previous studies, thereby encompassing a range of authentic contamination scenarios.<sup>19,38</sup>

However, for mechanical tests such as UCS and  $G_{max}$  tests, the soil sample with oil content in excess of 6 % was unable to manifest the requisite dry density during the sample preparation stage. This was a consequence of the excessive oil content, which filled the soil voids as compared to the wet soil conditions.<sup>5</sup> In addition, Al-Sanad, Eid, and Ismael<sup>24</sup> reported that excessive oil contamination could cause the qualities of the compaction curve to be lost. Additionally, extra crude oil during the compaction tests caused the samples to drain out of the compaction mold. Thus, the adopted oil content was capped at 16 % because further addition of oil beyond this limit resulted in highly saturated samples with oil being squeezed out during sample preparation, in line with the observation made by other researchers.<sup>7,38</sup> The amount of added oil was calculated based on the dry weight of the soil. The specimens were kept in an air-tight plastic bag for an extra seven days. This ensures the uniform distribution of pore water and oil in the soil matrix for the Atterberg limit tests, including liquid limit (*LL*) and plastic limit (*PL*) tests and the linear shrinkage test. This is shown to be sufficient time to reach equilibrium conditions in the mixture of hydrocarbon chemicals.<sup>11</sup>

For the UCS tests, direct shear tests, and BE tests to determine small-strain shear modulus, different oil contents ranging from 0 to 6 % were added to the soil mixture with PMCs of 0 % and 5 % and optimum water content (PMC)<sub>opt</sub> obtained from compaction tests corresponding to the field's three hypothetical initial moisture conditions. The weights of the added oil and water were calculated based on the dry weight of the soil mixture. The water-clay-oil mixture was initially cured for 24 h. Subsequently, an additional amount of water was added to the mixture, as required to reach the optimum moisture content, and cured for seven days in the air-tight plastic bags. The water-clay-oil mixture was then placed in the mold and compacted in three equal layers to achieve the target maximum dry density ( $\rho_d = 1.64 \text{ t/m}^3$ ) at an optimum moisture content of 19.7 %.

It should be noted that because this study assesses the impacts of occasional oil contamination that may not be visually apparent during construction, the adopted values for maximum dry density and optimum moisture content were determined from the standard compaction test conducted on uncontaminated soil mixture per the Australian standards (AS 1289.5.1.1:2017, *Methods of Testing Soils for Engineering Purposes, Method 5.1.1: Soil Compaction and Density Tests—Determination Of The Dry Density/Moisture Content Relation of a Soil Using*  Standard Compactive Effort).<sup>39</sup> Moreover, all compacted samples were prepared at the same maximum dry density of  $\rho_d = 1.64 \text{ t/m}^3$  to ensure consistency among all the samples and assess oil contamination impacts while maintaining the void ratio constant.

### **Testing Program**

#### MICROSTRUCTURAL ANALYSIS

SEM is one of the most prominent equipment for analyzing microstructural features of soils, particularly clays, providing high-resolution images.<sup>40</sup> It provides a magnified image explaining physical and chemical properties, including the mineral's size, shape, and composition. SEM imaging was utilized in this study to examine the contaminated soil microstructure and particularly the interaction of clay, water, and oil.

#### ATTERBERG LIMITS AND LINEAR SHRINKAGE

The Atterberg limit tests, including measuring the PL and LL and conducting the linear shrinkage (LS) test, were performed on the soil with and without oil contamination. The oil contents chosen were 0 %, 2 %, 4 %, 8 %, and 16 % in conjunction with four different initial moisture contents of 0 %, 5 %, 10 %, and 20 % inspired by other researchers to cover a range of contamination levels that may be observed in the field.<sup>5,19</sup> In this study, the PL test was conducted following the Australian standard AS 1289.3.2.1-2009, Methods of Testing Soils for Engineering Purposes, Method 3.2.1: Soil Classification Tests - Determination of the Plastic Limit of a Soil.<sup>41</sup> The PL was measured by a classical test apparatus including a glass plate, a 3-mm steel rod for thickness control of the sample, and small tins to quantify the moisture content. The LL test was conducted per the Australian Standard AS 1289.3.9.1:2015, Methods of Testing Soils for Engineering Purposes, Method 3.9.1: Soil Classification Tests - Determination of the Cone Liquid Limit of a Soil,<sup>42</sup> where the cone penetrometer was used and the penetration depth was recorded. The LL of the sample was determined at the moisture content corresponding to a cone penetration of 20 mm, at which the soil state changed from plastic to liquid. Herrick and Jones<sup>43</sup> suggested that the dependency of the LL test results on the test operator is expected to be negligible, which is the advantage of this method compared to the Casagrande device. For the LS test, the mixture at the LL was placed in a 250-mm length steel bar and subsequently placed in the oven at 105°C. Following the Australian standards AS 1289.3.4.1-2008, Methods of Testing Soils for Engineering Purposes, Method 3.4.1: Soil Classification Tests - Determination of the Linear Shrinkage of a Soil,<sup>44</sup> LS was determined by the ratio of the longitudinal shrinkage (i.e., change in the length) to the original length of the soil in the standard bar. Indeed, the results from the LS test can contribute to understanding the volume-change response of the oilcontaminated soil as a result of changes in moisture content.

#### UCS

The soil's *UCS* shows the maximum compressive stress that an unconfined specimen can sustain. Because of its economical and practical advantages, the *UCS* test is widely performed to measure soil strength. Moreover, Young's modulus (*E*) can be estimated using the stress-strain curve obtained from the *UCS* test. In this study, a series of *UCS* tests were conducted following the Australian standard AS 5101.4-2008, *Methods for Preparation and Testing of Stabilized Materials, Method 4: Unconfined Compressive Strength of Compacted Materials*,<sup>45</sup> with a constant compression rate of 1 mm/min on the contaminated sample with oil contents of 0 %, 1 %, 2 %, 4 %, and 6 % and *PMCs* of 0 % and 5 % and optimum moisture content (PMC)<sub>opt</sub> (i.e., 19.7 %). It should be noted that the dry density of all samples for *UCS* tests was maintained at 1.64 g/cm<sup>3</sup> by compacting the soil in three equal layers in a cylindrical mold with a height of 100 mm and diameter of 50 mm. During the sample preparation, it was observed that the soil sample with oil contents above 6 % could not achieve the required dry density because of excessive oil contents occupying the voids within the soil matrix, similar to the observations made by Khamehchiyan et al.<sup>5</sup>

#### SMALL-STRAIN SHEAR MODULUS (Gmax) OF THE OIL-CONTAMINATED CLAY

The small-strain shear modulus  $(G_{\text{max}})$  is one of the most significant and frequently used soil characteristics in geotechnical engineering, particularly during geophysical testing or when assessing the seismic or vibration

characteristics of the ground.<sup>46</sup> Among various laboratory methods to determine  $G_{max}$ , the BE method is highly capable, fast, and accurate.<sup>46</sup> In this study, BEs made from piezoelectric ceramic and embedded in the base pedestal were employed to estimate the shear wave velocity of the soil specimen. The BE test was performed by applying an excitation voltage from one element, sending a shear wave to travel within the soil sample, while the other element received the transmitted signal. The shear wave velocity was calculated based on the recorded travel time and the distance between the two elements. Equation (1) was used to calculate the soil's small-strain shear modulus (G<sub>max</sub>), considering the measured shear wave velocity and soil density.

$$G_{\max} = \rho V_s^2 \tag{1}$$

where  $\rho$  is the soil mass density (kg/m<sup>3</sup>) and V<sub>s</sub> is the shear wave velocity of the soil sample (m/s). BE tests were conducted on the compacted cylindrical soil samples (H = 100 mm and D = 50 mm) with various oil contents (i.e., 0-6 %) before conducting the UCS tests.

#### DIRECT SHEAR TESTS

Following the Australian standard AS 1289.6.2.2:2020, Methods of Testing Soils for Engineering Purposes, Method 6.2.2: Soil Strength and Consolidation Tests - Determination of Shear Strength of a Soil - Direct Shear Test Using a Shear Box,<sup>47</sup> the direct shear tests were conducted on the soil samples to investigate the impacts of the oil contamination on the shear strength parameters, namely friction angle and cohesion. Like the UCS tests, samples for the direct shear test were prepared with different oil contents (0-6 %) and different PMCs (i.e., PMC equal to 0 % and 5 % and optimum moisture content (PMC)<sub>opt</sub>). The compacted samples were placed in a 100 mm× 100 mm  $\times$  20 mm shear box and subjected to a shearing rate of 1 mm/min until failure.

#### AST

In this paper, an automatically controlled and executed AST was performed to study the rheological properties of the contaminated soil. Anton Paar Modular Compact Rheometer (MCR 302) and RheoCompass software were utilized to apply an oscillatory shear with a frequency of 0.5 Hz at an increasing strain amplitude from 0.001 to 100 %. Figure 3 shows the setup of the rheometer with the measuring plates. ASTs are used to measure the effect of oil contamination on the structure of the material. Storage (elastic) modulus (G') and loss (viscous) modulus (G'') are the two main parameters used in the test. The storage modulus is a measure of stored deformation energy in the

FIG. 3 (A) Rheometer (MCR302) test setup used in this study. (B) The sample is placed in the bottom measuring plate while the top measuring plate is lowered, (C) and the top measuring plate is raised after the test.



Resting time	30 s before test
Plate distance	3 mm
Shear deformation	0.001 to 100 %
Frequency	0.5 Hz
Measuring points	30 points, 4 s per point

 TABLE 3

 Settings details of the amplitude sweep test conducted in this study

system, while the loss modulus is a measure of the lost deformation energy. G' captures the elastic characteristics, while G'' captures the viscous characteristics of the sample.<sup>48</sup> The AST helps determine the linear viscoelastic (LVE) range of the sample and its length. This allows for a better understanding of the sample structural stability and initial movements. When a higher LVE state is detected, the sample is identified as a liquid.<sup>49</sup> The ASTs were conducted on samples with a *PMC* of 0 % and 5 % and with differing oil contents, i.e., 0 %, 4 %, and 16 %. The samples were prepared similarly as in the Atterberg limit tests mentioned earlier. A small amount of contaminated soil after curing was placed on the measuring plate of the rheometer. Subsequently, a profiled parallel plate measuring system was automatically lowered to reach the required plate distance. According to Markgraf, Horn, and Peth,<sup>49</sup> the choice of the distance between measuring plates depends on the texture of the material and a 1–2 mm gap is suitable for fine-grained soils with particle size  $\leq 2 \mu m$ , while a distance of 3–4 mm is recommended for a coarse-grain soil. A plate distance of 3 mm was chosen in this study because of the sample composition. Furthermore, before conducting the AST, a 30-second resting time was given to reduce the sample's disturbance effect on the measuring plate.<sup>49</sup> Further setting details of the AST are summarized in **Table 3**.

### **Results and Discussion**

#### MICROSTRUCTURAL ANALYSIS

The microstructural analysis results of the soil, which is composed of kaolin, bentonite, and sand mixture with various initial moisture contents and contamination levels, are depicted in **figure 4** with magnifications of 1,000 times (=  $10 \mu m$ ). Different degrees of agglomeration of particles can be observed, where bigger units were detected for the cases with higher contamination levels. **Figure 4***A* illustrates the microstructure of uncontaminated soil with 0 % *PMC* where the minerals are evident as flake-shaped particles. Although the interparticle forces tend to bring the particles closer, dispersed structure and distribution of particles were observed. As shown in **figure 4***A*, the particles were conglomerated into large domains and clusters because of the presence of water and corresponding high surface tension, bipolarity, and lower viscosity.<sup>16</sup>

However, the dispersion of the overall texture of the clay remained quite evident. On the contrary, the particles of oil-contaminated soil were organized more loosely because of their flocculation into larger units called domains,<sup>16</sup> as shown in **figure 4B**, compared to the uncontaminated soil (i.e., **fig. 4A**). More specifically, the volume of the particles was enhanced through the formation of an oil film on the particle surface, which promoted domain development. Because of high viscosity and low bipolarity, it had an insulating effect, but it divided the domains and hindered an uninterrupted texture.<sup>50,51</sup> Indeed, oil formed a thin film covering the clay particles. Compared to the observation of Ahmed, Abduljauwad, and Akram,<sup>50</sup> the negligible bipolarity of oil in conjunction with high viscosity has led to the decoupling of the individual subdomains, and a discontinuous texture was created, as evident in **figure 4B** and **4C**. This phenomenon can be seen more visibly and to a greater extent in **figure 4D** and **4E**, where the clay sample was highly contaminated (i.e., 16 % oil). **Figure 4C** and **4E** showed the combined effects of water and oil on clay texture within a highly contaminated soil sample. In these cases, the soil began to undergo flocculation and produced a highly compacted cluster. The clay manifested lumpy features at this level and started to be structurally similar to granular soil.

Mitchell and Soga<sup>26</sup> argued that the double layer becomes thinner when the dielectric constant of pore fluid declines, resulting in growing closeness between particles and their mutual absorption to give rise to a flocculated

FIG. 4 SEM images of (*A*) uncontaminated kaolin-bentonite-sand, (*B*) 4 % oil content with 0 % *PMC*, (*C*) 4 % oil content with 5 % *PMC*, (*D*) 16 % oil content with 0 % *PMC*, and (*E*) 16 % oil content with 5 % *PMC*.





structure (refer to **fig. 4***B* and **4***D*). **Figure 4***B* and **4***E* shows that adding water to the clay with oil contamination causes the breakdown of the bond between oil and clay, replacing the oil films encircling the particles and merging separate domains into uninterrupted clusters.<sup>51</sup> Such behavior has been legitimized in earlier research because water has a higher dielectric constant and bipolarity than other liquids.<sup>50</sup> The flocculation only ceases when there is equivalence between the attraction forces of the particles and their weight.<sup>27</sup> Subsequently, separation of the

continuous flocculated particles occurs, and the contaminated clay starts to look like clean wet soil structurally. **Figure 4***E* illustrates how the kaolinite material was affected by water content of 5 %. The clay remained relatively scattered structurally, even though the kaolinite particles were merged into distinct clusters by the surface tension of water. **Figure 4***D* presents soil fabric with 16 % contamination at the microscopic level. It is apparent that the separate kaolinite particles were covered by the used engine oil, forming a fabric of relative flocculation. The collective impact of water and oil on the kaolinite structure is revealed in **figure 4***E*. The oil enhanced the ability of water to produce a flocculated structure for kaolinite. Therefore, the microstructural properties are heavily impacted by oil content, which decreased the thickness of the DDL and caused the flocculation of clay particles. As the microstructural images in **figure 4** show, a decrease in the dielectric content results in a reduction in the DDL, which has led to the specimen behaving similar to a granular material or a material without cohesion.

#### ATTERBERG LIMITS AND LS

**Figure 5***A* shows the effect of contamination on the *LL* of soil with various *PMCs*. Regardless of the soil precontamination (or initial) moisture condition, the *LL* continuously declined with increased oil contents. For example, there was a 37 % reduction in the *LL* of oil-contaminated soil with PMC = 10 % when oil content increased from 0 to 16 %. Similarly, when the oil content of contaminated soil with PMC = 20 % increased from 0 to 16 %, the *LL* decreased from 71.2 to 51.9 %. **Figure 5***B* shows the relationship between the *LL* and the *PMC* ratio (i.e., *PMC/PI*). It indicates that the *LL* was reduced with increased *PMC* and the oil content. Moreover, the *LL* of oil-contaminated soil was reduced with an increase in *PMC/PI* up to the optimum point where a further increase in *PMC/PI* resulted in a slight variation of the *LL*. For example, the *LL* of contaminated soil with 8 % oil content was reduced from 64 to 59 % when the *PMC* ratio (i.e., *PMC/PI*) increased from 0 to an optimum point of 0.135. As observed in **figure 4***E*, the impact of oil content on the *LL* of the contaminated soil was more significant than the corresponding effect of *PMC*. This manifests the importance of the ground's *PMC* on the soil's liquidity when subjected to oil contamination.

The observed impact of *PMC* and contaminating oil content on the *LL* was because of the increase in the presence of contaminant fluid, deteriorating the thickness of the DDL between the soil particles. Furthermore, it is shown that the *LL* depends on the physicochemical factors (i.e., DDL) rather than mechanical factors, as also observed by other researchers (e.g., Omar, Fatahi, and Nguyen).<sup>38</sup>. With the reduction of the dielectric constant among clay particles, which decreased the *LL*, the sample tended to behave more like a granular or cohesionless material, as can be seen in the microstructural image reported in figure 4.



FIG. 5 (A) Effect of oil contents on the LL and (B) effect of normalized PMC / PI on the LL.

**Figure 6.4** captures the variations of the *PL* with oil content for various *PMCs*. As evident in these figures, the addition of contaminating oil resulted in a higher *PL*. Referring to **figure 6.4**, when oil content increased from 0 to 16 %, a 28 % increase in the *PL* of contaminated soil with *PMC* = 10 % was observed. Additionally, **figure 6.8** shows an increase in the *PL* of contaminated soil when the *PMC* ratio (i.e., *PMC/PI*) increased up to its optimum point, beyond which a slight decrease in the *PL* was observed. For instance, when *PMC/PI* increased from 0 to an optimum value of  $(PMC/PI)_{opt} = 0$ . 185, contaminated soil with 16 % oil content experienced up to a 25 % increment in *PLs* before decreasing gradually. The increase of the *PL*, which resembles the plasticity of the clay, is controlled by the adsorbed water in the DDL, whereas the nonpolar oil molecules prevent the water from reaching the DDL. Thus, the soil required more water to attain the plastic state, which resulted in the rise of the *PL* with the addition of used engine oil (**fig. 6.4**).

Following the results reported for the *LL* and *PL*, the plasticity index (*PI*) of the clay samples contaminated with various oil contents were determined and shown in figure 7. As observed in figure 7A, the *PI* of the contaminated soil was reduced with the oil content. For instance, a 48 % reduction (i.e., from 51.96 to 27 %) in the





*PI* of oil-contaminated soil with PMC = 5 % was observed when oil content increased from 0 to 16 %. According to Kermani and Ebadi,<sup>19</sup> the double-layer water influences the plasticity of oil-contaminated soil because of the dipolar nature of water; wherein there are positive and negative charges in the water molecules, which means that the water molecules are attracted to the hydrogen atoms on the surface of clay particles. Thus, a hydrogen bond is created with the clay particles. As previously mentioned, the clay is encompassed within the oil. Therefore, a further quantity of water is required to prompt the clay to demonstrate its plastic properties. Hence, high oil content correlates with diminished *PI*.<sup>38</sup>

The *PI* is determined by physicochemical factors (the DDL) and not mechanical factors.<sup>25</sup> Double-layer water is considered as the main contributor to the clayey soil plastic behavior and is electrically absorbed into the clay particle surface.<sup>16</sup> When oil contamination occurs, the clay becomes more like granular material because of its low dielectric constant value, decreasing the *PI*. Furthermore, dielectric constant changes of pore fluid might impact the double-layer thickness that forms in polluted soils.<sup>26</sup>

As the crude oil dielectric constant (ranges from 2 to 2.2) is less than that of water (80), the addition of oil to the soil thins the double layer, reducing soil cohesiveness.<sup>13</sup> Moreover, as reported in **figure 7B**, the contaminated soil with 16 % oil content showed a reduction in *PI* from 32.7 to 27 % when *PMC* increased from 0 to 5 %, correspondingly. It should be noted that the optimum *PMC* ratio  $(PMC/PI)_{opt}$ , which was in the range of 0.1–0.2, corresponded to *PMC* = 5 %, where a further increase in *PMC* resulted in an insignificant change of *PI*, as shown in **figure 7B**.

The results of the *LS* tests conducted on the oil-contaminated clay are reported in **figure 8**. The *LS* of the oilcontaminated sample increased with an increasing level of contamination. The slight addition of the oil content (e.g., 2 %) resulted in a sudden increase in *LS*. For example, the addition of 2 % oil to the sample with *PMC* = 0 % increased the *LS* from 9 to 16.5 %, while a reduction from 16.5 to 10.5 % was observed for *LS* with further addition of oil. Hence, the physical properties were heavily impacted by the addition of oil, because of the decrease in the DDL thickness causing flocculation of clay particles, resulting in reductions in *LL* and increases in *PL*, as evident in this study (see **figs. 5** and **6**).

#### UCS

The unconfined stress-strain responses of the oil-contaminated samples with different oil contents are presented in **figure 9**. The stresses mobilized in the soil skeleton increased to the peak  $(q_{\text{max}})$  and gradually decreased with increasing axial strain ( $\varepsilon$ ), indicating the strain softening. The  $q_{\text{max}} = 60.25$  kPa of the uncontaminated sample was recorded at  $\varepsilon = 0.065\%$ , while there was a significant loss of approximately 50 % in  $q_{\text{max}}$  for contaminated



FIG. 8 (A) Effect of oil contents on the LS and (B) effect of normalized PMC / PI on the LS.



FIG. 9 Effect of oil content on the stress-strain response of contaminated soil mixture with (A) PMC = 0 %, (B) PMC = 5 %, and (C) (PMC)<sub>oot</sub> = 19.7 %.

samples with 6 % oil content. However, the reduction rate in the peak strength depended on the *PMC* of the soil. As shown in **figure 9A**, samples with PMC = 0 % showed a dramatic drop in the peak strength compared to the samples with higher *PMC*.

Furthermore, the summary of the UCS of the compacted soil is depicted in figure 104. It can be observed that the strength of the contaminated high-plasticity clay was significantly reduced by more than 54 % when 6 % engine oil was added to the soil. For example, because of addition of oil, the UCS of samples with PMC = 0 % and 5 % decreased by 64 % (from 60.25 to 21.51 kPa) and 54.2 % (from 60.55 to 27.7 kPa), respectively. Furthermore, for samples with (PMC)<sub>opt</sub>, an approximately 60 % reduction was noted in the UCS when increasing oil content up to 6 %. (i.e., reduction from 60.85 to 24.65 kPa). The reduction in the UCS for the contaminated samples with PMC = 0 % was more significant compared to contaminated samples with PMC = 5 % and (PMC)<sub>opt</sub> in this study. For example, for soil contaminated with 6 % oil, a roughly 22 % reduction (i.e., from 27.72 to 21.51 kPa) in the UCS was measured for samples with PMC = 5 % and 0 %, respectively. This reduction in the UCS of samples is because of the addition of oil to the soil, which enhances the extent of pore fluid saturation, which can weaken the soil.<sup>13</sup> The reduction in the pore fluid dielectric constant was identified as the cause for reducing the shear strength of soil samples with organic chemical contamination.<sup>6</sup>

FIG. 10 Effect of oil content on the (A) UCS of the contaminated soil mixture, (B) Young's modulus of the contaminated soil mixture, and (C) small-strain shear modulus of contaminated soil mixture.



Consequently, foreign organic contaminants may penetrate the soil and modify its fabric. The pore fluid also affects mechanical factors like interparticle friction significantly. For instance, one study reported that oil contamination resulted in a reduction in the friction angle of the soil.<sup>52</sup> In reality, soil friction decreases when the pore fluid becomes more viscous because the pore fluid serves as a sort of interparticle lubricant.

Using the stress-strain curves in **figure 9**, Young's modulus could be determined by adopting the linear regression over the initial linear section of the stress-strain curve, where an elastic behavior was evident. As depicted in **figure 10***B*, a significant drop of about 70 % in Young's modulus was observed in all samples because of the addition of 6 % oil. Although Young's modulus of all samples converged at approximately 0.7 MPa, the samples with PMC = 0 % exhibited a highly sensitive response with the presence of oil, evident by a 45 % drop in stiffness with the addition of 1 % used engine oil, as shown in **figure 10***B*. It has been noticed that the reason for having a lower *UCS* for samples with PMC = 0 % compared to PMC = 5 % and  $(PMC)_{opt}$  can be because of the oil covering the soil particle surfaces. This can be explained by the impact of oil coating the soil particles, which reduces the specific surface area (SSA) because of the aggregation of clay soil particles.<sup>19</sup> Evidence has also been produced regarding a marked decrease in clay cation exchange capacity (CEC) because of oil addition.<sup>50</sup> The SSA decrease is reflected in the soil CEC decrease. In turn, SSA decrease causes

fewer water molecules to be assimilated in the clay particles and lower clay particle contact to diminish cohesion.

Moreover, soil cohesion increases by the addition of moisture to the soil, which increases the proportion of water molecules binding to the clay particle surface, leading to a proliferation of bonds between water molecules and various clay minerals. Nevertheless, cohesion is less influenced by moisture at higher levels of contamination, as also observed in previous research studies.<sup>19</sup> Furthermore, as the total strain energy of the soil samples can be determined using the area under the axial stress-strain relationship (e.g., fig. 9), the addition of oil decreased the absorbed energy as a significant portion of the applied energy was transferred to the increased free water, rather than the soil structure. Thus, the effect of oil contamination has a deteriorating impact on the mechanical properties of oil-contaminated soil.

#### SMALL-STRAIN SHEAR MODULUS (Gmax) OF THE OIL-CONTAMINATED CLAY

The results of the BE test are reported in figure 10C. A progressive decrease of the small-strain shear modulus was detected because of the addition of contaminated oil. For the sample with PMC = 0 %,  $G_{max}$  experienced a 7 % reduction as oil content increased from 0 to 2 %, while a 22.5 % reduction was observed when oil content increased from 2 to 6 %. The extent of reduction in  $G_{\text{max}}$  for the samples with PMC = 5 % was relatively moderate for up to 4 % for oil contents. However, there was a significant drop in the small-strain shear modulus with further oil contamination (i.e., 6 % oil content). For instance, referring to figure 10C, for  $PMC = (PMC)_{opt}$ ,  $G_{max}$  was reduced by 15 % as oil content increased from 4 to 6 %. The contaminated samples with an optimum PMC exhibited a higher small-strain shear stiffness than the samples with PMC = 0 %. For example, for a soil sample with 4 % oil content,  $G_{\text{max}} = 29.05$  MPa was observed for (PWC)<sub>opt</sub>, while  $G_{\text{max}} = 27.17$  MPa was measured for PMC = 0 %. Moreover, the repulsive forces of the double layer and van der Waals attractive forces exist between two individual clay particles.<sup>53</sup> The former is degraded by the interference of the oil particles, which reduces the dielectric constant. Subsequently, this causes the clay particles to move closer to each other. Hence, larger clay aggregates associated with larger voids in the pore space were formed (e.g., fig. 4C and 4E). Moreover, replacing the adsorbed water by the contaminating oil increases the free water in the soil structure, and subsequently, more conductive soil is achieved. The increased hydraulic conductivity and lubricating effect because of the addition of oil led to less frictional resistance between soil particles and, therefore, a lower  $G_{\text{max}}$  was observed in figure 10C.

#### DIRECT SHEAR TESTS

The variations of internal friction angle and cohesion of the samples are reported in figure 11. An inverse correlation between the shear strength parameters and the level of contamination was observed. It is known that the shear strength of granular materials reduces when the viscosity of the pore fluid increases.<sup>14</sup> Referring to figure 114, there was a 62 % reduction in the friction angle reported for soil with PMC = 0 % as the oil content increased from 0 to 6 %, whereas the samples with PMC = 5 % experienced a smaller reduction in friction angle (i.e., 44 %) for the same increase in the oil content. Moreover, a significant drop of 80 % in friction angle was observed for the samples with (PMC)<sub>opt</sub> because of the addition of oil, consistent with the observation made by Ahmadi, Ebadi, and Maknoon.<sup>52</sup> Indeed, the pore spaces within the fabric of the fine-grained soil are filled with pore fluid and a thin fluid layer covering the minerals. Increased pore fluid viscosity changes the characteristics at the interface between the mineral and the pore fluid, leading to softening behavior.<sup>14</sup> On the other hand, figure 11B shows that for samples with PMC = 0 %, a 60 % reduction (from 29.2 to 11 kPa) in cohesion was noted when the engine oil contamination level increased from 0 to 6 %, while for the sample with PMC = 5 %, a 54 % reduction in cohesion was observed. Furthermore, a lower reduction in cohesion (i.e., by 52.7 %.) was noted in the samples with  $(PMC)_{opt}$  as compared to the sample with PMC = 0 % and 5 % (see fig. 11B). Referring to Ahmed, Abduljauwad, and Akram,<sup>50</sup> the CEC between clay particles can be diminished because of the presence of the oil particles.

FIG. 11 (A) Effect of oil content on the friction angle of contaminated soil mixture and (B) effect of oil content on the cohesion of contaminated soil mixture.



Moreover, the plasticity of fine-grained soils is governed by the surface tension from the water molecules within the DDL. The displacement of water molecules by the oil molecules led to a reduction in cohesion because of the lower surface tension of the oil molecules. Indeed, the specific area of the clay-oil interface was reduced because of the aggregation of clay-oil particles, thus reducing the cohesion of the clayey soil. It has been noticed that the reason for having a lower friction angle and cohesion for samples with PMC = 0 % compared to PMC = 5 % and  $(PMC)_{opt}$  can be because of the oil coating the soil. Thus, adding oil to the sample with PMC = 0 % may prevent water from getting in contact with soil. Coating the clay particles with oil prevents water molecules from reaching the double-layer water so that a more significant amount of water is required for the soil to achieve plastic properties. The water present in the pore space that is not absorbed by the clay particles and can move unhindered within the soil is known as free water, which impacts the liquid condition of the soil. The assimilated water on the clay particle surface was substituted by the added oil so that the soil structure had a higher content of free water and the interparticle friction resistance was diminished. Another consequence is that soil hydraulic conductivity is enhanced.<sup>11</sup> The addition of water beyond a specific quantity results in an inordinately high volume of free water and enlarges the interparticle space, thus having a negative effect on cohesion. Furthermore, increasing the water quantity to saturation eliminates capillary suction, and the apparent cohesion diminishes.

#### AST

The viscoelastic behavior of soils can be described using a Kelvin-Voigt model, which is widely used for solids simultaneously exhibiting elastic and viscous behaviors.<sup>49</sup> In an AST where an oscillatory shear is applied to the sample, the shear modulus (*G*) for an ideal elastic material derived from Hooke's law turns into a complex dynamic modulus (*G*<sup>\*</sup>) to account for both elastic and viscoplastic behaviors.<sup>49</sup> There are two components in *G*<sup>\*</sup>: an elastic component presented by a storage modulus (*G*') and an imaginary loss modulus (*G*") representing the viscous behavior of the sample. The ratio between *G*" and *G*' is defined as the loss factor (tan  $\delta = G''/G'$ ). When tan  $\delta < 1$  (i.e., G' > G''), the elastic component of the material predominates, while a viscous flow occurs when tan  $\delta > 1$  (i.e., G'' > G'). At tan  $\delta = 1$  (i.e., G' = G''), a yield point is reached where further increase in strain results in plastic deformation. Figure 12 shows a typical result of the AST conducted on clay with *PMC* = 0 % and 4 % oil content. As shear strain ( $\gamma$ ) increased to 0.1 %, *G*' and *G*" approached an LVE limit, where the shear strain within this range is elastic and fully recoverable. Beyond the LVE limit, the sample entered into

#### FIG. 12

A typical result of an AST on oil-contaminated clay.



a pre-yielding stage where G' and G'' started to reduce with increasing shear strain. The crossover of G' and G'' occurred at  $\gamma = 3.4$  %, indicating that the sample was at the yield point where the breakdown of soil structure began because of plastic deformation with increasing strain.

**Figure 13** summarizes the results from an AST conducted on soil mixture with PMC = 0 % and 5 % and contaminated with 0 %, 4 %, and 16 % used engine oil. The black square and circle symbols represent the storage modulus (G'), while the loss modulus (G'') for 0 % and 5 % *PMC* was shown by the red square and circle symbols. Oil-contaminated clay with PMC = 5 % showed an increase in storage modulus G' and loss modulus G'' with increasing oil content at the LVE limit. In contrast, there was a decrease of 37 % in the initial storage modulus ( $G'_{\text{initial}}$ ) and 13 % in G' at the LVE limit (i.e.,  $G'_{\text{LVE}}$ ) when the oil content increased from 4 to 16 % for samples with 0 % *PMC*. A similar observation was made when the yield point was reached (G' = G''). Hence, referring to figure 13, it was observed that increasing levels of oil contamination expanded the range in which the sample behaved elastically. Moreover, it has shown that samples with PMC = 5 % have higher values of G' than samples with *PMC* = 0 %. This means the sample behaves more elastically when it is in a wet condition before contamination.

The plots of loss factor (tan  $\delta$ ) against the shear strain ( $\gamma$ ) are presented in **figure 14** to compare samples with different *PMCs*. The yield points occurred at the shear strain of  $\gamma = 1.4-3.4$  %, where the loss factor curves crossed the tan  $\delta = 1$  line. As evident in **figure 15**, as 16 % of used engine oil was added to the sample, a contrasting behavior was observed where the sample with *PMC* = 5 % showed a lower shear strain at yield (i.e.,  $\gamma = 1.4$  %) as compared to  $\gamma = 2.6$  % corresponding to the sample with *PMC* = 0 %. This may suggest a reduction in the stability of the soil structure because of a higher *PMC* and the presence of oil in pores, which has a high viscosity and acts as a lubricant, leading to various changes between soil particles and pore fluid, causing softening behavior.

#### DISCUSSION ON THE STABILITY OF MICROSTRUCTURE

An important rheological parameter to quantify the stability of the soil structure is the Integral Z ( $I_z$ ), which is highly dependent on the shear strain at yield. Integral Z is determined by calculating the area between tan  $\delta$  and the line tan  $\delta = 1$  from the initial shear strain ( $\gamma$ ) to the yield point. Markgraf, Horn, and Peth<sup>49</sup> described the

#### FIG. 13

Results from an AST on oil-contaminated soil in this study, namely G' and G" at: (A) initial state, (B) LVE limit, and (C) yield.



integral Z as a semi-quantifiable parameter to measure materials' elastic behavior or rigidity. At a higher value of  $I_z$ , the soil exhibits higher elasticity and greater structural stability at the microscale.<sup>49</sup> The calculated  $I_z$  of the contaminated samples are summarized in **figure 15A**. The sample with *PMC* = 0 and 4 % oil content showed the highest value of  $I_z = 1.3$ , followed by a drop in  $I_z$  with further addition of oil (e.g.,  $I_z = 0.9$  for 16 % oil content).

Similarly,  $I_z$  of soil with PMC = 5 % was reduced with an increasing amount of oil; indeed, a drop of 44 % was observed when oil content increased from 4 to 16 %. Furthermore, as shown in **figure 15**, the  $I_z$  and the shear strain at yield follow a similar pattern where the shear strain is reduced with the addition of oil. A reduction of 42 % in a shear strain for soil with PMC = 5 % was observed when oil content increased from 4 to 16 %. Hence,  $I_z$  was highly dependent on the shear strain values at yield as both  $I_z$  and  $\gamma_{yield}$  capture properties of the material in the elastic range. Furthermore, the higher addition of oil resulted in a reduction in the stability of the soil because of the increase in viscosity of the soil mixture. Indeed, hydrocarbon from the oil interacted with clay particles and reduced the direct contact between clay particles, resulting in lower friction and higher slippage. Moreover, the effect of oil contamination might have increased the thickness of the diffused double layer and particle dispersion within the soil matrix. Consequently, the stability of the microstructure is reduced as similar to the observation reported in the literature.<sup>54</sup>

#### FIG. 14

Results from an AST on oil-contaminated soil in this study, namely loss factor,  $tan(\delta)$  for samples with *PMC* = 0 % and *PMC* = 5 % mixed with (*A*) 0 %, (*B*) 4 %, and (*C*) 16 % oil content.



#### FIG. 15

Sweep test results on oilcontaminated soil: (A) Integral Z and (B) shear strain at yield.



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The AST results show that the stability of microstructure of the sample with PMC = 0 % was higher than the corresponding values for PMC = 5 % at 4 % and 16 % oil contents. Indeed, the soil mixture with PMC = 0 % showed a dispersed distribution of clay particles, as shown in **figure 4B**. When the oil particles entered the soil matrix with PMC = 0 %, a thin layer of oil film was developed, covering the clay particles and subsequently forming larger flocculated clay particles through hydrogen bridges and van der Waals forces.<sup>55</sup> It should be noted that the aggregation of the soil-oil mixture contributed to a greater extent of elasticity in the soil microstructure. For the soil with PMC = 5 %, the added oil replaced the adsorbed water on the surface of clay particles, resulting in an increase of free water in the soil structure and thus reducing frictional resistance between soil particles. Subsequently, referring to **figure 15A**, for contaminated soil with PMC = 5 %, a weaker microstructure was observed compared to the sample with PMC = 0 %. Therefore, a lower PMC resulted in a wider elasticity range and greater microstructural stability of the oil-contaminated soil, which is also consistent with the findings reported by Ghezzehei and Or.<sup>56</sup>

#### DISCUSSION ON THE PRACTICAL IMPORTANCE OF THE PROPOSED STUDY

The relationship between normalized shear strength (*UCS/E*) and different *PMC* and oil contents is reported in **Table 4**. The results showed that the normalized shear strength (*UCS/E*) increased from 0.029 (without oil contamination) to 0.041 when 6 % of oil content was added to the sample with *PMC* = 5 %. Furthermore, it can be noted that the highest ratio of *UCS/E* = 0.046 was obtained from a contaminated sample with *PMC* = 5 % and 4 % oil content. With *PMC* = 0 %, the maximum value of *UCS/E*, which was 0.044, was obtained at 1 % oil content while further increases in oil content showed negligible effect on *UCS/E* ratio. Moreover, the normalized shear strength increased with increasing oil content for samples with optimum *PMC*. It should be noted that although the normalized shear strength increased by increasing the oil content, both *UCS* and *E* decreased with *E* having a higher reduction rate as compared to *UCS*. Consequently, a higher *UCS/E* ratio was observed with higher oil content for both *PMC* = 5 % and *PMC*<sub>opt</sub>.

Moreover, the variations of  $G_{\text{max}}/E$  ratio with *PMC* and oil contents are reported in **Table 5**. It was observed that the ratio  $G_{\text{max}}/E$  increased with increasing oil contents and different pre-contamination contents. Increasing

#### TABLE 4

Relationship between normalized shear strength (UCS/E) and different PMCs and oil contents

Oil, %		PMC, PMC %	
	PMC = 0 %	<i>PMC</i> = 5 %	$PMC_{\rm opt} = 19.7 \%$
0	0.03	0.029	0.029
1	0.044	0.039	0.032
2	0.039	0.042	0.033
4	0.041	0.046	0.036
6	0.036	0.041	0.039

#### TABLE 5

Relationship between ratio of shear modulus ( $G_{max}/E$ ) and different pre- PMCs and oil contents

Oil, %		PMC, PMC %	
	PMC = 0 %	<i>PMC</i> = 5 %	<i>PMC</i> <sub>opt</sub> = 19.7 %
0	16.2	16.12	16.03
1	27.94	24.17	19.97
2	29.63	29.20	22.73
4	35.49	37.32	31.86
6	40.5	35.98	39.60

oil content up to 6 % resulted in a 151 % increase of normalized shear modulus  $(G_{\text{max}}/E)$  (i.e., from 16.2 to 40.5) for the sample with PMC = 0 %. It should be mentioned that values of  $G_{\text{max}}$  and E decreased with oil content, while E reduced at a higher rate as compared to  $G_{\text{max}}$ . Practicing engineers can refer to these ranges (i.e.,  $G_{\text{max}}/E$  ranges from 16.2–40.5 with various oil contents) to estimate Young's modulus and the shear modulus using BE tests. This method is based on shear wave velocity or small-strain shear modulus values identified in nondestructive tests. It is important to note that engine oil migration into the soil skeleton is expected to saturate the voids, which ultimately increases soil plasticity and causes the clay to lose stiffness and strength. This is a significant issue that practicing engineers must consider when remediating and stabilizing oil-contaminated clays.

The results obtained in this study are useful in guiding further investigations related to effects of site moisture levels before contamination in order to evaluate the implications of soil contamination by used engine oil soil. Any accidental oil leaks caused by exploration, operations, and transportation before construction can cause oil contamination. Thus, understanding the impacts of oil contamination on soils is critical, particularly on the foundations of critical infrastructures such as roads, railways, and heavy machinery. The in situ moisture content of soils can vary from dry to wet based on site and environmental conditions, which can ultimately impact soil properties. Therefore, it is vital to perform experimental studies to investigate the effects of oil contamination on soft clay, and geotechnical practitioners can further enhance their understanding on how to effectively and accurately evaluate the stability of foundations and earthwork during design and construction.

In a case study performed around the Nigerian Niger Delta,<sup>57</sup> laboratory tests were carried out to assess the impacts of oil spillage on the shear strength of lateritic soil. The tests included Atterberg limits, compaction, and shear strength. The results showed that the oil covers and agglomerates the lateritic soil particles, which lowers the SSA. This enhances the bonding strength of the lateritic soil. The findings also showed that the value of the *LL* parameter decreased. The drop in *LL* values was attributed to a decrease in the soil's CEC. Moreover, the decrease in the angle of internal friction values was attributed to the building up of oil-coated films around the lateritic soil particles, which caused particles to constantly glide over each other, which in turn decreased the relative internal friction between the lateritic soil particles. However, to the best of the authors' knowledge, there is no study on the *PMC* of oil-contaminated clay. In addition, further studies are recommended to investigate the properties of oil contamination on the different types of soils.

Oil contamination has an impact on the environment, and it is critical to study its relationship with environmental remediation programs. Specifically, when there is a need to clean soil contaminated, experimental testing on soils can be conducted after cleaning to verify that the land is no longer hazardous. There have been multiple studies about the stabilization or remediation of fine-grained soils that have been subjected to oil contamination.<sup>7,25,27</sup> There is a consensus among all these studies that it is preferable to remediate contaminated soils rather than abandon them. The use of chemical additives, such as cement and lime, is a feasible solution because they can reinstate the geo-mechanical characteristics of contaminated clay soil. In recent decades, the combination of cement and lime is often employed as a stabilizer to counteract the adverse effects of soil contamination and the effects on backfill/highway pavements.<sup>22,34</sup>

### Conclusions

While significant studies have targeted the behavior of contaminated sandy soils, investigations on engineering properties of the oil-contaminated clayey soil and impact of *PMC* on the response of the soil remain very limited. In this paper, extensive laboratory tests were conducted on a high-plasticity soil composed of kaolin, bentonite, and sand mixture with different pre-contamination moisture conditions subject to different contamination levels because of the addition of used engine oil, ranging from 0 to 16 %. Variations of the plasticity characteristics, shear strength, and clay stiffness were investigated in detail. The comprehensive experimental plan consisted of micro-structural analysis by SEM, the Atterberg limit and *LS* tests, the *UCS*, the BE, the direct shear, and the ASTs. Considering the *PMC* of the soil, significant variations in the microstructural, physical, and mechanical properties of the soil were observed. It was shown that the addition of the used engine oil decreased the *LL* and the *PI* of clay,

which very much depended on the pre-contamination moisture condition. Moreover, the increase in the *PL* could be correlated with the significant decrease in the shear strength parameters and the small-strain shear modulus of the contaminated soil. In addition, the rheological properties from the AST provided an assessment for the stability of the contaminated soil. It was found that an increase in the oil content and the *PMC* had adverse impacts on the elasticity or the rigidity of the microstructure of the oil-contaminated clayey soils. This study shows that to assess the risk of adverse effects of soil contamination with used engine oil, it is essential for practicing engineers to know the pre-contamination moisture condition of the site. Dry soil (i.e., PMC = 0 %) has shown the lowest value for *UCS*,  $G_{max}$ , and shear strength. On the other hand, wet soil with PMC = 5 % showed a critical point for *LL* and *PI*, as a further increase in *PMC* had no notable impact on the Atterberg limits of the contaminated soil. Moreover, soil with the range of PMC = 5 % to  $(PMC)_{opt}$  showed the highest soil mechanical properties. Therefore, the *PMC* of the site soil is critical and can impact the severity of adverse effects of oil contamination on microstructure, physical, mechanical, and stability characteristics of the soil.

#### ACKNOWLEDGMENTS

The first author would like to thank Saudi Arabian Cultural Mission (SACM), Cultural Mission of Royal Embassy of Saudi Arabia for sponsoring this research study.

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