Geomechanics and Engineering, *Vol. 18*, *No. 2 (2019) 179-188*
DOI: https://doi.org/10.12989/gae.2019.18.2.179

Effect of palm oil on the basic geotechnical properties of kaolin

Anirudh Subramanya Sriraam[1a], Mavinakere Eshwaraiah Raghunandan*[1,4], Tey Beng Ti[2,4b] and Jayantha Kodikara[3c]

[1]Civil Engineering Discipline, School of Engineering, Monash University Malaysia, 47500 Bandar Sunway, Selangor, Malaysia
[2]Chemical Engineering Discipline, School of Engineering, Monash University Malaysia, 47500 Bandar Sunway, Selangor, Malaysia
[3]Department of Civil Engineering, Monash University, Clayton, Victoria 3800, Australia
[4]Advanced Engineering Platform, Monash University Malaysia, 47500 Bandar Sunway, Selangor, Malaysia

(Received June 2, 2018, Revised May 13, 2019, Accepted May 14, 2019)

Abstract. This paper presents an experimental study to evaluate the effect of palm oil on the selected basic physical-chemical and geotechnical properties of kaolin. The experimental findings are further compared with literature outcomes investigating similar properties of fine grained soils subjected to contamination by different types of oils. To this end, palm oil was mixed with oven dried kaolin samples–aiding oil's interaction (coating) with dry particles first, in anticipation to emphasize the effect of oil on the properties of kaolin, which would be difficult to achieve otherwise. Oil content was limited to 40% by dry weight of kaolin, supplemented at intervals of 10% from clean kaolin samples. Observations highlight physical particle-to-particle bonding resulting in the formation of pseudo-silt sized clusters due to palm oil's interaction as evinced in the particle size distribution and SEM micrographs. These clusters, aided by water repellency property of the oil coating the kaolin particles, was analyzed to show notable variations in kaolin's consistency–measured as liquid and plastic limits. Furthermore, results from compaction tests indicates contribution of oil's viscosity on the compaction behavior of kaolin – showing decrease in the maximum dry unit weight ($\gamma_{d,max}$) and optimum moisture content (w_{opt}) values with increasing oil contents, while their decrease rates were directly and inversely proportional in $\gamma_{d,max}$ and w_{opt} values with oil contents respectively. Comparative study in similar terms, also validates this lower and higher decrease rates in $\gamma_{d,max}$ and w_{opt} values of the fine grained soils respectively, when subjected to contamination by oil with higher viscosity.

Keywords: oil contamination; ground improvement; laboratory analysis; plasticity; soil behavior

1. Introduction

Rising demand for palm oil and related products over the recent decades has enthused higher production, processing, transport, and storage of crude palm oil globally. Malaysia and Indonesia are two major producers accounting to 81.7% of the total global production during 2004 - 2013 (FAO 2016, MacNeill 2016). Increased milling and post-production process for this purpose in anticipation, and following few reported cases, result in severe geoenvironmental concerns due to accidental spills, improper disposal practices, uncontrolled milling and others (Lim *et al.* 2016, Mohammadi-Sichani *et al.* 2018). Some of the recent reports of accidental palm oil spills observed during its transportation (Yu 2017) and seven other instances on the British waters (Mowat 2018) are the typical examples of such concerns. Not limiting to these, multiple cases of persistent oil spills on ground or soil at milling sites have been frequently reported in Malaysia and Indonesia (Mohammadi-Sichani *et al.* 2018, Olafisoye *et al.* 2017, Soleimaninanadegani and Manshad 2014). One such possible case of contamination in mills is during the processing and/or disposal of the palm oil mill effluent (POME). POME contains an estimated 500-700 mg/l of residual palm oil (MPOB 2017, Olafisoye *et al.* 2017), which most likely accumulates in the soil's structure–either by physically adhering to the soil particles and/or accumulating within the voids, when spilled or contained on soils. Palm oil spills on the ground surface hypothetically affects the soil's surface characteristics aided by the impermeable film of oil formed, which limits, sometimes curtails the interaction of surface water with soil. This convincingly suggests another likely case wherein the presence of oil within the soil matrix potentially alters the water-soil interaction in the soil mass.

Literature over the past decade evince the influence of oil, generally referred to as oil contamination, on this water-soil interaction, where the interaction is commonly measured in terms of varying basic physical-chemical properties of the soils. These include, but not limited to visual identifications, thermal behavior, micro-structure or texture, particle size distribution, consistency limits, and compaction behaviors. Soils generally favor attraction to both hydrophilic and hydrophobic pore fluids, mainly due to their negatively charged surface characteristics (Żbik and Horn 2003). These characteristics though justifies the physical adhesion of oil onto the dry soil particles, the same behavior with moist particle surface is rarely encountered

*Corresponding author, Senior Lecturer
E-mail: mavinakere.raghunandan@monash.edu
[a]Former Ph.D. Student
[b]Professor
[c]Professor

http://www.techno-press.org/?journal=gae&subpage=7
ISSN: 2005-307X (Print), 2092-6219 (Online)

(Rimmer *et al.* 1996). Which basically demarks the process of oil's interaction with wet or moist soil particles due to water's hydrophobic nature which is believed to further dictate the complex soil particle-water-oil interaction.

Particle size distribution has been predominantly used as a tool to study the effect of increasing oil on the gradation of the soil samples (Chang and Cho 2014, Trzciński *et al.* 2015). Notwithstanding this, the results also highlight the clogging of pore spaces due to excessive oil accumulation, despite varying initial packing and particulate structure. The presence of oil within the pore space, particularly in clays and fine-grained soils, result in large pseudo-silt sized particles (for example see Ijimdiya 2012). The formation of these pseudo-silt sized particles in clays in presence of oil can be related to the depleting double layer thickness due to the relatively lower dielectric constant of oils (Mitchell and Soga 2005). This increase in particle size results in increased pore diameter altering the density and the porous structures. Discussion on the standard compaction behavior in the literature show decreasing trends of the maximum dry density of clayey soils with increasing oil content due to additional lubrication offered by the oil (Meegoda *et al.* 1998; Nasehi *et al.* 2016; Saberian and Khabiri 2018). This lubrication effect is believed to aid in improving the particle packing via the particle-particle attraction because of the surface adhesive van der Waals forces (Lai *et al.* 2015). Furthermore, oil introduces soil-water repellency causing reduction in the interfacial tension allowing particles to slide over each other (Takawira *et al.* 2014). This perhaps justifies the decreasing amount of water required to reach the maximum dry density despite significant reduction in the fraction of available pore space within the compacted soil samples.

On the other hand, utilization of various industrial wastes and by-products to ensure eco-friendly utilization of these wastes and/or their effect on the basic soil properties has been the key focus of various research project recently (example Yu *et al.* 2016; Mohanty *et al.* 2017). In general, the consistency limits (or Atterberg limits) are one amongst the basic soil properties which have been traditionally used to assess the soil-water interaction, based on the consistency of soil. Literature present such attempts to also discuss the effect of oil content by utilizing the Atterberg limits as a tool. As examples, Khosravi *et al.* (2013) studied the effect of gas oil contamination on kaolin which reported an inverted parabola-shaped trend in the liquid limit (w_L) curves with increasing oil content. Similar trends were reported by other researchers including Kermani and Ebadi (2012), Khamehchiyan *et al.* (2007), Ijimdiya (2012) and Rahman *et al.* (2010) mainly focusing on the field samples contaminated with crude, gas and motor oils respectively. Contrarily however, the findings presented by Pandey and Bind (2014) showed a gradual decrease in w_L with increasing engine oil contents, unlike the parabolic trends. Authors in general, justify these trends as a result of oil, a non-polarizing fluid, eventually causing reduction in the thickness of water film around the clay minerals. Similar behavior or trends were reported for the plastic limits (w_p) with increasing oil contents. Literature discussing on the shrinkage limit (w_s) however were very scare. Nonetheless, inconsistent results encountered at various instances during the literature review cautioned our limited understanding of the oil's influence on the behavior of clays, particularly with changing oil amount, type, and properties. Moreover, only fewer literatures investigate the effect of palm oil on the engineering properties of clays, with not all the crucial properties required to explain the water-oil-soil interactions being discussed in detail. This therefore led to the research question–which trend and behavior available from the literature suffice to explain the behavior of clays supplemented with palm oil? Should a similar inverted parabolic trend be observed in the properties of clay with palm oil, what is the oil content associated with this inflexion point?

On the other hand, rapid developments in infrastructure and connectivity to the palm plantations or mills, and other potential sites exposed to increasing palm oil contents, perhaps mandates for a detailed and engineered understanding on the basic physical-chemical interaction and engineering (or soil) properties of the palm oil mixed clays. The main focus of this study therefore is to address this issue and answer the above research questions. Selection of clay for this study was in relation to the natural soil deposit mostly encountered in the palm plantations and/or mills in Malaysia and Indonesia. The geological surveys and reports from the Southeast Asian regions though highlights a range of soil types, the abundance of kaolinite mineral in these soil types was a common observation (Liu *et al.* 2012). Thus it was felt appropriate to use a controlled kaolinite rich soil sample (referred to as kaolin hereafter) in contrast to using natural soil deposits–owing to its varying material characteristics. Therefore, the main aim of this paper is to evaluate the effect of increasing palm oil content on the basic physical-chemical and soil (or geotechnical) properties of kaolin using a series of laboratory experimentation. The discussions in this paper mainly focusses on comparing the measured response of increasing palm oil contents in kaolin with the observation from literatures using other types of oils and attempt to generalizing the effects of oil on clays. The paper concludes with details on the material characterization of palm oil mixed kaolin (POMK) samples and elaborating their basic soil properties in comparison with the existing knowledge.

2. Materials

Commercially available kaolin and refined palm oil was used in this study. The refined oil instead of field samples of either POME or palm oil was used to limit the interaction of any impurities–as the study aims to evaluate the effect of palm oil only. The POMK samples were prepared by hand mixing palm oil with the oven dried (at 105°C for 24 hours) clean kaolin sample (K_{00}). Oil was further supplemented at increments of 10 (K_{10}), 20 (K_{20}), 30 (K_{30}) and 40 (K_{40}) per cent measured by the weight of dry kaolin. The samples were then sieved through a 0.2 mm opening sieve to exclude any clogs and lumps of POMK samples. The samples were later stored in air-tight plastic bags for about a week to facilitate stabilization and/or surface charge equilibration. The oil content in the POMK samples were ensured by periodically testing the samples using the hexane solvent oil extraction method–see Urum *et al.*

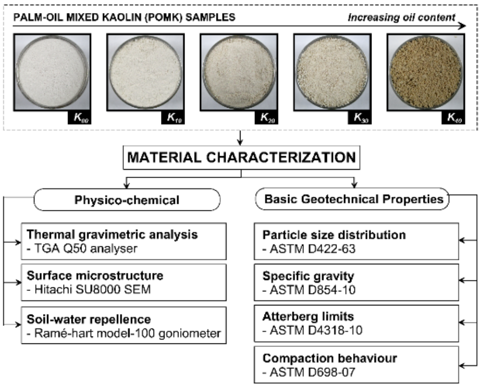

Fig. 1 Flow chart showing the experimental program used in this study

(2006) for detailed procedure. The addition of palm oil to the pale white colored kaolin particles showed visually evident physical changes, including the color and texture–formation of aggregates, clogs and lumps. The POMK's showed increasing reddish-orange color with increasing oil content, which typifies the overlapping or coating of carotenoids–red fat-soluble pigments present in the palm oil (Nagarajan *et al.* 2017, Goh *et al.* 1985). Fig. 1 shows the image of the POMK samples from this study. The figure also summarizes the flow and details of the experimental program adopted in this study.

3. Experimental methodology

The thermal gravimetric analysis (TGA) of the POMK samples was conducted using a TGA Q50 V20.10 Build 36 analyzer to investigate the behavior of the palm oil in the soil with temperature. The outcomes from TGA was anticipated to evaluate any temperature restrictions to be adapted in the laboratory tests on the POMK samples. Test procedure involved a constant temperature increase at 10ºC/min from an ambient temperature of 25ºC up to 800ºC aided by nitrogen purging at a rate of 20 and 40 l/min as balance and sample gasses respectively. Test samples included 2-5 g of fine portions of the air dried POMK sample, spread on a platinum pan. The TGA outcomes are presented as the variation of mass loss and their first derivative with temperature. The surface microstructure of the kaolin before and after supplementing palm oil was observed using Hitachi SU8000 high resolution field emitting scanning electron microscope (SEM) operated at 5-15kV and working pressure of 50-70 a. The POMK samples were initially coated using platinum to enable conductance and then placed on the aluminum holders of 12 m diameter using carbon tapes. The specific surface area (S_a) of POMKs was estimated using the BET method. Approximately 0.1-0.2 of POMK sample was placed in the glass sample tube of an accelerated surface area and porosimetry equipment. The S_a values thus estimated were averaged based on three trails, which showed less than 2% variation in the results.

Soil water repellency was studied using the water drop penetration time (WDPT) and static contact angle

Table 1 Basic properties of POMK samples

Description	Units	Procedure	K_{00}	K_{10}	K_{20}	K_{30}	K_{40}
BET surface area, S_a	m^2/g	Brunauer *et al.* (1938)	6.76	7.00	4.03	2.76	1.48
Water droplet penetration time, WDPT	s	Doerr *et al.* (2000)	1	684	1148	2824	3652
Contact angle, ø	º	Doerr *et al.* (2000)	< 90	106.7	109.4	112.3	128.0
Specific gravity, G_S		ASTM D854-10	2.44	2.18	1.91	1.82	1.71
Liquid limit, w_L	%	ASTM D4318-10	64	71	67	55	31
Plastic limit, w_P	%	ASTM D4318-10	41	36	28	22	14
Shrinkage limit, w_S	%	ASTM D4943-02	6	17	20	18	13
Plasticity index, I_P	%	ASTM D4318-10	24	34	39	37	17
Activity			1.7	2.0	3.2	5.1	2.6
Maximum dry unit weight, $\gamma_{d,max}$	kN/m^3	ASTM D698-07	13.3	12.8	11.9	11.7	11.2
Optimum moisture content, w_{opt}	%	ASTM D698-07	24.0	11.7	8.6	7.9	6.4

measurements. For this purpose, 50 g air dried POMK sample was levelled in a petri-dish. Using a hypodermic needle attached to a 20 ml syringe, 3 μl drops (spherical droplet about 1 mm radius) of distilled water was gently deposited onto the soil surface in an air environment. Static contact angle measurements were carried out using a Ramé-hart model goniometer. Images of the droplet were obtained by a digital camcorder with 10x optical and 120x digital zoom and analyzed for the contact angles using an image tool software. In the WDPT test, a 3 μl droplet of distilled water was placed on the horizontal surface of the samples, and the time taken for the water drop to infiltrate or move completely into the soil was recorded, as longer durations suggests higher water repellency. Repeatability and any error due to heterogeneity at micro levels was ensured with five test trials conducted at different positions on the sample's surface.

Table 1 tabulates the laboratory tests conducted to evaluate the basic geotechnical properties of the POMK's. The table also details the standard procedure followed in the laboratory tests and their outcomes. However, one major modifications from the standard practice (ASTM D2216-10) was in the water content measurements for the POMK samples. This modification primarily relates to the presence of oil forming a four phase (soil-oil-water-air) system now,which otherwise should have been a traditionally used three phase (soil-water-air) system. To this end, the relation suggested by Khamehchiyan *et al.* (2007) shown below, is used to estimate the water content values of all the samples in this study.

$$w = \left[(1 + mn)\frac{m_1}{m_2}\right] - (1 - m) \qquad (1)$$

where, m_1 is the wet mass of oil-mixed soil, m_2 is the dry mass of oil-mixed soil, and m and n are the oil content in the soil samples before and after drying. Main reason using

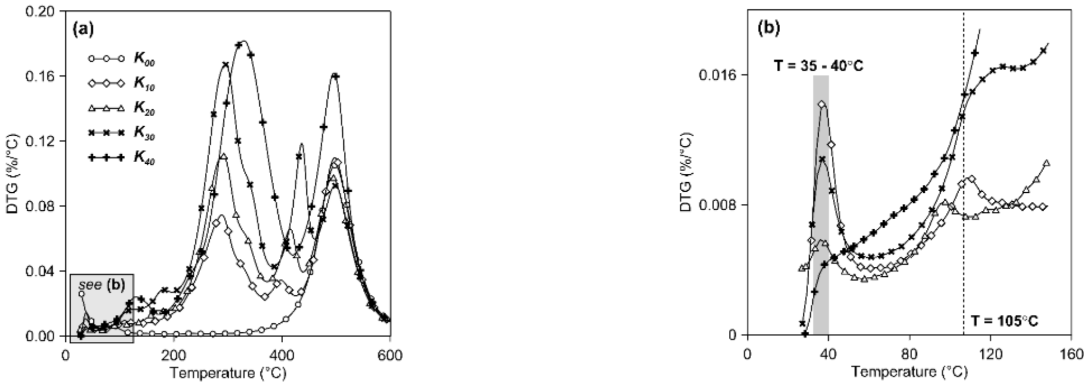

Fig. 2 Comparison of POMK samples response to increasing temperature

the relation proposed by Khamehchiyan *et al.* (2007) was the close comparison in the evaporation behaviors of the oil at higher temperatures in oven drying observed in this study. The trend of evaporation curve shows an inverse correlation of oil loss with increasing oil content, explaining the possibility of higher effort required to break the bonding between the oil and soil as stated by Khamehchiyan *et al.* (2007). The oil content was measured using the hexane solvent oil extraction method (Urum *et al.* 2006).

4. Results and discussion

Oil properties in general are sensitive to the temperature (T). Sensitivity for example ranges from varying physical properties–viscosity, evaporation etc. at moderate T of 40ºC-100ºC, to complete decomposition at higher temperatures (greater than 100ºC). Thermal analysis of POMK's in this study initially focused to map ΔT associated with the traditional oven drying method (ASTM D2216-10), that is at T ≤ 105ºC. Nonetheless, TGA was later considered appropriate as to contemplate oils' fate not only at T ≤ 105ºC, but also when exposed to higher T values, though impractical in normal operational conditions. Fig. 2 shows the results of TGA in this study. Fig. 2(a) adopts the derivative thermo-gravimetric weight (DTG) replacing the conventional weight loss in the analysis to capture even minor changes in the sample weight. Relatively lower weight loss with DTG of 0.004-0.014%/ºC was observed at 40ºC–100ºC, which basically relates to the structural reorganization in POMK samples–water molecules and fatty acids responding to increasing temperatures (de Souza *et al.* 2014). The next peak at around 293ºC is associated with the degradation of unsaturated fatty acids and low-molecular weight triglycerides present in palm oil, which is generally reported to degrade between 190ºC and 310ºC (Santos *et al.* 2014). Further peaks at 400ºC to 440ºC most likely marks completion in decomposition of triglyceride in POMK samples (Santos *et al.* 2014).

Fig. 2(b) magnifies the TGA curves limiting to T values at 105ºC, as to address its purpose in this study. It should be noted that the K_{00} sample is not considered for the analysis due to absence of oil in it. Observations highlight distinct peaks at 35ºC-40ºC which perhaps depicts the release of physically absorbed water molecules. Despite the boiling point for water close to 100ºC, vaporization is even expected at lower temperatures following the low activation energy – at 43.5kJ/mol required for the vaporization of water (Holt *et al.* 1962). However, in the POMK samples the pore fluid consists of both water and palm oil, hence vaporization of the palm oil also needs to be considered for T ≤ 105ºC. Tan *et al.* (2001) estimates the activation energy required to oxidize the lipid constituents in palm oil as 89.3±0.3kJ/mol, which is clearly 2-folds for that of water. Although this observation tends to rule-off the possibility of the vaporization of palm oil fraction in the POMK's, it should be noted that this inference in this study is limited to a constant heating rate of 10ºC/min as per the procedure followed in the TGA measurements. This means the test samples were exposed to a certain temperature only for only a minute before the T was increased by another 10ºC.

Nonetheless, duration of exposure at a given temperature also plays an important role. The oil in general, has shown to have evaporated with increasing evaporative exposure – function of mass transfer coefficient under prevailing wind conditions, gas constant, and environmental temperature, as demonstrated in Stiver and Mackay (1984). For examples, Stiver and Mackay (1984) justified possible evaporation of crude oil samples at temperatures as low as 0ºC. Similar evaporation behavior of the palm oil is thus anticipated in this study. In-spite of the degradation temperature of palm oil at around 293ºC, vaporization of palm oil from the POMK samples was observed, rather sensed, during oven drying at 105ºC for 24 hours (ASTM D2216-10) in this study. Observations presented here were based on the strong essence of palm oil felt (smell) in the laboratory when the POMK's were subjected to overnight drying in oven. These discuss strongly substantiates the possible evaporation or loss of oil during drying, and thereby our choice of using the method proposed by Khamehchiyan *et al.* (2007) to estimate the water content of POMK samples in this study.

Palm oil by its physical nature is a viscous fluid. Its interaction with clay particles is mostly dominated with adhesion at micro- and macro-structural levels, associated with low and high oil contents respectively. SEM micrographs were attempted to study and discuss possible interaction of palm oil with the kaolin particles at the micro-structural level. The noticeable hexagonal shape forming vermicular stacks varying sizes with low crystal width-to-thickness ratio and sharp-edged profiles are amongst the most general particulate characteristics of kaolinite samples

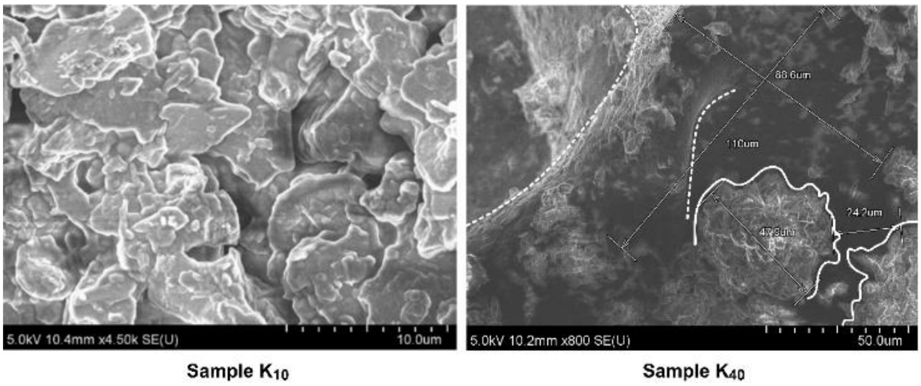

Fig. 3 SEM image of K_{10} and K_{40} samples. Image of the K_{40} sample highlights the smooth-fluidic surface and the clusters

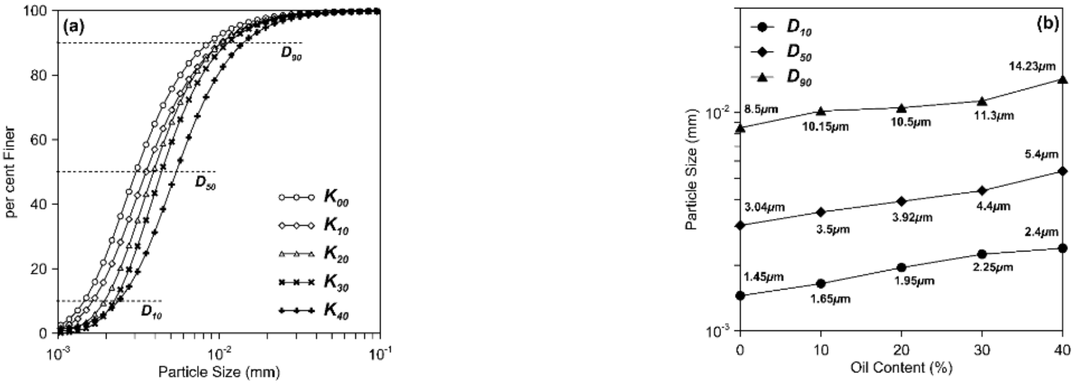

Fig. 4 Particle size distribution curves for POMK samples

(Murray 2000), which were also observed from the SEM micrographs for K_{00} samples. However, SEM analysis in this study was mainly attempted to obtain a visual detection of the oil's presence in the micro-structure of the POMK's. Fig. 3 shows the SEM images captured using K_{10} and K_{40} samples. The SEM images for the K_{40} sample is believed to highlight the presence of a layer of oil. In comparison to the typical SEM features of oil coated kaolin particles discussed in literature (Khosravi *et al.* 2013; Liu *et al.* 2015; Murray 2000), observations in this study also highlights the formation of clay clusters. This clearly relates to the physical adhesion of palm oil traces attracting clay particles to form clusters, as discussed in the literature. The SEM images and observations from this study also showed several such irregularly arranged clusters with diameters ranging between 0.05 mm and 0.075 mm. The SEM image for the K_{40} in Fig. 3 highlights one such cluster (diameter $\approx$ 0.05mm) embarked within the solid white line. However a unique, yet distinct feature from the figure is the adulated smooth fluidic surface with clay particles adhered to it (highlighted by broken line), which can be visualized as the layer of palm oil. Multiple efforts to confirm this, and ensure repeatability, was not successful in this study due to oil traces which very observed to be unstable when exposed to the working conditions of the SEM equipment. Nonetheless the cluster formation or aggregation of the clay particles with palm oil was observed in most of the SEM trials signifying the role of oil in adhering the particle-to-particle and coating the clay particles.

In addition to the surface morphologies, corresponding elemental weight compositions of each POMK sample were determined using the Energy Dispersive X-Ray (EDX) spectroscopy. Clean kaolin followed the typical form of kaolinite with aluminosilicate together with higher concentrations of Al and Si peaks. The oxides of these elements basically accounted to over 90 per cent of the total mass of the sample analyzed perhaps. The EDX spectra for samples with increasing palm oil content showed low signal of noise ratio – based on the observed disturbed spectra. Since oil coating impedes elemental quantification of the samples, the quantification of the elements in this study was believed to be vastly affected in samples with higher oil content. It was in this way the layer observed in Fig. 3 was determined as a layer of oil. Nonetheless due to limited repeatability of tests being achieved–related to the instability of oil under the working conditions of SEM apparatus, further investigations are advised to establish a more precise determination of oil's presence in the micro-structure of clays.

Kaolin particles generally are hydrophilic and oleophilic (Saada *et al.* 1995), which was also observed in the laboratory when dry kaolin samples were mixed with water and oil separately in this study. However, palm oil traces coating the clay particles are likely to affect the wettability of the particles and the flow due to the water repellency offered by the oil. Thus, presence of water in kaolin induces oleophobic surface behavior perhaps preventing oil from attaching onto the surface of kaolin particles. It is for this reason palm oil was mixed with oven dried kaolin to prepare POMK samples in this study–as to study the effect of oil on kaolin. Table 1 shows the solid-liquid contact angle (ø) and the water drop penetration time (WDPT) measurements for the POMK samples. The measured ø values for samples mixed the oil content (as less as 10%)

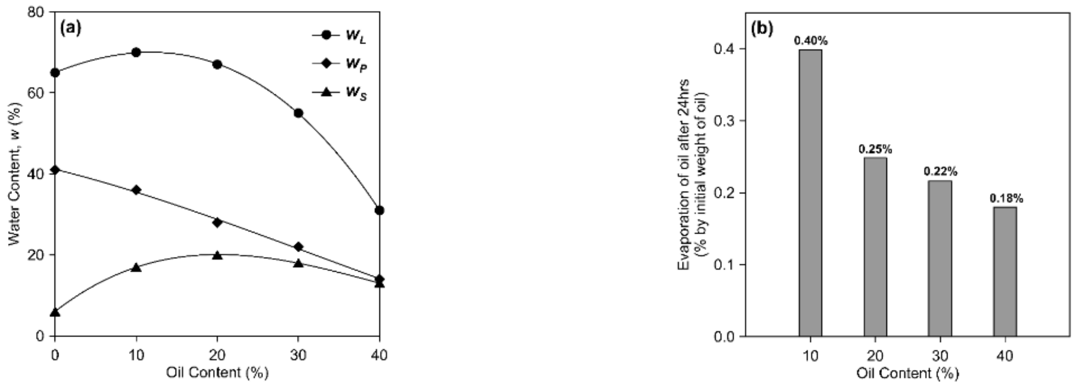

Fig. 5 Effect of palm oil content on the (a) consistency limits of POMK samples and (b) evaporation of oil

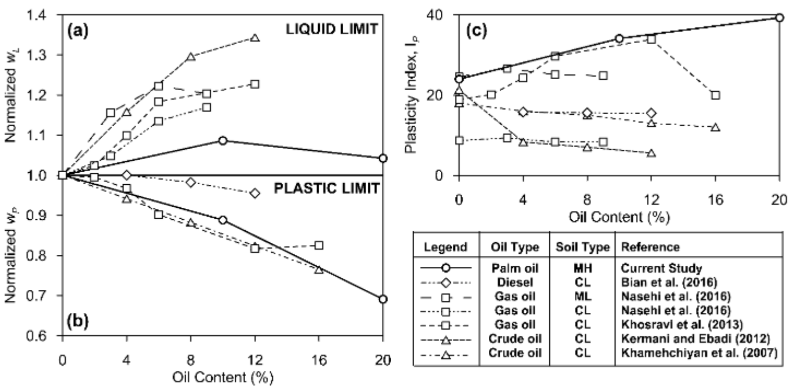

Fig. 6 Comparison of type and amount of oil on the consistency limits of fine graded soils

was ≥ 90º, which clearly describes the non-wettable surface of the samples. The wettability, generally expressed using water repellency, is well explained based on the varying surface tension at the water-air interface, oil-air interface and oil-water interface, which were 72.7 mN/m, 32.7 mN/m, and 31.3 mN/m respectively for palm oil in this study. It is unlikely in many trials to obtain a planar soil surface that readily allow ø measurements to define wettability. It is for this reason that the WDPT measurements were adopted henceforth to explain water repellency. The average time for the WDPT increased linearly with increasing palm oil content with coefficient of determination (r^2) of 0.94. According to the classification system by Dekker and Ritsema (1994), the control sample K_{00} was classified as wettable with very low WDPT time (< 1s). Based on these evidences, the POMK samples (K_{10} - K_{40}) were classified as extremely hydrophobic with WDPT ≥ 600s, thereby proving possibility of intrinsic hydrophobicity in POMKs with increasing oil content.

Fig. 4(a) shows the particle size distribution (PSD) curve for POMK samples – *only the best fit lines drawn to the experimental data*. In comparison to clean kaolin (K_{00}), the PSD for POMK's show a distinct reduction in per cent fines with increasing palm oil content. This reduction certainly relates to the formation of pseudo-silt sized particles due to the adhering palm oil and adjacent particles or cluster formation as discussed above. Virtually it can be hypothesized that the palm oil sleeks to wrap around the kaolin forming larger particles due to surface forces and adhering adjacent particles henceforth. Similar observation and explanation was presented by Ijimdiya (2012) and Trzciński *et al.* (2015) discussing on motor oil contaminated soil. In addition, Williams *et al.* (2004) discussed the contribution of the upper end of the PSD curve to aggregation or flocculation. Hence to track the possible increase in the particle sizes due to any of the above-mentioned conditions, the particle diameter of K_{00} and the POMK samples are separately plotted using particle sizes (diameter) at 10% (D_{10}), 50% (D_{50}), and 90% (D_{90}) finer from Fig. 4(a). Fig. 4(b) shows the variation of these particle sizes with increasing oil content. Observations from the estimated values show an increase in particle sizes ranging up to 65-75% of the K_{00} samples. The PSD data however is limited to particle shape being considered as spherical unlike as expected in kaolin particles – generally hexagonally shaped and flaky with high width-to-thickness ratio and other assumptions in the procedure (ASTM D422-63). However, the decreasing S_a values (area per unit gram of the POMK samples) with increasing palm oil content (see Table 1) is an added information to justify this formation of pseudo-silt sized particles or clusters.

The formation of cluster, varying pore fluid (emphasizing on the presence of oil in pore unlike the ideal condition of water alone) and other possible interaction of palm oil with clay particles is certainly expected to also influence the consistency of the remolded POMK samples. For this purpose, the basic limits suggested by Atterberg - liquid limit (w_L), plastic limit (w_P), and shrinkage limits (w_S), are used in this study. Fig. 5(a) quantifies the influence of palm oil on the w_L, w_P, and w_S values of the POMK samples. Observations in general shows an initial increase in the w_L and w_S values up to around 10% and 20% oil contents respectively, followed by decrease with further increasing oil content. While w_P linearly decreases with increasing oil content. The minor increase observed in w_L - ranging to about 8.5% of the w_L for K_{00} may relate to the adhesive force from the palm oil bonding the clay particles. Hence a slightly higher amount of water being required for the POMK samples to flow on its own weight when tested using Casagrande's apparatus/method (ASTM D4318-10).

While with increasing oil content the bonding tends to get weaker either due to a thick film of oil coating the particles or clay particles weakly adhering to thick, smooth and fluidic surface of palm oil as demonstrated in the SEM images in this paper (see Fig. 3). The presence and increasing oil and its lubricating effect perhaps makes the soil softer and thereby a relatively lower pore water to reach its liquid limit and the decreasing w_P values. The variation of w_S is basically independent of the soil's plasticity characteristics (Sridharan and Prakash 1998). At lower oil contents (< 20% by weight of kaolin) the cluster formation and flocculation effect is believed to allow larger pore space, thereby increasing the w_S (from about 6% to 20%). On the other hand, palm oil being relatively a viscous (see Table 1) fluid occupies additional space within the void thereby limiting the pore space for water in the soil skeleton at higher oil contents.

Owing to the evaporation of both pore water and palm-oil in the conventional oven drying method as per the discussions presented above with TGA data in this paper, the effect of evaporating oil was captured during the calculation of water content (using Eq. (1)). In addition to this, the values of w_L, w_P, and w_S were also calculated based on the per cent evaporation of palm oil measured in the laboratory at 105ºC for 24 hours. Fig. 5(b) shows the evaporation of oil expressed as per cent by initial weight of oil in POMK samples used in the calculations. The w was then calculated by subtracting the total weight of water lost with the estimated weight of oil evaporated in the process. However, no significant differences in the values of w_L, w_P, and w_S was observed. Nonetheless the w_P, and w_S values for K_{40} was observed to coincide with each other (see Fig. 5(a)). This perhaps marks as the limiting value of palm oil in POMK samples that can be tested using the standard laboratory geotechnical testing procedure to determined Atterberg limits as per ASTM D4318-10.

Fig. 6(a)-6(c) compares the w_L and w_P values from this study with the values from the literature. The comparison was extended over 4 different types of oils including palm oil from this study, diesel, gas and crude oils from the literature. The literature selected were limited to fine grained soils that were classified predominantly under the clay of low plasticity (CL), silt of high plasticity (MH), and silt (ML) categories based on the Unified Soil Classification System (USCS). The w_L and w_P values were further normalized with the w_L and w_P values of the clean soil samples (zero oil content) respectively for the purpose of comparison. The observation from Fig. 6(a) and 6(b) shows a good comparison in the trends of both w_L and w_P curves respectively. Irrespective of the type of oil and/or the soil used, the curves show an initial increase followed by a decrease in the values of w_L which perhaps suggests the presence of oil to significantly influence the consistency of the soil sample. Same conclusion can also be drawn based on the variation of w_P curves. To further strengthen the discussion, the plasticity index (I_P) values for the data from current study and the literature are calculated. Fig. 6(c) presents the variation of I_P with oil content. Although observations show random distribution of curves, for discussion, the variation of the soil samples with palm, gas oil and diesel can be visualized to be similar, that is an initial increase followed by dropping I_P values with increasing oil contents, while the results with crude oil showed continued decrease in I_P values with increasing oil content. Hence based on the comparison and the discussion, it is suggested that a further elaborate investigation considering different types of oil, their physical and chemical properties is essential.

Reducing I_P due to increasing oil content is likely to promote better compaction because oil potentially alters the soil's structural arrangement (Meegoda *et al.* 1998). Standard Proctor compaction tests were therefore conducted on POMK samples to determine the influence of increasing palm oil content on the molding dry unit weight (γ_d) and optimum moisture content (w_{opt}) of kaolin. Fig. 7 presents the compaction curves for POMK samples. The compaction curve developed for clean kaolin sample (K_{00}) were determined from three independent trials, highlighting a maximum unit weight ($\gamma_{d,max}$) of 13.2 kN/m^3 at w_{opt} of 24.7%. The figure also shows tangential lines based on the G_S of kaolin (2.44) cutting across the compaction curves which represents the different degree of saturation (S) of the specimen. A low S observed for the K_{00} samples prepared at dry of optimum is a result of relatively large pore spaces filled with air, due to the irregular soil-particle arrangement (Yoshinaka and Kazama 1973). Increasing the S through addition of water in K_{00} softens the particles by means of absorption and forms a thin double-layer water film (Lawton et al. 1992; Sridharan and Jayadeva 1982). Water-film lowers the resistance to deformation and allows particles to slide over one another during compaction (Delage *et al.* 1996) resulting in a reduction of the void ratio (e) with increasing w up to w_{opt}. Further addition of w collapses the structure as the water film comes in contact with adjacent water film layers (Sridharan and Jayadeva, 1982). Collapse of kaolin structure in samples prepared at wet of optimum form flat edge-to-edge structure in a two-dimensional arrangement (Yoshinaka and Kazama 1973). Therefore, samples compacted at wet-of optimum observed an increase in e and S. Above all, it is important to comprehend the distribution of voids under wet-of optimum condition is significantly different to dry-of optimum in relation to arrangement of particles.

Increasing the palm oil content from 0 to 40% decreased the $\gamma_{d,max}$ of POMK samples by 16% to about 11.2kN/m^3. The w_{opt} to attain $\gamma_{d,max}$ for POMK samples reduced by a greater margin with increasing palm oil content. For example, the reduction in wopt for an increase in 10% palm oil content was observed close to 51%. Subsequent increase in palm oil content up to K_{40} sample revealed 73% reduction in the required w_{opt} to attain $\gamma_{d,max}$. The reduction in the compaction characteristics can be associated to the degree of flocculation of the clay aggregates (Lambe 1958) due to increasing palm oil. This is also demonstrated from the overall reduction observed in γ_d for POMK samples in comparison with K_{00} across the molding w. Furthermore, increasing oil content significantly reduce the pore spaces

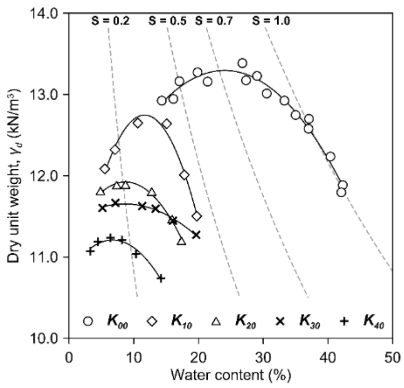

Fig. 7 Effect of palm oil content on the compaction characteristics of POMK samples

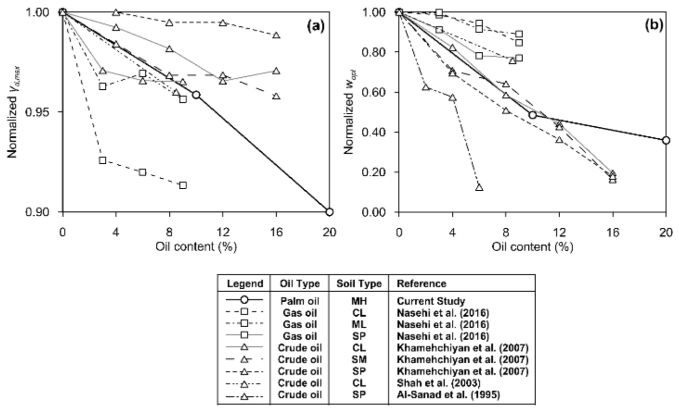

Fig. 8 Comparison of type and amount of oil on the consistency limits of fine graded soils

within the soil sample. This further reduced the available spaces for water in the soil skeleton, thereby reduced the wopt for $\gamma_{d,max}$ of samples with increasing palm oil content. It can also be argued that the reduction in OMC is a consequence of reduced capillary forces due to oil-water repellency (Abousnina *et al.* 2015). The results also suggest the higher viscosity of palm oil (77.2mPa.s) in comparison to water (1.0mPa.s) may provide better lubrication to facilitate clay particles sliding over each other. Hence, a denser packing arrangement of kaolin perhaps require lower molding *w* with increasing palm oil content.

Fig. 8(a) and 8(b) compare the $\gamma_{d,max}$ and w_{opt} values from this study with that presented in the literature. The comparison was extended over three different types of oils including palm oil – limited to 20% oil content from this study, and gas and crude oils from the literature. The literature selected were extended across different soil types that were classified with in CL, MH, ML, silty sand (SM) and poorly graded sand (SP) categories from the literature. The $\gamma_{d,max}$ and w_{opt} values were normalized with the respective $\gamma_{d,max}$ and w_{opt} values of clean soil samples for comparison. Observation from the figures show good comparison in the trends for both $\gamma_{d,max}$ and w_{opt}. Literature focusing on crude oil unlike other oil types show higher $\gamma_{d,max}$ and w_{opt} values of cleans soils in comparison to oil mixed soils. Though increasing oil content reduced the compaction behavior of fine grained soils, a consistent variation however was noted with other oil types which is believed to relate with the higher viscosity values of the crude oil. The reduction in normalized $\gamma_{d,max}$ of soil with increasing gas oil (up to 20%) is twice that for crude oil. On the contrary, the order of oil type is reversed for the w_{opt} for the soils respectively. This certainly relates to the differences in viscosity for the oil type in consideration – gas oil (1.7-2.0 mPa.s), palm oil (77.2 mPa.s) and crude oil (20.5-424.0 mPa.s). Lower viscosity of oil tends to show higher rate of decrease in $\gamma_{d,max}$, while comparatively lower rate of decrease in the w_{opt} for fine grained soils used in this study for comparison. Based on the comparison, it is apparent that the viscosity of oil perhaps has a major role dictating the compaction characteristics of oil supplemented fine grained soils.

5. Conclusions

Following the experimental results and discussions aided by comparison with literature, it can be concluded that the supplementation of oil in soil, generally referred to as oil contamination, pose a significant influence on the basic properties of fine grained soils. The physical adhesion of palm oil traces on the particles as estimated in this study based on the SEM images, which eventually brings the clay particles together leading to the formation of pseudo-silt sized clusters. These pseudo-silt sized clusters, in collaboration with the water repellency nature of oil coating the clay particles, pose notable variation in the consistency of the kaolin samples as measured by the variations observed in Atterberg limits. The consistency limits from this study further showed good comparison as observed in the trends with oil contamination of fine grained soil from the literature. This evince that the oil coating the soil particles is most likely to dominate the influence. Results from the compaction tests signifies the contribution of oil's viscosity to influence the compaction behavior of the soil, owing to decreasing maximum dry unit weight ($\gamma_{d,max}$) and optimum moisture content (w_{opt}) values observed in the experimental results for palm oil and kaolin. Lower viscosity of oil tends to show higher rate of decrease in $\gamma_{d,max}$ unlike lower decrease rates in the wopt values for fine grained soils used in this study for comparison. To this end, the current experimentation was limited to the condition where oil interacts with the dry soil particles first (oil coating soil particles), while it is convincingly true that the behavior may change when oil interacts with the wet soil – owing to the oleophilic nature of the kaolin particles, which certainly requires further consideration and in-depth study.

Acknowledgements

This study was supported by the Advanced Engineering Platform, Monash University Malaysia under Multi-disciplinary Research Competitive Grant (AEP-15-004). First author acknowledges Monash University Malaysia for the financial support under the Higher Degree by Research Scholarship program for his doctoral program.

References

Abousnina, R.M., Manalo, A., Shiau, J. and Lokuge, W. (2015), "Effects of light crude oil contamination on the physical and mechanical properties of fine sand", *Soil Sediment Contamin. Int. J.*, **24**(8), 833-845. https://doi.org/10.1080/15320383.2015.1058338.

Al-Sanad, H.A., Eid, W.K. and Ismael, N.F. (1995), "Geotechnical properties of oil-contaminated Kuwaiti sand", *J. Geotech. Eng.*, **121**(5), 407-412. https://doi.org/10.1061/(ASCE)0733-9410(1995)121:5(407).

Bian, H., Liu, S., Cai, G. and Chu, Y. (2016), "Influence of diesel pollution on the physical properties of soils", *Jap. Geotech. Soc. Special Publ.*, **2**(14), 552-555. https://doi.org/10.3208/jgssp.CHN-16.

Brunauer, S., Emmett, P.H. and Teller, E. (1938), "Adsorption of gases in multimolecular layers", *J. Amer. Chem. Soc.*, **60**(2), 309-319. https://doi.org/10.1021/ja01269a023.

Chang, I. and Cho, G. (2014), "Geotechnical behavior of a beta-1, 3/1, 6-glucan biopolymer-treated residual soil", *Geomech. Eng.*, **7**(6), 633-647. https://doi.org/10.12989/gae.2014.7.6.633.

de Souza, C.M.P., dos Santos, D.P.G., Soares, B.S.A., de Santana, C.P., Apolinário, A.C., da Cunha, E.V.L., Damasceno, B.P.G., da Veiga Jr, V.F., Santana, D.P. and da Silva, J.A. (2014), "Physicochemical characterization and in vitro evaluation of the photoprotective activity of the oil from Opuntia ficus-indica (L.) Mill. Seeds", *Afr. J. Pharm. Pharmacol.*, **8**(33), 824-831. https://doi.org/10.5897/AJPP2014.4012.

Delage, P., Audiguier, M., Cui, Y. and Howat, M.D. (1996), "Microstructure of a compacted silt", *Can. Geotech. J.*, **33**(1), 150-158. https://doi.org/10.1139/t96-030.

Doerr, S.H., Shakesby, R.A. and Walsh, R.P.D. (2000), "Soil water repellency: its causes characteristics & hydro-geomorphological significance", *Earth-Sci. Rev.*, **51**(1-4), 33-65. https://doi.org/10.1016/S0012-8252(00)00011-8.

FAO (2016). *World Food Programme – 2015, The State of Food Insecurity in the World 2015. Meeting the 2015 International Hunger Targets: Taking Stock of Uneven Progress*, Food and Agriculture Organization Publications, Rome, Italy. http://www.fao.org/3/a-i4646e.pdf.

Goh, S.H., Choo, Y.M. and Ong, S.H. (1985), "Minor constituents of palm oil", *J. Amer. Oil Chem. Soc.*, **62**(2), 237-240. https://doi.org/10.1007/BF02541384.

Holt, J.B., Cutler, I.B. and Wadsworth, M.E. (1962), "Rate of thermal dehydration of kaolinite in vacuum", *J. Amer. Ceramic Soc.*, **45**(3), 133-136. https://doi.org/10.1111/j.1151-2916.1962.tb11101.x.

Ijimdiya, T.S. (2012), "Effect of oil contamination on particle size distribution and plasticity characteristics of lateritic soil", *Adv. Mater. Res.*, **367**, 19-25. https://doi.org/10.4028/www.scientific.net/AMR.367.19.

Izdebska-Mucha, D., Trzcińsk, J., Żbik, M.S. and Frost, R.L. (2011), "Influence of hydrocarbon contamination on clay soil microstructure", *Clay Miner.*, **46**(1), 47-58. https://doi.org/10.1180/claymin.2011.046.1.47.

Kermani, M. and Ebadi, T. (2012), "The effect of oil contamination on the geotechnical properties of fine-grained soils", *Soil Sediment Contamin. Int. J.*, **21**(5), 655-671. https://doi.org/10.1080/15320383.2012.672486.

Khamehchiyan, M., Charkhabi, A.H. and Tajik, M. (2007), "Effects of crude oil contamination on geotechnical properties of clayey and sandy soils", *Eng. Geol.*, **89**(3-4), 220-229. https://doi.org/10.1016/j.enggeo.2006.10.009.

Khosravi, E., Ghasemzadeh, H., Sabour, M.R. and Yazdani, H. (2013), "Geotechnical properties of gas oil-contaminated kaolinite", *Eng. Geol.*, **166**, 11-16. https://doi.org/10.1016/j.enggeo.2013.08.004

Lai, O.M., Tan, C.P. and Akoh, C.C. (2015), *Palm Oil Production, Processing, Characterization, and Uses*, Academic and AOCS Press, https://doi.org/10.1016/C2015-0-02411-X.

Lambe, T.W. (1958), "The engineering behaviour of compacted clay", *J. Soil Mech. Found. Div.*, **84**(2), 1-35.

Lawton, E.C., Fragaszy, R.J. and Hetherington, M.D. (1992), "Review of wetting-induced collapse in compacted soil", *J. Geotech. Eng.*, **118**(9), 1376-1394. https://doi.org/10.1061/(ASCE)0733-9410(1992)118:9(1376).

Lim, M.W., Lau, E.V. and Poh, P.E. (2016), "A comprehensive guide of remediation technologies for oil contaminated soil– Present works and future directions", *Mar. Pollut. Bull.*, **109**(1), 14-45. https://doi.org/10.1016/j.marpolbul.2016.04.023.

Liu, Z., Liu, S., Cai, Y. and Fang, W. (2015), "Electrical resistivity characteristics of diesel oil-contaminated kaolin clay and a resistivity-based detection method", *Environ. Sci. Pollut. Res.*, **22**(11), 8216-8223. https://doi.org/10.1007/s11356-014-3964-7.

Liu, Z., Wang, H., Hantoro, W.S., Sathiamurthy, E., Colin, C., Zhao, Y. and Li, J. (2012), "Climatic and tectonic controls on chemical weathering in tropical Southeast Asia (Malay Peninsula, Borneo, and Sumatra)", *Chem. Geol.*, **291**, 1-12. https://doi.org/10.1016/j.chemgeo.2011.11.015.

MacNeill, M. (2016). *Annual Report, Vienna, Austria: Organization of the Petroleum Exporting Countries*.

Meegoda, J.N., Chen, B., Gunasekera, S.D. and Pederson, P. (1998), *Compaction Characteristics of Contaminated Soils: Reuse as a Road Base Material* in *Recycled Materials in Geotechnical Applications, ASCE Geotechnical Special Publication No. 79*, 195-209.

Mitchell, J.K. and Soga, K. (2005). *Fundamentals of Soil Behavior*, John & Wiley Sons.

Mohammadi-Sichani, M., Assadi, M.M., Farazmand, A., Kianirad, M., Ahadi, A. and Hadian-Ghahderijani, H. (2018), "Ability of Agaricus bisporus, Pleurotus ostreatus and Ganoderma lucidum compost in biodegradation of petroleum hydrocarbon-contaminated soil", *Int. J. Environ. Sci. Technol.*, **16**(5), 1-8. https://doi.org/10.1007/s13762-017-1636-0.

Mohanty, S.K., Pradhan, P.K. and Mohanty, C.R. (2017), "Stabilization of expansive soil using industrial wastes", *Geomech. Eng.*, **12**(1), 111-125. https://doi.org/10.12989/gae.2017.12.1.111

Mowat, L. (2018), British coast LEGALLY used as dumping ground of palm oil by foreign tankers: Express, https://www.express.co.uk/news/world/914723/palm-oil-ships-poison-waters-British-waters-Cornwall-European-Maritime-Safety-Agency.

MPOB (2017). *Overview of the Malaysian Oil Palm Industry, Kuala Lumpur*: Malaysian Palm Oil Board.

Murray, H.H. (2000), "Traditional and new applications for kaolin, smectite, and palygorskite: A general overview", *Applied Clay Sci.*, **17**(5), 207-221.

Nagarajan, J., Ramanan, R.N., Raghunandan, M.E., Galanakis, C.M. and Krishnamurthy, N.P. (2017), *Chapter 8 – Carotenoids. Nutraceutical and Functional Food Components*, Academic Press, 259-296, https://doi.org/10.1016/B978-0-12-805257-0.00008-9.

Nasehi, S.A., Uromeihy, A., Nikudel, M.R. and Morsali, A. (2016), "Influence of gas oil contamination on geotechnical properties of fine and coarse-grained soils", *Geotech. Geol. Eng.*, **34**(1), 333-345. https://doi.org/10.1007/s10706-015-9948-7.

Nyer, E.K. (2000). *In situ Treatment Technology*, CRC Press.

Olafisoye, O.B., Oguntibeju, O.O., and Osibote, O.A. (2017), "Trace elements and radionuclides in palm oil, soil, water, and leaves from oil palm plantations: A review", *Crit. Rev. Food Sci. Nutr.*, **57**(7), 1295-1315. https://doi.org/10.1080/10408398.2014.886032.

Pandey, A. and Bind, Y.K. (2014), "Effects of oil contamination on geotechnical properties of alluvial soil Naini, Allahabad", *Int. J. Innov. Technol. Explor. Eng.*, **3**(8), 39-42.

Rahman, Z., Umar, H. and Ahmad, N. (2010), "Geotechnical characteristics of oil-contaminated granitic & metasedimentary soils", *Asian J. Appl. Sci.*, **3**(4), 237-249. http://dx.doi.org/10.3923/ajaps.2010.237.249.

Rimmer, A., Parlange, J.Y., Steenhuis, T.S., Darnault, C. and Condit, W. (1996), "Wetting and nonwetting fluid displacements in porous media", *Transport Porous Media*, **25**(2), 205-215. https://doi.org/10.1007/BF00135856.

Saada, A., Siffert, B. and Papirer, E. (1995), "Comparison of the hydrophilicity/hydrophobicity of illites and kaolinites", *J. Colloid Interfac. Sci.*, **174**(1), 185-190. https://doi.org/10.1006/jcis.1995.1381

Saberian, M. and Khabiri, M.M. (2018), "Effect of oil pollution on function of sandy soils in protected deserts and investigation of their improvement guidelines (case study: Kalmand area, Iran)", *Environ. Geochem. Health*, **40**(1), 243-254. https://doi.org/10.1007/s10653-016-9897-y.

Santos, A.G.D., Souza, L.D., Caldeira, V.P.S., Farias, M.F., Fernandes Jr, V.J. and Araujo, A.S (2014), "Kinetic study and thermoxidative degradation of palm oil and biodiesel", *Thermochimica Acta*, **592**, 18-22. https://doi.org/10.1016/j.tca.2014.08.006

Shah, S.J., Shroff, A.V., Patel, J.V., Tiwari, K.C. and Ramakrishnan, D. (2003), "Stabilization of fuel oil contaminated soil-A case study", *Geotech. Geol. Eng.*, **21**(4), 415-427. https://doi.org/10.1023/B:GEGE.0000006052.61830.1a.

Soleimaninanadegani, M. and Manshad, S. (2014), "Enhancement of biodegradation of palm oil mill effluents by local isolated microorganisms", *Int. Scholar. Res. Notices*, 1-8. http://dx.doi.org/10.1155/2014/727049

Sridharan, A. and Jayadeva, M.S. (1982), "Double layer theory and compressibility of clays", *Geotechnique*, **32**(2), 133-144. https://doi.org/10.1680/geot.1982.32.2.133.

Stiver, W. and Mackay, D. (1984), "Evaporation rate of spills of hydrocarbons and petroleum mixtures", *Environ. Sci. Technol.*, **18**(11), 834-840. https://doi.org/10.1021/es00129a006.

Takawira, A., Gwenzi, W. and Nyamugafata, P. (2014), "Does hydrocarbon contamination induce water repellency and changes in hydraulic properties in inherently wettable tropical sandy soils?", *Geoderma*, **235-236**, 279-289. https://doi.org/10.1016/j.geoderma.2014.07.023.

Tan, C.P., Che Man, Y.B., Selamat, J. and Yusoff, M.S.A. (2001), "Application of arrhenius kinetics to evaluate oxidative stability in vegetable oils by isothermal differential scanning calorimetry", *J. Amer. Oil Chem. Soc.*, **78**(11), 1133-1138. https://doi.org/10.1007/s11746-001-0401-1.

Trzciński, J., Williams, D.J. and Żbik, M.S (2015), "Can hydrocarbon contamination influence clay soil grain size composition?", *Appl. Clay Sci.*, **109-110**, 49-54. https://doi.org/10.1016/j.clay.2015.03.014.

Urum, K., Grigson, S., Pekdemir, T. and McMenamy, S. (2006), "A comparison of the efficiency of different surfactants for removal of crude oil from contaminated soils", *Chemosphere*, **62**(9), 1403-1410. https://doi.org/10.1016/j.chemosphere.2005.05.016

Williams, N.D., Walling, D.E. and Leeks, G.J.L. (2004), "The settling behaviour of fine sediment particles: Some preliminary results from LISST instruments", *Proceedings of the International Symposium on Sediment Transfer through the Fluvial System*, Moscow, Russia, Russia.

Yoshinaka, R. and Kazama, H. (1973), "Microstructure of compacted kaolin clay", *Soil. Found.*, **13**(2), 19-34. https://doi.org/10.1016/0012-8252(72)90112-2.

Yu, E. (2017), *Hong Kong cleans up greasy beaches after palm oil spill: Phys.org*. https://phys.org/news/2017-08-hong-kong-greasy-beaches-palm.html.

Yu, H., Huang, X., Ning, J., Li, Z. and Zhao, Y. (2016), "Improving performance of soil stabilizer by scientific combining of industrial wastes", *Geomech. Eng.*, **10**(2), 247-256. https://doi.org/10.12989/gae.2016.10.2.247.

Żbik, M. and Horn, R.G. (2003), "Hydrophobic attraction may contribute to aqueous flocculation of clays", *Colloid. Surface. A: Physicochem. Eng. Aspects*, **222**(1-3), 323-328. https://doi.org/10.1016/S0927-7757(03)00250-4.

CC