Interaction studies between high-density oil and sand particles in oil flotation technology

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ABSTRACT

In the event of a sand contamination, the first course of action would be to ensure that a successful flotation is through the detachment of oil from sand for the ease of flotation. It is widely recognized that the initial oil–sand contact is crucial for oil removal and recovery. Due to its high viscosity and adhesive nature, high density bunker oil could pick up any silica particles (sand) of any size at a short contact time as low as several milliseconds. Nevertheless, the resulting detachment of sand particles from oil would vary under different conditions. Therefore, this study aims at investigating the interactions between oil and sand to further understand the detachment process between oil and sand in a flotation process under various conditions including pH, temperature, sand particle size and wettability. An increase in the water content in the sand sample from 0 wt% to 12 wt% aids the liberation of oil from contaminated sand from 0.7% to 65%, due to the presence of thin film of water which weakens the attachment forces between the oil and sand particles. On the other hand, the coarse sand particles of 1.0 mm easily detach themselves from the oil layer compared to finer sand particles of 0.125 mm which implicate that the attachment forces between oil and sand particles increase with the decrease in sand particle size. An increase in the solution pH from pH 6 to pH 14 and temperature from 20 °C to 60 °C also showed an increase in the sand detachment efficiencies from 25.1% to 60.9%, and from 15.2% to 85.1% respectively for 1 mm sand particle size. Further verification experiments including the differential zeta potential results and the DLVO theory supported the results of these former detachment studies, whereby differential zeta potential results showed that increase in pH increased the repulsive forces between particles, while the increase in temperature did not significantly affect the interparticle forces. Hence, the enhanced detachment efficiency due to increase in temperature is mainly attributed to the decrease in oil viscosity which reduces the adhesiveness of bunker oil which also facilitates oil liberation. Finally, the results are in good agreement with the oil flotation efficiencies.

1. Introduction

Oil spills on land are a common problem in the petroleum industry, as there are many ways where oil can spill and contaminate the land. In a petroleum refinery, accidental oil spillages may occur at drill rigs, storage tanks, tankers, pipelines and well heads. On the other hand, oil spillage on beach shorelines may also occur during transportation of oil on seas, where tidal wave action could sweep the oil on shore and contaminate the beach shoreline. In the event of an oil contamination on land, the oil tends to adhere to the sand/soil, making it difficult for immediate cleaning which could cause devastating impacts in many sectors (Wang et al., 2009). Absorption, inhalation and ingestion of oil could cause irreversible damage to the environment, economy and society. Therefore, this could lead to a long-term contamination if not removed in time.

The devastating impact of oil spills in the petroleum industries highlights the necessity and urgent need for removal of oil contamination. However, the removal of oil from contaminated shoreline, soils and sands is still currently an ongoing problem in the petroleum industry. Therefore, different methods had been developed to remove oil contaminant from soil, which have been implemented throughout the oil contamination sites in the past few years. The methods include physical removal via the high pressure hot water and combustion method (Bellier and Massart, 1979; Broman et al., 1983; Smith et al., 2001; Rushton et al., 2007), chemical removal using surfactants (Goi et al., 2006; Goi et al.,

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2009; Ahmadi and Shadizadeh, 2013), the biological method via bioremediation, and natural attenuation (Hoff, 1993; Merkl et al., 2005; Peng et al., 2009; Zheng et al., 2012; Rufino et al., 2013). Although successful rates of oil removal were shown through these studies, these methods, however, also exhibit their own disadvantages in terms of cost and feasibility. Therefore, there is a need to find other environment-friendly, cost-effective technologies to separate oil-contaminated soil which are simple to operate yet effective in a short period of time.

At present, one of the commonly studied oil removal technology is the flotation technology. Flotation technology is a combination of chemical, physiochemical and physical method which uses gas bubbles to remove contaminants. The advantages of flotation technology is that it could separate very small or light weight particles with low settling velocities, with minimal interference with the surrounding environment. In addition, it is also easily applied and implemented, with minimal operational cost. The success of the flotation process lies in three different mechanisms: liberation of oil from sand; aeration and attachment of air bubbles onto the bubbles; and flotation of bubble-oil particle (Tao, 2005).

Therefore, the first course of action to ensure a successful removal of oil from sand is through the detachment of oil from sand for the ease of flotation. It is widely recognized that the initial oil–sand contact is crucial for oil removal and recovery. Bunker oil is a naturally adhesive liquid due to its high viscosity. Due to its adhesive nature, oil could pick up any sand particle of any size at a short contact time as low as several milliseconds (Dai and Chung, 1995). Hence, the fundamental studies on the interactions between oil and sand are particularly important to investigate which parameters could influence the oil detachment, which would then subsequently aid the flotation process in the removal of oil contaminant from contaminated sands.

The interactions study between oil and silica particles, which was initially performed by Dai and Chung (1995) via a bitumen pickup test, showed that the bitumen–sand interaction was affected by pH, particle size, temperature and solvent dilution. Liu et al. (2003, 2005) also measured the interaction forces between bitumen, silica, clays and fines using atomic force microscope (AFM) and found that the energy of adhesion between the oil–sand components are affected by pH of solution, calcium ions additions, salinity and temperature (Liu et al., 2003; Liu et al., 2005). Similar AFM studies were also conducted by Zhou et al. (2006) on the effect of calcium and magnesium cations, surfactants, and the combination on sand–bitumen interactions in industrial-plant process water.

From past studies, it can be observed that there have been limited studies in the area of liquid–solid detachment, particularly in the separation of high density oil from sand particles. Thus, the aim of this study focuses on the effect of temperature, pH, sand particle size and wettability in the oil–sand interaction via an oil–sand detachment test, which is independent of the effects of bubbles. These results are also explained using the zeta potential measurements, and verified via the Derjaguin–Landau, Verwey–Overbeek (DLVO) theory. The implications of these results are then imposed to the understanding of the flotation technology used during the removal of bunker oil from contaminated sand.

2. Experimental work

2.1. Materials

Sand samples were obtained from a clean designated site from the shores of Port Klang, Malaysia. The chemical used to adjust the slurry pH is analytical grade sodium hydroxide solution (NaOH, Sigma-Aldrich). High density bunker oil was obtained from KIC Oil Terminals in Port Klang, Malaysia. The type of oil used was fuel oil with a maximum viscosity of 380 cSt.

2.2. Experimental methodology

2.2.1. Oil characterization

The oil density was evaluated using a Mettler–Toledo Density Meter DM40, while the oil dynamic viscosity was evaluated using a Anton Paar Stabinger Viscometer SVM3000 over a range of temperature. The flash point was measured using a Normalab NPM 440 Pensky-Martens Flash Point Tester, while the oil calorific value was evaluated using the Perr 6100EF semi-auto-bomb calorimeter. The results reported are an average of three consecutive readings.

2.2.2. Oil–sand detachment test

Oil–sand detachment tests were carried out using a specially fabricated crank-shaft mechanism as demonstrated in Fig. 1. The 4cm × 4cm square plate was coated with a layer of oil, and was subsequently driven down onto a sand bed of a specific particle size ranging from 0.125 mm to 1 mm and specific wettability ranging from 0 wt% to 8 wt%, for a contact time of 2 s. The plate which is now coated with a layer of sand was then submerged into a test solution of specific pH and temperature ranging from pH 6 to pH 14 and 20 °C to 60 °C respectively. The plate was then oscillated at an amplitude of 2 cm and 2.5 Hz for 2 min to detach the sand attached to the oil layer. The detached sand particles were collected, while the remaining attached sand samples were collected by scrapping them off the square plate. The experiment was repeated to collect 5 replicate samples which was subsequently combined and washed using chloroform to remove the residual bunker oil and to obtain the net weight of the sand particles which were detached from the bunker oil. The chloroform-washed sand samples were dried in a pre-heated oven at 100 °C overnight, and the weight of the dried sand particles were then obtained.

The results of the oil–sand detachment test are expressed as sand detachment efficiency (%), and are calculated as a weight ratio of detached sand particles over total initial sand particles attached on the square plate as

$$\text{Detachment efficiency}(\%) = \frac{\text{Weight of detached particles }(g)}{\text{Weight of total particles (attached + detached) }(g)} \times 100\% \quad (1)$$

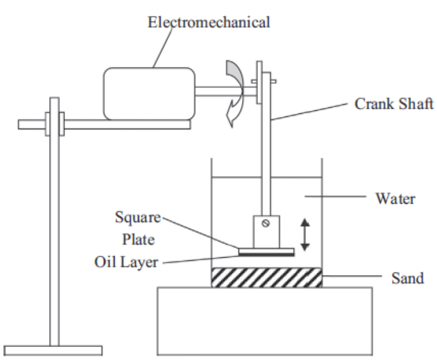

Fig. 1. Schematic diagram of crank–shaft mechanism for oil–sand interaction studies.

2.2.3. Zeta potential measurements

The electrokinetic phenomenon was studied to experimentally determine the zeta potential, in place of the immeasurable surface potential, ψ_o. The zeta potential represents the surface charge of the particles whereby a positive/negative charge shows an attraction model while a negative/negative charge demonstrates a repulsive model. The zeta potential distribution measurement was carried out using a Zeta Sizer Nano S90 (Malvern Instruments Ltd).

The zeta potential measurements of both oil and sand mediums were measured by first preparing both mediums in suspensions. The oil droplet suspensions were obtained by shearing 0.01–0.05 wt% of oil droplets using a high speed stirrer in 1 kg of prepared solution for 30 min. Likewise, the fine sand particles suspension was also prepared by stirring 0.01–0.05 wt% of sand particles using a high speed stirrer for 30 min. The oil droplets and sand particles suspensions were allowed to cream for 15 min prior to zeta potential readings. The temperature during the measurements was maintained at 25 °C, with the exception of zeta potential readings with respect to temperature whereby the equilibrium temperature was set to the required temperature and allowed to stabilize for 30 min prior to experiment runs. Each test was replicated five independent times.

2.2.4. Oil flotation studies

To evaluate the effect of operating parameters on the flotation efficiency of oil from contaminated sand, flotation studies were carried out at different temperatures, pH and wettability with the aid of microbubbles generated using a venturi system in a closed loop system as shown in Fig. 2. A known mass of bunker oil (200 g) was introduced into a 5 l beaker containing 500 g of soil sample with constant particle size ratio, and the mixture is allowed to mix homogeneously overnight before analytical use.

The experiments were conducted at different temperatures (°C), pH and wettability (%) in the sand sample as per the sand detachment tests.

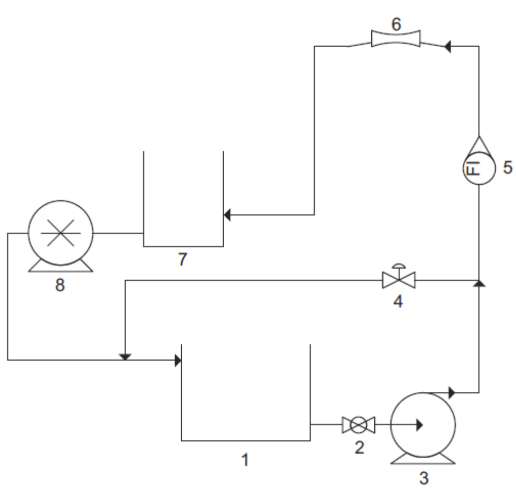

Fig. 2. Schematic diagram of flotation experiment setup using microbubbles [1 – water source, 2 – ball valve, 3 – centrifugal pump, 4 – diaphragm valve, 5 – rotameter, 6 – venturi, 7 – customized flotation cell, 8 – peristaltic pump].

Table 1
Oil characteristics at 25 °C

Density	kg/m³	976.5 [0.7]
Viscosity	mPa s	5247.2 [203.3]
Calorific value	MJ/kg	41.6 [0.2]
Flash point	°C	97.2 [0.6]

*Values are average of triplicates while values in bracket [ ] represent the standard deviation of the data.

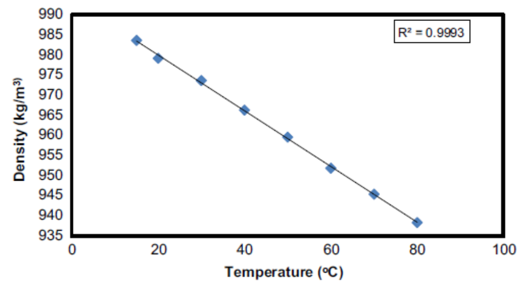

Fig. 3. Effect of temperature on bunker oil density.

3. Results and discussion

3.1. Oil characteristics

Table 1 summarizes the bunker oil characteristics at 25 °C. Figs. 3 and 4 show the effect of temperature on the density and viscosity of oil respectively. An increase in temperature showed a linear decrease in oil density while an exponential decay trend in oil viscosity was observed.

3.2. Oil–sand detachment test

Fig. 5 shows the effect of wettability on the sand detachment efficiency for particle sand size of 0.125 mm. As observed, the lowest sand detachment efficiency of less than 1% occurred at 0 wt % water content. This agreed well to Hupka et al. (2008) and Painter et al. (2010) who both noted that "oil-wet" sand or 0 wt% water content in sand is considered to be difficult to remove due to the difficulty in dislodging oil from the oil-wet sand surfaces. Nonetheless, the sand detachment efficiency was then observed to increase by approximately 40% with the increase of water content from 2 to 8 wt%. The presence of water indicates that the oil contaminant was not in direct contact with the sand due to the presence of a thin film of water. Thus, the attachment forces between sand particle and oil weakens with the increase in water content which leads to higher removal efficiencies. However, further increase in water content in sand from 8 wt% to 12 wt% showed insignificant change in sand detachment efficiency.

Fig. 6 shows the effect of temperature on the oil–sand detachment test with respect to particle size at the pH 7 under oil-wet conditions. Similar trends are observed in all sand particle size whereby the increase in temperature from 20 °C to 60 °C increases the detachment efficiency of sand. High detachment efficiencies of approximately 70% and 59% were observed in sand particle size of 1 mm and 0.5 mm respectively with increase in temperature up to 60 °C. High operating temperatures would increase the sand detachment efficiency due to the effect of temperature on the characteristic of the oil. An increase in the temperature decreases

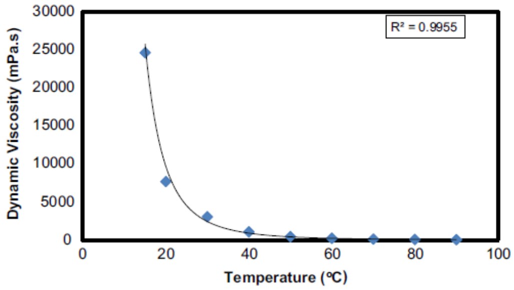

Fig. 4. Effect of temperature on bunker oil viscosity.

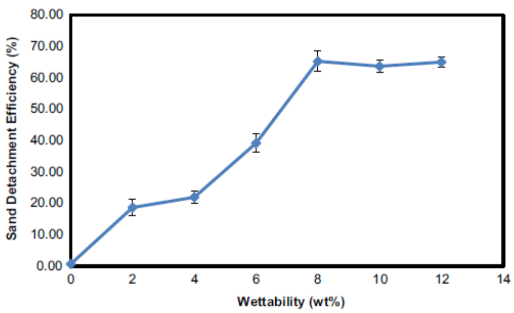

Fig. 5. Sand detachment efficiency (%) as a function of wettability (wt%) at pH 7 and room temperature.

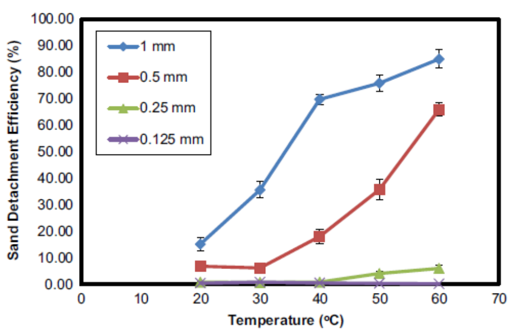

Fig. 6. Oil-wet sand detachment efficiency (%) as a function of temperature (°C) with respect to different sand particle sizes at constant pH 7.

the oil density (Fig. 3) and viscosity (Fig. 4), which therefore reduces the adhesiveness of oil and therefore leads to an increase in sand detachment efficiency as observed (Alomair et al., 2014). In industrial practices, high temperatures are favored for the recovery of oil as it is able to enhance oil recovery in minimal amount of time (Pathak et al., 2011; Lu et al., 2014).

On the other hand, the increase in temperature does not appear to significantly affect the detachment efficiency of sand of smaller sizes. A low detachment efficiency of 6% and 0.25% was observed for 0.25 and 0.125 mm sand particle size respectively.

The effects of pH on oil–sand (oil-wet) detachment test with respect to sand particle size are shown in Fig. 7. The detachment efficiency was observed to increase drastically beyond pH10. This

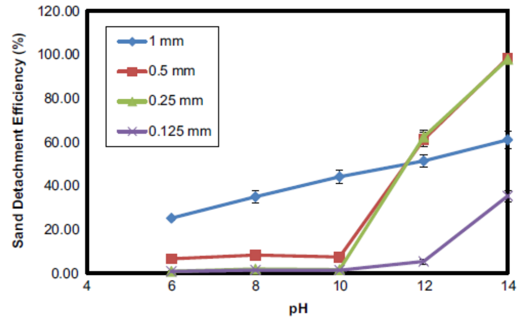

Fig. 7. Oil-wet sand detachment efficiency (%) as a function of pH with respect to different particle sizes at room temperature.

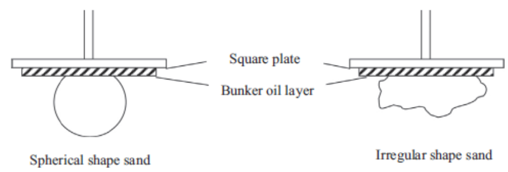

Fig. 8. Schematic diagram of sand attachment on oil layer with respect to particle shapes.

is suggested to be attributed to the release of natural surfactants from saponification reaction between NaOH and oil. This results in the weakening of interaction forces between oil and sand particles due to the lower surface tension at oil/NaOH solution interface at high pH, which therefore increases the detachment efficiency (Almalik et al., 1997; Türksoy and Bağci, 2000). Similar to Fig. 6, finer particles (0.125 mm) exhibit a lower detachment efficiency as compared to coarser particles.

From Figs. 5 to 7, similar trends were observed whereby coarser sand particles exhibit greater sand detachment efficiency compared to finer sand particles. This could be attributed to the sand surface area available for immediate contact with oil. Under the assumption that the sand particles are perfect spherical shaped, fine sand particles have a larger total sand surface area available for oil contact as opposed to coarse sand particles. Therefore, the total absolute amount of oil attached to the sand particles is greater for fine sand particles than coarse sand particles. The increase in the absolute amount of oil attached to sand particles increases the attachment forces between the oil and sand, which therefore leads to the decrease in the sand detachment efficiency for fine sand particles as observed.

However, at pH 12 and 14, the detachment efficiencies of sand particle size of 1 mm was lower compared to the detachment efficiencies at similar pH for 0.5 mm and 0.25 mm sand particle sizes. This is suggested to be attributed to irregularity in the coarse sand particle shapes as depicted in Fig. 8. The experimental coarse sand particle samples are not perfectly spherical and possess irregular shapes with flat surfaces Therefore the irregular shaped surface tends to adhere to the oil layer better compared to the rounder surface, which leads to a higher surface area available for oil–sand attachment during sand pick up in the detachment test. Hence, the attachment forces between the coarse sand particles and oil are stronger due to the larger surface area available, which leads to lower sand detachment efficiencies as shown in Fig. 7.

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