ARTICLE IN PRESS

Chemical Engineering Journal xxx (2016) xxx-xxx



Contents lists available at ScienceDirect

Chemical Engineering Journal

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Short communication

Submergence patterns of floating crude oil by granular particles

Daria Boglaienko*, Berrin Tansel

Department of Civil and Environmental Engineering, Florida International University, Miami, FL, USA

HIGHLIGHTS

- Granular particles added to floating oils transform floating oils to submerged state.
- Oil thickness, particle size and addition rate affect oil submergence patterns.
- Six different zones with specific submergence patterns were identified.
- At critical thickness the floating oil can hold large amounts of particles.
- At high particle application rates oil submerges rapidly forming a vertical channel.

ARTICLE INFO

Article history: Received 27 October 2016 Received in revised form 1 December 2016 Accepted 2 December 2016 Available online xxxx

Keywords: Floating oils Granular particles Aggregation Liquid-liquid interface Oil spills

1. Introduction

Understanding the behavior of light hydrophobic liquids in water and their interaction with granular particles is needed for controlling the fate of oil spills in the environment, as well as for development of treatment processes and oil spills mitigation strategies [1,2].

The aggregation of light hydrophobic liquids with granular materials is similar to liquid marble formation, however, unlike

E-mail address: dbogl003@fiu.edu (D. Boglaienko).

http://dx.doi.org/10.1016/j.cej.2016.12.012 1385-8947/© 2016 Elsevier B.V. All rights reserved.

G R A P H I C A L A B S T R A C T



ABSTRACT

Granular particles added to floating oils can transform the floating layer to submerged state. Here we show that there is a critical thickness at which the floating oil can hold large amounts of particles by forming an elastic oil-particle bowl while keeping some particles in completely dry state. Furthermore, we observed that at high particle application rates, a vertical channel forms in water through which floating oil submerges rapidly. Transformation of floating liquid to high strength elastic state has important implications for particle induced reinforcement at liquid-liquid interfaces; while formation of liquid-in-liquid channels by granular particles enables pipeless and rapid transfer of floating liquids. © 2016 Elsevier B.V. All rights reserved.

liquid marbles, which are liquid droplets encapsulated by hydrophobic particles exposed to air [3–5], the particle-oil aggregates are hydrophobic liquids encapsulated by granular solids in water [6–8].

Encapsulation or, in our case, aggregation of hydrophobic liquids with granular particles depends on factors that can be divided into two major categories: physical and chemical properties, and application mode. Physical properties of hydrophobic liquids, such as density, viscosity, and surface and interfacial tension affect aggregation rates with granular materials [6,9]. Particles size, wettability, and surface morphology are related to aggregation efficiency [7,10], and capillary cohesion [11]. Chemical composition of both liquids and granular materials influences transport and

Please cite this article in press as: D. Boglaienko, B. Tansel, Submergence patterns of floating crude oil by granular particles, Chem. Eng. J. (2016), http://dx. doi.org/10.1016/j.cej.2016.12.012

^{*} Corresponding author at: 10555 W. Flagler Street, Florida International University, EC 3781, Miami, FL 33174, USA.

positioning of particles at the hydrophobic liquid and water interface [12]. Application mode is a less studied category of parameters, as it depends on the experimental design and test conditions. Particle to liquid ratio [2], or volume fraction [8], has been considered as the most important application factor for the particle-oil formation. However, application rate has not been evaluated for this type of particle and oil interactions. Application rate (mass of particles per unit of time) was controlled in this study as the main parameter. Different submergence and aggregation patterns of oil occurred with granular particles (i.e., vertical liquid bridging, or roping state, and capillary bridging for particle retention, or bowl state). Thus, the main objective was to investigate the interactions of granular particles, which were applied at different flow rates, with floating oil layer of different thicknesses.

Regarding the application mode of particles, different flow regimes have been studied which affect mobility characteristics of the granular particles [13–16]. These flow regimes have been classified as solid, liquid, and gaseous types depending on interaction of particles [13]. Behavior of particles is very different in each flow regime and in the transition states (i.e., jamming transition from liquid to solid flow [16]). However, the interaction of the granular particle flow with immiscible liquids involving a hydrophobic liquid–water interface and different submergence patterns have not been reported in the literature.

The investigation of submergence patterns of floating oil by granular particles opens up the possibilities for transformation of light hydrophobic liquids to different states. These transformations can be used to explore novel technologies for capturing and stabilizing light hydrophobic liquids in a controlled manner. Further investigations of the particle-oil interactions can lead to development of efficient treatment methods which can be as effective as the conventional methods employed for oil and grease removal [17]. Additionally, the submergence patterns and characteristics of the particle-oil aggregates provide insight into the mechanisms for breaking the floating oils under the influence of surface tension and gravity forces.

2. Materials and methods

Experiments were conducted with South Louisiana crude oil (MC 252), obtained from the BP America Production Company (Houston, TX), and quartz sand with two different particle sizes. Pure quartz sand with fine particle size (40–100 mesh that corresponds to particle size range of 0.42–0.15 mm; average 0.28 mm) was obtained from Acros Organics; pure quartz sand with medium particle size (20–30 mesh with particle size range of 0.85–0.60 mm; average 0.72 mm) was obtained from Spectrum Chemical MFG.

Submergence experiments were performed in a 50 mL beaker filled with tap water and placed on an electronic balance to monitor the addition rate of the particles. Quartz sand was applied to the floating South Louisiana crude oil layer through a funnel equipped with a stopper. The application rate of sand was controlled by using funnels with different size openings. The distance from the funnel opening to the floating oil surface was maintained the same (8 cm). The experiments were carried out at the room temperature (22 °C).

Sand particles, reaching the floating layer, initiated instantaneous aggregation followed by retention and/or submergence of the particle-oil aggregates. Transition time for the submergence patterns varied, but for the majority of the experiments it occurred within several seconds. The granular particles passed through the floating oil without aggregation in less than one second (Supplementary Movie 1). Vertical transfer of the floating oil by roping also occurred within one second (Supplementary Movie 2). The time needed for the transition of oil from the floating to the submerged state as a small aggregate was 1–2 s (Supplementary Movie 3, first aggregate), and, as a large aggregate with a significant amount of sand retained (bowl formation), was 10 s on average (Supplementary Movies 3 and 4).

The thickness (*h*, mm) of crude oil layer was varied during the experiments. The amount (volume, *V*, mL) of oil added was estimated by mass (*m*, g) and density (ρ , g/cm³) of the oil and the radius (*r*, mm) of the oil slick ($h = V/\pi r^2$, or $h = m/\rho \pi r^2$).

Impact force of a particle was calculated as $F = m_p g h_l/d$, knowing mass of a particle $(m_p, \text{ kg})$; acceleration due gravity $(g, m/s^2)$; distance from a funnel opening to liquid surface (h_l, m) ; and travel distance of a particle in oil after impact (d, m).

3. Results and discussion

The experiments conducted with crude oil and granular sand particles showed that there were six distinct submergence patterns of the particle-oil aggregates depending on the floating oil thickness, particle size, and particle addition rate (Table 1). These patterns were described as pass through (particles coated with thin oil film and submerged discretely), small aggregates (particles

Table 1

Submergence patterns of floating crude oil with granular particles. Identified regions refer to Fig. 1a.

Submergence pattern	Dominant effects and forces	
Pass through (Region I)	Gravitational	
Small aggregates (Region IV)	Gravitational Cohesion Capillary bridging	
Small and large aggregates (Regions IV and V)	Gravitational Cohesion Capillary bridging Viscous stretching and pinch off	
Bowl formation (Region II)	Gravitational Cohesion Capillary bridging Viscous stretching (no pinch off) Elastic/Plastic stretching	
Aggregates followed by bowl formation (Region III)	Gravitational Cohesion Capillary bridging Viscous stretching and pinch off Followed by: Viscous stretching (no pinch off) Elastic/Plastic stretching	
Roping (Region VI)	Gravitational Cohesion Inertial effects and jetting Viscous stretching (no pinch off)	7

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