## **ARTICLE IN PRESS**

#### Marine Pollution Bulletin xxx (2014) xxx-xxx

Contents lists available at ScienceDirect

# Marine Pollution Bulletin

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

## Instantaneous stabilization of floating oils by surface application of natural granular materials (beach sand and limestone)

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#### ARTICLE INFO

Article history: Available online xxxx

Keywords: Oil spills Granular particles Aggregation Oil separation Floating oil Natural particles

#### ABSTRACT

When granular materials are applied to hydrophobic liquids floating over another liquid (i.e., water), particles form aggregates which can be separated from the floating phase. This concept can be used for controlling mobility of floating oils, especially after oil spills near coastal areas. The objectives of this research were to characterize oil capture efficiency and determine effectiveness of particles for converting the floating phase to a heavier phase for effective separation. Experiments were conducted with South Louisiana crude oil contaminated salt water, limestone and quartz sand. Although the oil removal efficiency increased with the increasing amount of granular material applied, it did not increase linearly. About 50% of the floating oil was removed by aggregates, regardless of the material used, when granular material to floating oil ratio was about 1 g/g. The aggregates separated had higher amounts of oil content when smaller amounts of granular materials were added.

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#### 1. Introduction

Addition of a small amount of liquid to dry granular particles changes their behavior significantly as the liquid forms bridges between the particles. The process may be described by induced cohesion between particles due to the surface tension of the liquid (Fingerle and Herminghaus, 2008; Mitarai and Nori, 2006; Herminghaus, 2005; Iverson and Vallance, 2001); as adhesion takes place due to solid and liquid bridges (Simons, 1996; Rossetti et al., 2003); or by capillary forces (Abkarian et al., 2013); or due to free surface energy changes (Bhushan, 2003). Similarly, the reversal to the process (i.e., adding granular particles to liquid) also results in changes in the characteristics of the liquid when the small amount of granular material is added into the liquid phase. The primary bonding mechanisms between the granular particles and liquid include: 1. adhesion and cohesion forces in the liquid phase molecules with granular particles; 2. interfacial forces in liquid phase with granular particles; 3. formation of liquid bridges and coating of granular particles; 4. attractive forces between granular particles and liquid molecules; and 5. physical interlocking of hydrophobic liquid the granular particles due to surface tension.

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http://dx.doi.org/10.1016/j.marpolbul.2014.12.020 0025-326X/© 2014 Elsevier Ltd. All rights reserved. Researchers Abkarian et al. (2013) define such oil-particle aggregates as rafts that are a close-packed layer of particles in oil formed due to long-range capillary attractions. The raft sinks when the number of particles is large enough to alternate the lower density of oil and the balance between buoyancy force and capillary force is changed.

Crude oil sedimentation by adhesion of hydrophobic liquid after collision with solid particles is a natural mechanism, which can be utilized for phase separation of floating oils. The phenomenon is similar to the interaction of suspended particles and their flux through the hydrophobic phase, which occur in the environment. In muddy waters with sediment concentration over 0.5 kg/m<sup>3</sup>, oil sedimentation may exceed normal dispersion. Oil droplets that adhere to suspended particles are considered to be removed from the oil slick (Lehr et al., 2002). According to Payne et al. (1987), at suspended particles concentration 1-10 mg/L, no oil sedimentation occurs; at 10-100 mg/L, oil sedimentation in the sufficient turbulence mixing; and at >100 mg/L, massive oil transport may occur. Payne et al. (1987) developed a mathematical model for oil droplets and suspended particulate matter interaction kinetics using an experimental setup consisting of a stirred tank with suspended oil droplets and particulate matter. Two main mechanisms considered were the interaction of oil sorption on suspended solids and oil collision with suspended solids. Similarly, collision of granular particles with floating oil layer (rather than the dispersed oil droplets) can be an effective mechanism for transformation of floating oils into an immobile phase by utilizing the sticky nature





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Please cite this article in press as: Boglaienko, D., Tansel, B. Instantaneous stabilization of floating oils by surface application of natural granular materials (beach sand and limestone). Mar. Pollut. Bull. (2014), http://dx.doi.org/10.1016/j.marpolbul.2014.12.020

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Granular particles Particles coated with oil

Bridging and aggregation (oil-particle-water)

Particle-oil aggregate (oil-particle)

Fig. 1. Formation of oil-particle aggregates.



**Fig. 2.** Transitioning of the floating oil phase from stable raft to unstable raft phase by addition of granular particles.

oil onto the solid particles when the granular particles mix with the floating oil layer.

For a highly hydrophobic liquid (i.e., oil) floating over another liquid (i.e., water), addition of granular particles into the hydrophobic floating phase gradually increases the density of the floating phase. As the hydrophobic liquid retains the granular particles, particles form aggregates due to the cohesive forces as illustrated in Fig. 1. As the density of the oil-particle aggregates reaches a critical point, the oil-water aggregates separate from the floating phase while retaining a significant amount of the hydrophobic phase as presented in Fig. 2. This concept can be used for controlling surface mobility of floating oils, especially during the initial stages while the floating oil layer is intact and dispersant application may not be suitable, mostly near coastal areas where transport of floating oils can significantly impact coastal ecosystem. Once the oil-particle phase separated, additional measures can be taken for remediation of the sediments or separation of the aggregates as they form.

The objectives of this research were to determine:

#### Table 1

Particle density of the granular materials used.

Granular material	No. of particles per g
Medium limestone Fine limestone Fine quartz	$\begin{array}{c} 2.9 \times 10^{3} \\ 1700 \times 10^{3} \\ 600 \times 10^{3} \end{array}$

Table 2	
Characteristics of the materials used	

Material and property	Typical value
South Louisiana crude oil	
API gravity	32.72 <sup>a</sup>
Density (g/mL)	0.856 (15 °C) <sup>a</sup>
Dynamic viscosity (cP)	10.1 (15 °C) <sup>a</sup>
Limestone	
Medium particle size	<2.00 mm and >0.300 mm
Fine particle size	<0.300 mm and >0.075 mm
Bulk density (g/cm <sup>3</sup> )	2.5 (ave) <sup>b</sup>
Quartz	
Particle size	<0.300 mm and >0.075 mm
Bulk density (g/cm <sup>3</sup> )	2.2 (ave) <sup>b</sup>

<sup>a</sup> EPA/600/R-03/072, 2003. Characteristics of spilled oils, fuels, and petroleum products.

<sup>b</sup> Manger, 1963. Porosity and bulk density of sedimentary rocks. http://pubs.usgs.gov/bul/1144e/report.pdf.

- 1. characteristics of granular materials suitable for capturing floating oils at sea;
- 2. identify natural materials which can be suitable for controlling transport of floating oils;
- 3. determine suitable oil/granular material ratios in relation to particle size;
- 4. characterize oil capture efficiency in relation to granular particle characteristics; and
- 5. determine effectiveness of natural granular particles for converting the floating phase to a heavier phase for effective separation.

Experiments were conducted with South Louisiana crude oil contaminated salt water. Limestone sand with medium and fine particle sizes and quartz sand with fine particle size were used. The characteristics of the granular particles were controlled by sieving through No. 4, 16, 50, and 270 sieves. The oil-particle aggregates were separated by filtering the solutions through a coarse fiberglass filter to determine the capture efficiencies of the granular particles in relation to the oil-particle ratios (w/w).

#### 2. Theoretical analyses

Rate of collision and adhesion of an oil droplet onto a suspended particle to produce oil-particulate agglomerate (i.e., loss or settling of free oil droplet) can be estimated by the following equation (Payne et al., 1987):

$$R = kC_o C_p \tag{1}$$

where *R* is the collision rate,  $C_o$  is oil-droplet concentration,  $C_p$  is total particulate concentration, and *k* is the rate constant for the reaction which depends on the turbulence or energy dissipation rate.

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