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# Self assembly, mobilization, and flotation of crude oil contaminated sand particles as granular shells on gas bubbles in water



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#### HIGHLIGHTS

#### GRAPHICAL ABSTRACT

- Mobilization of oil contaminated sand particles with gas bubbles was studied.
- Oil contaminated sand formed tightly packed granular shells on air bubbles.
- Oil contaminated can sand particles transferred from sediments to water column.
- Oil contaminated particles were carried as granular shells with gas bubbles.
- Gas bubbles provide a mobilization mechanism for oil contaminated sand in sediments.



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#### ABSTRACT

Contaminant fate and transport studies and models include transport mechanisms for colloidal particles and dissolved ions which can be easily moved with water currents. However, mobilization of much larger contaminated granular particles (i.e., sand) in sediments have not been considered as a possible mechanism due to the relatively larger size of sand particles and their high bulk density. We conducted experiments to demonstrate that oil contaminated granular particles (which exhibit hydrophobic characteristics) can attach on gas bubbles to form granular shells and transfer from the sediment phase to the water column. The interactions and conditions necessary for the oil contaminated granular particles to self assemble as tightly packed granular shells on the gas bubbles which transfer from sediment phase to the water column were evaluated both experimentally and theoretically for South Louisiana crude oil and quartz sand particles. Analyses showed that buoyancy forces can be adequate to move the granular shell forming around the air bubbles if the bubble radius is above 0.001 mm for the sand particles with 0.28 mm diameter. Relatively high magnitude of the Hamaker constant for the oil film between sand and air (5.81 × 10<sup>-20</sup> J for air-oil-sand) indicates that air bubbles have high affinity to attach on the oil film that is on the sand particles in comparison to attaching to the sand particles without the oil film in water ( $1.60 × 10^{-20}$  J for air-water-sand). The mobilization mechanism of the contaminated granular particles with gas bubbles can occur in natural environments resulting in transfer of granular particles from sediments to the water column.

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#### 1. Introduction

Dissolved air flotation is a well known technology for capturing colloidal particles and oil droplets at the gas-water interfaces and removal

\* Corresponding author. *E-mail address:* tanselb@fiu.edu (B. Tansel). from water. The colloidal particles or oil droplets attach to the small gas bubbles which are released from pressurized water. The small particles and oil droplets remain attached and move with the gas bubbles towards to water surface forming a froth layer which can be skimmed from the water surface. However, mobility of much larger granular particles (e.g., sand) by gas bubbles have not been considered as a possible mechanism due to the larger size of the particles and high bulk density which result in the gravitational forces to be too large in comparison to the buoyancy of the gas bubbles and inability of the particles to remain attached to the gas bubbles by relatively small forces during particle- bubble interactions (e.g., electrical forces, van del Waals forces). However, hydrophobic granular particles can remain attached to the gas bubbles in aquatic environments and can mobilize into the water column.

Oil contaminated granular particles exhibit hydrophobic characteristics which allows them to attach together forming clusters. Oil coating on particle surface can also reduce the gravitational forces if the density of the oil is less than that of water and the oil film covering the particles is thick enough. Once oil coated, and if the granular particles are small enough, large amounts of particles can remain floating on water surface indefinitely because of their hydrophobic nature (due to oil film) and aggregation of the oil coated particles which create conditions that overcome the gravitational forces.

The oil contaminated sand particles in sediments can exhibit different aggregation behaviors depending on the oil characteristics, and oil content in the sediments. The purpose of this study was to examine the possible mobilization and resurfacing mechanisms of crude oil contaminated granular particles in sediments by self assembly around gas bubbles. We examined the conditions for mobilization of submerged oil contaminated sand particles after they came into contact with air bubbles by experimental observations and theoretical analyses. Experiments were conducted to evaluate the interactions and forces necessary for the oil contaminated granular particles to self assemble around the gas bubbles and mobilize from the sediment phase to the water column. Here we show that gas bubbles can allow self assembly of oil contaminated sand particles around them to form tightly packed granular shells; therefore, mobilizing the oil contaminated granular particles from sediments to the water phase.

#### 2. Materials and methods

Experiments were conducted in 50 mL beakers which contained 40 mL of tap water and prespecified amount of South Louisiana crude oil added to water (resulting in a floating oil layer between 0.03 and 0.80 mm thickness). South Louisiana crude (SLC, MC 252) oil was obtained from BP America Production Company (Houston, TX). Quartz sand with 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 (Pittsburgh, PA, USA).

The beaker was placed on an electronic balance for monitoring the amount of sand added (Fig. 1a). Sand was added through a glass funnel equipped with a flow control valve which allowed the sand to be added at a uniform rate ( $1.6 \pm 0.1$  g/s). Sand addition was controlled such that the floating oil submerged by roping due to the small area of contact with sand. Fast submergence of oil and sand allowed air to be entrapped within the submerging oil-sand mixture, especially after most of the floating submerged and as sand entered the water phase. After all the free floating oil submerged, the sand addition was stopped. The experimental set up and the progression of the sand-oil-water interactions from one experimental run are provided in Fig. 1.

#### 3. Results and discussion

#### 3.1. Experimental observations

The floating crude oil, regardless of the thickness of the oil layer, submerged very quickly with sand addition, leaving no visible free floating



**Fig. 1.** Submergence of floating crude oil with sand addition in roping state: (a) experimental set up, (b) progression of oil submergence with sand addition (frames 1–8 were sequentially photographed). Frame 1 shows the floating crude oil layer on the water surface, frames 2–4 show submergence of oil by roping, frames 5–6 show lower oil content and increasing sand content, frames 7–8 show the formation and movement of air bubbles with granular shells (almost uniform in size) and large air bubbles without granular shell remaining attached to the oil at the bottom.

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