

Research article

Partitioning of fresh crude oil between floating, dispersed and sediment phases: Effect of exposure order to dispersant and granular materials

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ABSTRACT

When three or more high and low energy substrates are mixed, wetting order can significantly affect the behavior of the mixture. We analyzed the phase distribution of fresh floating Louisiana crude oil into dispersed, settled and floating phases depending on the exposure sequence to Corexit 9500A (dispersant) and granular materials. In the experiments artificial sea water at salinity 34‰ was used. Limestone (2.00–0.300 mm) and quartz sand (0.300–0.075 mm) were used as the natural granular materials. Dispersant Corexit 9500A increased the amount of dispersed oil up to $33.76 \pm 7.04\%$. Addition of granular materials after the dispersant increased dispersion of oil to $47.96 \pm 1.96\%$. When solid particles were applied on the floating oil before the dispersant, oil was captured as oil-particle aggregates and removed from the floating layer. However, dispersant addition led to partial release of the captured oil, removing it from the aggregated form to the dispersed and floating phases. There was no visible oil aggregation with the granular materials when quartz or limestone was at the bottom of the flask before the addition of oil and dispersant. The results show that granular materials can be effective when applied from the surface for aggregating or dispersing oil. However, the granular materials in the sediments are not effective neither for aggregating nor dispersing floating oil.

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1. Introduction

Dispersion of floating crude oil is one of the main processes linked to the degradation of the spilled oil in marine environment. This process has been extensively studied, resulting in development of chemical dispersants and their use after large oil spills as a spill response practice.

Besides mechanical dispersion (wave turbulence), chemical dispersion (surfactants, or dispersants), interaction of oil with mineral particles and sediments has been recognized as an important factor in oil dispersion. Oil-mineral aggregates may eventually settle trapping oil on the bottom (Lee, 2002; Sterling et al., 2004; Gong et al., 2014). Moreover, fine sediments on shorelines add to natural oil removal (surf-washing) by formation

of oil-mineral aggregates (Jahns et al., 1991; Bragg and Owens, 1995; Stoffyn-Egli and Lee, 2002). A method that uses dispersion of mineral solid particles in aqueous phase for oil removal via formation of buoyant particle-oil flocs was patented (Bragg and Yang, 1996). Direct application of granular materials on the floating oil surface, followed by immediate oil sedimentation and stabilization, has been recently studied and identified as a potential oil spill remediation method (Abkarian et al., 2013; Boglaienko and Tansel, 2015).

Gordon et al. (1973) reported that adsorption of oil on the surface of suspended particulate matter may result in removal of oil from the water surface by 87–98%. Later, Muschenheim and Lee (2002) reviewed and discussed the role of additional to adsorption processes that influence surface oil removal by particulate matter, including flocculation of fine particles with surface oil. Passow et al. (2012) pointed on marine snow formation in the Gulf of Mexico after the Deepwater Horizon accident and the role of coagulation of oily particulate matter. In general, two main oil-particle interactions have been specified: adsorption of hydrocarbons and direct aggregation of oil with particulate matter (Gong et al., 2014; Bandara et al., 2011; Lee, 2002). A numerical model

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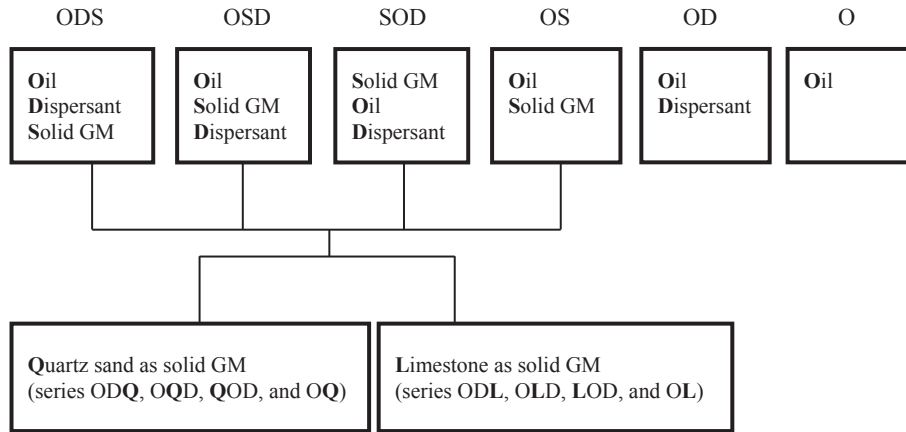


Fig. 1. Experimental design (O: crude oil, D: dispersant, S: Solid granular material, GM: granular material).

that simulates oil and suspended sediment interaction, and estimates the fraction of removed oil was presented by Bandara et al. (2011), according to which the amount of oil removed as oil-sediment aggregates can reach up to 65%.

Colloidal solid particles (clay) can attach on oil droplet surfaces (Dommersnes et al., 2013). At high clay concentrations (above about 2% w/w), the droplet surface can be covered by the solid particles. The chemical dispersants decrease the oil-particle interaction. Physically dispersed oil (i.e., emulsion formed by mixing) associates easily with particulate matter in comparison to the chemically dispersed oil (i.e., using dispersants) (Mackay and Hossain, 1982; Payne et al., 2003). Similarly, another study showed that addition of a dispersant decreased the adsorption of oil to suspended particulate matter (Sørensen et al., 2014). Dispersant Corexit 9500A contains anionic surfactants, nonionic surfactants, and organic solvents (Nalco, 2014). Such nonionic components of the Corexit dispersants as Tween 80 adsorbs to oil droplets irreversibly, resulting in long coalescence time (Reichert and Walker, 2015).

High energy substrates are more easily wet than low energy substrates (De Gennes, 1985). Also the wetting process is more complete when the substrate has a relatively higher surface energy than the liquid (Kern et al., 1986). Addition of a surfactant to water decreases wetting and adhesion work of high energy polar solid materials (i.e., quartz) (Zdziennicka and Jańczuk, 2010). A dispersant, designed to lower surface tension (surface free energy), contributes to smaller rates of oil-mineral aggregation, as it becomes less beneficial (i.e., smaller decrease in free energy) for the system to go from “oil and solids in water” to “oil-solid aggregates in water.”

$$\Delta G = \gamma_{so} - \gamma_{wo} - \gamma_{sw} \quad (1)$$

where γ is surface free energy ($\gamma_i = \partial G / \partial A_i$, A is area); subscripts: s is solids, o is oil, and w is water.

For the system to experience decrease in free energy for oil-solids aggregation (i.e., $\Delta G \leq 0$):

$$\gamma_{so} \leq \gamma_{wo} + \gamma_{sw} \quad (2)$$

Addition of the dispersant decreases γ_{wo} , so that condition (2) may not be satisfied.

The change in surface free energy, ΔG , can be described in two steps: 1. ΔG_1 for activation of energy barrier to the adhesion of solid particle to oil, and 2. ΔG_2 for work of adhesion, w_a (Adamson, 1990). These steps can be expressed as follows:

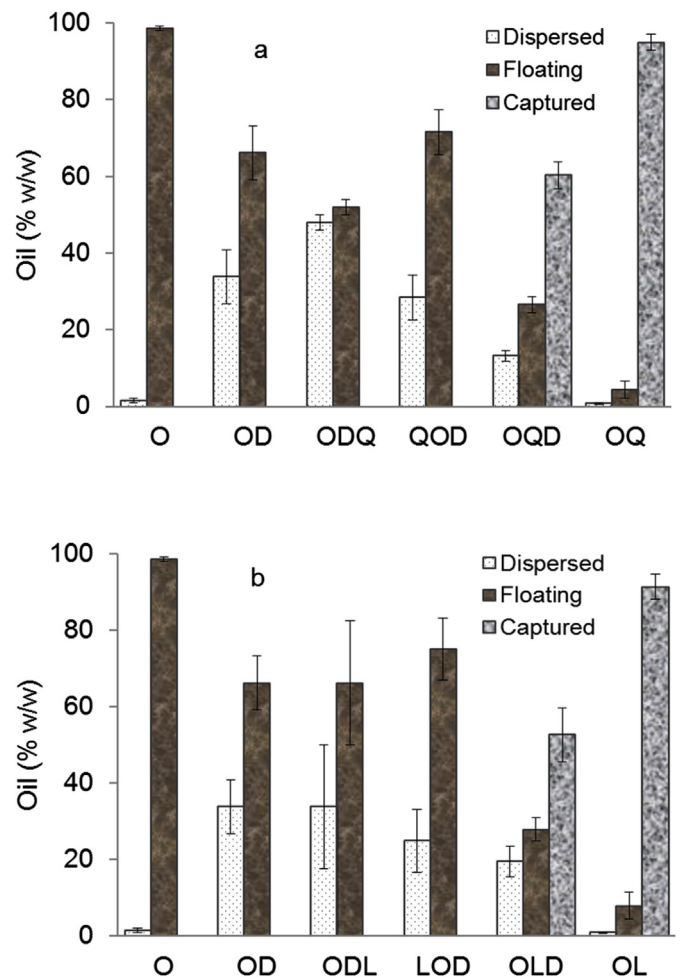


Fig. 2. Phase distribution of oil depending on the presence and absence of the dispersant Corexit 9500A and granular materials (error bars: ± 1 SD): a. fine quartz used as solid granular material, b. Medium limestone used as solid granular material.

$$\Delta G = \Delta G_1 + \Delta G_2 \quad (3)$$

where,

$$\Delta G_1 = \gamma_{wo} \Delta A_{wo} \quad (4)$$

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