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Enhanced microbubbles assisted cleaning of diesel contaminated sand

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ABSTRACT

In this article, we investigated the effect of low intensity pulsed ultrasound (US), temperature and salinity on cleaning efficacy of fine bubbles with diameter < 50 μm for diesel contaminated sands. About 47% and 76% diesel removal was achieved from 10% (w/w) diesel contaminated fine and medium sands respectively, after 30 min treatment with 40 kHz low intensity intermittent pulsed US together with MBs in contrast to 41% and 68% diesel removal while treatment with MBs alone. The effect of high temperature was found to be prominent during the initial stages of cleaning. In addition, MBs generated in 599 mM saline water efficiently removed 85% diesel from fine sand within 30 min in contrast to only 41% diesel removal with MBs in fresh water. This study provides evidence for developing highly efficient MBs based chemical free technology for diesel contaminated sediments.

1. Introduction

Recently, microbubbles (MBs) with diameter < 50 μm also known as fine bubbles have been explored as a chemical free cleaning technology for removal of press oil from steel plate (Tano et al., 2013), cleaning of human skin (Lee et al., 2013) and biologically fouled membranes (Agarwal et al., 2012a) etc. Characteristic physicochemical properties of MBs such as their proficiency to generate pressure waves while contracting and collapsing under water surface (Takahashi et al., 2007), large interfacial surface area and high negative zeta potential (Agarwal et al., 2011; Takahashi, 2005) qualifies them for numerous cleaning applications. Recently, the combination of low intensity pulsed ultrasound (US) with MBs have shown great potential over the use of MBs alone for cleaning of biologically fouled membrane surface (Agarwal et al., 2014). Factors assisting in enhancing the cleaning efficiency of MBs with diameter < 50 μm generated without adding any chemical for remediation of diesel contaminated sand is still unknown. Though water temperature (Urum et al., 2004) and salinity (Wang et al., 2010) have been known to boost the efficiency of physical cleaning for diesel contaminated sediments, the effect of temperature and salinity in combination with MBs is yet to be explored. In this article we investigated the effect of temperature, salinity and low intensity pulse US of 40 kHz on cleaning efficiency of MBs for diesel contaminated sands.

Contamination of marine sediments by petroleum hydrocarbon is a widespread environmental problem (Schrope, 2010). For remediation of diesel contaminated sediments several physicochemical methods have been employed however these methods suffer from one or more drawbacks (Agarwal and Liu, 2015). Among all the remediation techniques, bioremediation remains as the preferred technique for cleaning of polluted sediments though it suffers from drawbacks such as slow response in cold climatic conditions and limited application for degradation of biorefractory contaminants (Goi et al., 2006). A novel eco-friendly technology is urgently required for cleaning of diesel contaminated sediments. In this article, we explored the effect of low intensity intermittent pulse US, temperature and salinity for improving the cleaning efficiency of MBs with diameter < 50 μm for diesel contaminated fine and medium sands. It is expected that optimization of these parameters would be helpful in future for further developing an on-site, highly efficient chemical free cleaning technology based on collapsing air MBs for remediation of diesel contaminated sediments.

2. Materials and method

2.1. Preparation of diesel contaminated sand

Ultra-low sulphur diesel (10 ppm) was mixed with adequate quantities of pre-washed and dried fine sand (grain size range 0.15–0.25 mm; d_{50} 0.18 mm) and medium sand (grain size range 0.25–0.60 mm; d_{50} 0.39 mm) separately, to obtain 10% (w/w) diesel-

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sand mixtures. For optimum adsorption of diesel over sand surface, diesel-sand mixtures were mixed vigorously for 30 min and then left untouched for 24 h for ageing.

2.2. Experimental setup

A MB generator (model: AS-MA5S; make: Riverforest Corporation, USA) capable of generating MBs with diameter < 50 μm at a flow rate of 220 ml/min was used to treat diesel contaminated sands. The generation of MBs in tap water developed milky appearance which was attributed towards reflectance of light through millions of MBs with diameter < 50 μm (Agarwal et al., 2012b). Diesel contaminated sand (10% (w/w)) was treated with MBs in a glass column reactor with an internal diameter of 3 cm and a height of 25 cm. Treated sand samples were collected after every 10 min and analysed for residual diesel. All the experiments were conducted thrice unless otherwise stated.

a) *Treatment of diesel contaminated sand with MBs + US method:*

Glass column reactor filled with diesel contaminated sand was placed in an ultrasonic bath (model: ALDS-40200-60H-C; make: Alstron Pvt. Ltd., Singapore) capable of generating programmable pulsed US of 40 kHz. Fig. 1 shows schematic of the experimental setup for remediation of diesel contaminated sand with the combined effect of US and MBs. Intermittent pulsed US of 3 s were initiated after every 57 s while MBs generated in tap water at room temperature were continuously introduced at a flow rate of 220 ml/min from the bottom into the column reactor. An optimal time of 3 s was chosen for ultrasonic pulse as this was sufficient enough to collapse all the MBs in the column reactor while the entire column could be filled up with MBs in 57 s. Nine trials were carried out for the above experiment.

b) *Treatment of diesel contaminated sand with MBs at different temperatures:*

Glass column reactor filled with diesel contaminated sand was placed in a temperature controlled water bath while the MBs were continuously introduced into the column reactor at a flow rate of 220 ml/min. The schematic of this experimental setup was similar to the one used for MBs + US method except the ultrasonic bath was replaced by a temperature controlled water bath. The experiments were conducted while maintaining the temperature of the water bath at 25 and 65 °C.

c) *Treatment of diesel contaminated sand with MBs in saline water:*

Saline water (599 mM) mimicking salinity equivalent to that of seawater was prepared by dissolving adequate amount of sodium chloride (AR grade) in tap water. MBs generated in saline water at room temperature were continuously introduced into the column reactor at a flow rate of 220 ml/min for the treatment of diesel contaminated sand. The schematic of the experimental setup for this method was similar to Fig. 1 excluding ultrasonic bath.

2.3. Analytical methods

Diesel content of the treated sand samples was determined by solvent extraction technique and cross checked by thermogravimetric (TG) analysis. For this purpose, 0.1 g of diesel contaminated sand sample was treated with 10 ml n-hexane (Sigma-Aldrich, HPLC grade) and vortexed for 5 min. Samples were then centrifuged at 3000 rpm for another 5 min to remove any suspended particles. Absorbance was measured at 264 nm (Levy, 1972) using UV–Visible spectrophotometer (model: UV-1800; make: Shimadzu, Japan). Percentage diesel removal was determined by Eq. (1):

$$\text{Oil removal (\%)} = ((O_i - O_r)/O_i) \times 100\% \quad (1)$$

where O_i and O_r are initial and remaining oil in the oil-sand mixtures, respectively.

TG analysis of the diesel contaminated sand was performed by PerkinElmer TGA 4000 equipped with Pyris™ software. For this purpose, nearly 20 mg of the sand sample was used for analysis. TG analysis was performed under nitrogen environment at a flow rate of 20 ml/min to prevent diesel oxidation. During analysis the temperature of the furnace was increased from 30 to 600 °C at a constant heating rate of 20 °C/min. All the data obtained was processed by Pyris™ software.

2.4. Statistical analysis

All the experiments were conducted thrice while nine trials were carried out for the treatment of diesel contaminated sand with MBs + US method. Results were expressed as mean value ± absolute deviation. Student *t*-tests were performed for analyzing the significance of results at the level of $P < 0.05$.

3. Results and discussion

Fig. 2a and b shows percentage diesel removal for 10% (w/w) diesel contaminated fine and medium sands respectively, upon treatment with MBs and MBs + US method. The percentage diesel removal from both diesel contaminated fine and medium sands was higher upon treatment with MBs + US method in comparison to treatment with MBs alone (Student's *t*-test, $P < 0.05$). For instance, after 30 min treatment with MBs + US method nearly 47% and 76% diesel removal was achieved for diesel contaminated fine and medium sands respectively, in comparison to 41% and 68% diesel removal upon treatment with MBs. Fig. 3 shows schematic of the pulsed US assisted microbubble collapse for treatment of diesel contaminated sand. It is well known that the cleaning action of US is mainly due to the formation of cavitation

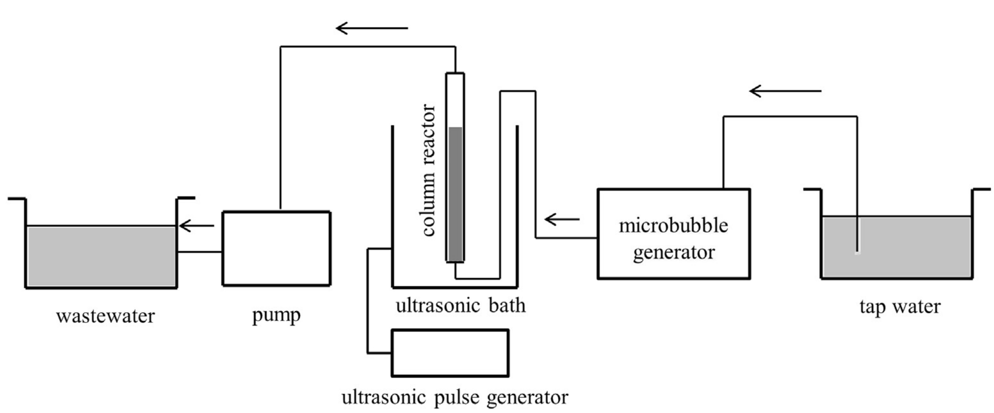

Fig. 1. Schematic of the experimental setup for cleaning of diesel contaminated sand with US and MBs.