Marine Pollution Bulletin xxx (2015) xxx–xxx

Contents lists available at ScienceDirect

Marine Pollution Bulletin

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

Review

Remediation technologies for oil-contaminated sediments

Ashutosh Agarwal [a], Yu Liu [a,b,*]

[a] Advanced Environmental Biotechnology Centre, Nanyang Environment and Water Research Institute, Nanyang Technological University, 1 Cleantech Loop, Singapore 637141, Singapore
[b] School of Civil and Environmental Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore

ARTICLE INFO

Article history:
Received 11 May 2015
Received in revised form 21 August 2015
Accepted 3 September 2015
Available online xxxx

Keywords:
Remediation
Crude oil
Sediment
Contamination

ABSTRACT

Oil-contaminated sediments pose serious environmental hazards for both aquatic and terrestrial ecosystems. Innovative and environmentally compatible technologies are urgently required to remove oil-contaminated sediments. In this paper, various physical, chemical and biological technologies are investigated for the remediation of oil-contaminated sediments such as flotation and washing, coal agglomeration, thermal desorption, ultrasonic desorption, bioremediation, chemical oxidation and extraction using ionic liquids. The basic principles of these technologies as well as their advantages and disadvantages for practical application have been discussed. A combination of two or more technologies is expected to provide an innovative solution that is economical, eco-friendly and adaptable.

Contents

1. Introduction

During the past few decades, large volumes of oil have been released into the environment due to marine crude oil tanker collisions, leakages from pipelines/storage tanks and other accidental spills (Albaigés Riera et al., 2006; Araruna et al., 2004; Schrope, 2010; Trevors and Saier, 2010). Of these, marine oil spills are very common occurrences. These oil spills not only destroy aquatic habitants but also adversely affect the health of the population near oil spill shorelines (Diaz, 2011; Major and Wang, 2012). Due to the rise in oil spills in the past few decades, the management and remediation of crude oil-contaminated sites containing a significant amount of polycyclic aromatic hydrocarbons have been a major environmental concern due to their carcinogenic nature (Labud et al., 2007; Samanta et al., 2002). Marine oil spills and inland spills usually involve heavy crude oil and light petroleum products, respectively (USEPA, 1999). While light oil tends to evaporate quickly, heavy oil forms a thick oil–water mixture commonly known as mousse (Araruna et al., 2004). On being exposed to sunlight and wave action, heavy oil forms sticky substances such as tar balls and asphalt. Carried by waves, heavy oil reaches the shorelines and sticks to sand and pebbles. On coming in contact with sandy beaches, the oil starts to percolate down the spaces between sediments up to a few centimetres. In some cases, due to the hydromorphological dynamics of the beach, oil could be buried up to a depth of 4 m, which can be

---

* Corresponding author at: School of Civil and Environmental Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore.
E-mail address: cyliu@ntu.edu.sg (Y. Liu).

http://dx.doi.org/10.1016/j.marpolbul.2015.09.010

Please cite this article as: Agarwal, A., Liu, Y., Remediation technologies for oil-contaminated sediments, Marine Pollution Bulletin (2015), http://dx.doi.org/10.1016/j.marpolbul.2015.09.010

more difficult to clean (Bernabeu et al., 2009). Some of the oil from an oil-contaminated beach is removed naturally by environmental processes such as evaporation, oxidation and biodegradation. Natural biodegradation of oil is a very slow process often requiring months or even years in comparison with evaporation and oxidation processes (Atlas and Hazen, 2011). Lightweight oil evaporates easily, whereas the majority of heavy oil rich in carcinogenic compounds remains attached to the sediments. Only the superficial oil is slowly broken down into simpler compounds via air oxidation (USEPA, 1999). Biodegradation by oil-consuming bacteria occurs slowly at the initial stages, due to the toxicity of crude oil (USEPA, 1999). As a result, the oil content of oil-contaminated sediments is significantly reduced by natural processes over a long duration. Therefore, various methods have been studied to achieve rapid and complete removal of oil from sediments.

Developing novel and effective technologies for separating oil from oil-contaminated sediments is of considerable environmental interest. In the past few decades, various physical, chemical and biological methods have been proposed for the remediation of oil-contaminated sediments (Couto et al., 2009; Do et al., 2009; Harvey et al., 1990; Shin and Shen, 2011). Some of these approaches have also been demonstrated on-site (Chien, 2012; Lee et al., 2014; Szulc et al., 2014). This review provides up-to-date information on the various approaches developed for the remediation of oil-contaminated sediments.

2. Methods for remediation of oil-contaminated sediments

2.1. Flotation and washing

For the past few decades, the flotation technique has been commonly used to separate hydrophobic minerals from hydrophilic materials (Schramm et al., 2003; Zhang et al., 2001). This inexpensive technology has been extensively used to separate bitumen from oil-contaminated sediments in the petroleum industry (Chou et al., 1998). The interfacial and electrostatic properties of the oil-contaminated sediment system are essential to oil recovery in the flotation technique (Schramm et al., 2003). The overall efficiency of the flotation process in oil recovery from oil-contaminated sediments depends on such parameters as the viscosity of crude oil, the adhesion forces between the oil and the associated sediment surface, ageing of sediment–oil mixtures and the separation temperature (Wang et al., 2010). An increase in temperature not only reduces the oil viscosity but also increases the negative zeta potential of sediment particles. This in turn enhances the repulsion between the oil and the sediment, thus enabling easier detachment of oil from the sediment (Dai and Chung, 1995). At low temperatures, the adhesion of light oil to the sediment surface plays a key role in the separation process, whereas the viscosity of heavy oil is the main governing factor in the overall flotation separation process (Urum et al., 2004; Wang et al., 2010). Nonionic surfactants effectively enhance flotation separation in saline water for both light and waxy oils; however, for heavy crude oil, the viscosity must be significantly lowered by adding hydrocarbon solvents (Wang et al., 2010). The weathering of oil-contaminated sediments significantly lowers the oil removal efficiency of the flotation process. This is caused by the evaporation of lighter oils, leaving behind heavy oil components attached to the sediment surface in their concentrated form (Urum et al., 2004).

In the early 1990s, the US Environmental Protection Agency (USEPA) investigated the hot water washing of oily sediments (Davis and Lien, 1993). This method has certain limitations: considerable energy is required to heat water to the desirable temperature; moreover, only a fraction of the oil is removed and the unattached residual oil requires other remediation techniques (Davis and Lien, 1993). Unlike the flotation separation method, the sediment-washing technology requires large amounts of surfactants, typically ranging from 0.5% to 2% significantly above the critical micelle concentration (CMC). Surfactant regeneration becomes crucial for lowering the overall cost of the washing process. However, surfactant regeneration is practically inefficient (Underwood et al., 1995). In general, the flotation separation method has a higher oil removal efficiency than the sediment-washing technology (Chou et al., 1998). This is because a stabilized oil-in-water emulsion is formed on continuously washing the oil-contaminated sediment with surfactants, making it difficult to separate from the oil–water mixture. Sediment washing further becomes ineffective when sediments are contaminated with metal ions (Ko et al., 2006) or when they contain a large amount of silt and clay particles (Van Rijt, 1993).

Several studies on the use of cationic, anionic as well as nonionic surfactants for removing oil from oil-contaminated sediments with both the flotation separation and washing methods have been reported (Ceschia et al., 2014; Deshpande et al., 1999; Duffield et al., 2003). Surfactants reduce the hydrophobicity of the oil phase so that the oil can be easily wetted by the water phase and thus detach from the sediment surface. Nonionic and anionic surfactants are preferred over cationic surfactants, as the latter bind readily with the anionic sites of sediments and are attached to the sediment surface with great adhesion forces (Chu, 2003). In addition, prolonged sediment washing with a surfactant solution often tends to form a stable oil-in-water emulsion, making it difficult to separate from the aqueous phase. To overcome this limitation, next-generation surfactants commonly known as switchable anionic surfactants (SAS) have been studied in the remediation of oil-contaminated sediment by washing (Ceschia et al., 2014). Switchable surfactants can be switched ‘on and off’ with triggers such as light, pH or redox reagents (Datwani et al., 2003; Dexter et al., 2006; Liu et al., 2006; Sakai et al., 1999; Scott et al., 2012). SASs triggered by CO_2 are promising candidates for sediment washing. These surfactants could act as both emulsion stabilizers and destabilizers in the presence and absence of CO_2, respectively (Penth, 1995). SASs are also more efficient in oil removal than non-switchable surfactants (Ceschia et al., 2014).

In contrast to synthetic surfactants, biosurfactants are considered as potential candidates for the remediation of oil-contaminated sediments through the flotation separation and washing methods due to their excellent surface activities, lower toxicity, easy availability, possible regeneration, high biodegradability and specificity (Christofi and Ivshina, 2002; Mulligan, 2005; Ron and Rosenberg, 2002). Various biosurfactants such as aescin, lecithin, rhamnolipid, saponin, tannin, surfactin and sophorolipid have been investigated in the removal of oil from oil-contaminated sediments through the washing method (Lai et al., 2009; Mnif et al., 2014; Pacheco et al., 2010; Santa Ana et al., 2007; Urum and Pekdemir, 2004). Lai et al. (2009) found that biosurfactants (rhamnolipid and surfactin) are more efficient in removing total petroleum hydrocarbons than synthetic surfactants such as Tween 80 and Triton X-100. In an another study, Urum and Pekdemir (2004) found that rhamnolipid removed as much oil as the synthetic surfactant sodium dodecyl sulphate (SDS). Batista et al. (2010) showed that biosurfactants produced by *Candida tropicalis* recovered 84% of oil from motor oil-contaminated sediments. This was considerably greater than that reported by Lai et al. (2009) for rhamnolipid and surfactin produced by *Pseudomonas aeruginosa* and *Bacillus subtilis*, respectively. Increasing the biosurfactant concentration up to the CMC increases the oil removal efficiency, due to mobilization resulting from a reduction in interfacial tension (Urum and Pekdemir, 2004). However, increasing the biosurfactant concentration above the CMC does not further enhance the oil removal efficiency, as a result of the micelle instability caused by the change in micelle shape and size (Urum and Pekdemir, 2004).

Washing oil-contaminated sediments with seawater solutions of synthetic and biosurfactants is more efficient in removing oil than surfactant solutions in distilled water (Urum et al., 2005). The addition of seawater leads to a reduction in the electrostatic repulsion of surfactants. Introduction of air bubbles while washing oil-contaminated

Please cite this article as: Agarwal, A., Liu, Y., Remediation technologies for oil-contaminated sediments, Marine Pollution Bulletin (2015), http://dx.doi.org/10.1016/j.marpolbul.2015.09.010