Environmental Technology, 2013
Vol. 34, No. 3, 407–416, http://dx.doi.org/10.1080/09593330.2012.698648

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Polycyclic aromatic hydrocarbon removal from petroleum sludge cake using thermal treatment with additives

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(*Received 12 September 2011; final version received 25 May 2012*)

Petroleum sludge is a hazardous waste that contains various organic compounds including polycyclic aromatic hydrocarbons (PAHs) which have carcinogenic–mutagenic and toxic characteristics. This study focuses on the thermal treatment (indirect heating) of petroleum sludge cake for PAH degradation at 250, 450, and 650 °C using $Ca(OH)_2 + NaHCO_3$ as an additive. The treatment was conducted in a rotary drum electric heater. All experiments were carried out in triplicate. Concentrations of the 16 priority PAHs in gas (absorbed on Amberlite XAD-4 adsorbent), particulate (on quartz filter) and residue phases were determined using gas chromatography-mass spectrometry (GC-MS). The samples were extracted with acetonitrile by ultra-sonication prior to GC-MS analysis. The use of additive was beneficial and a temperature of 450 °C was suitable for PAH degradation. Low levels of PAH emissions, particularly carcinogenic PAH and toxic equivalent concentration (ΣTEC), were observed in gas, particulate and residue phases after treatment.

Keywords: petroleum sludge cake; polycyclic aromatic hydrocarbons; additive; thermal treatment; indirect heating

Introduction

Petroleum refineries produce huge amounts of waste sludge worldwide. The sludge is included in the hazardous waste listings of the Resource Conservation and Recovery Act (RCRA) [1]. In Malaysia, a typical petroleum refinery with a production capacity of 105,000 barrels per day produces about 50 tons oil sludge per year [2]. Due to its hazardous nature, the sludge has to be suitably treated prior to discharge into the environment. Typically, oil sludge contains more than 33% total petroleum hydrocarbons (TPH) with about 550 mg/kg polycyclic aromatic hydrocarbons (PAHs) [3]. PAHs are known for their toxic and carcinogenic–mutagenic characteristics. The US Environmental Protection Agency (USEPA) has identified 16 PAHs as priority compounds (Table 1) [3]. The toxic equivalence factor (TEF) of PAHs reflects their relative carcinogenic potential with respect to the specific compound benzo[a]pyrene (BaP) [4,5].

The threat posed to the environment by petroleum sludge disposal has led to conventional disposal techniques such as land-farming to be banned in certain countries [6]. Biological treatment (bioremediation) uses microbes or living organisms for the removal of contaminants. Using a batch aerated bioreactor, Soriano and Pereira [7] achieved 83% PAH removal from petroleum sludge in two weeks of treatment. Harmsen *et al.* [8] conducted a field-scale land-farming experiment on petroleum sediment for over 10 years. The desorbable PAH results showed about 90, 75, 55 and 28% for 3-, 4-, 5- and 6-ring PAHs respectively. Kriipsalu *et al.* [9] obtained 72% PAH degradation in 62 days using an aerated static pile for composting oily sludge amended with remediated oil-contaminated soil and non-mature garden waste compost. The phytoremediation of oil sludge using rye, crested wheatgrass, and couch grass reduced PAHs by 44, 37, and 36% respectively in 72 days [10]. Bioremediation can treat a wide range of organic compounds; low molecular weight (LMW) PAHs are removed more extensively than high molecular weight (HMW) PAHs [11]. Zahed *et al.* [12] concluded that bioremediation is more efficient for low hydrocarbon concentrations and is not recommended for matrices highly contaminated with oil (>2000 mg/l). Bioremediation is also associated with performance unpredictability and may require long treatment durations.

Chemical treatment methods employ oxidants for PAH removal. Ferrarese *et al.* [13] reported 98, 96, 96, 92, 88 and 74% total PAH removal from contaminated sediments using modified Fenton's reagent, hydrogen peroxide, potassium permanganate, activated sodium persulfate + hydrogen peroxide, activated sodium persulfate and potassium

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