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Research article

Microwave (MW) remediation of hydrocarbon contaminated soil using spent graphite – An approach for waste as a resource

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ABSTRACT

In this study, we have explored the possibility of using an industrial waste for remediation of heavy fuel oil contaminated soil. Microwave (MW) heating in the presence of spent graphite (SG) from an auto forging industry has been used for the remediation. The physico-chemical characterization of SG and contaminated soil were done. Microwave remediation experiments were conducted in a lab scale unit and the effect of different parameters like microwave power, susceptor loading and treatment time were studied and optimized. The contaminated and decontaminated soils were analysed using GC-MS for total petroleum hydrocarbons (TPH), Total Organic Carbon and CHNS analyzers. Batch experiments of soil remediation showed that the TPH removal efficiencies (%) of 41.25, 87.77 and 91.18 at 300, 450 and 600 W respectively at SG concentration of 2.5 (wt. %) for a reaction time of 60 min. The addition of SG as susceptor enhanced the desorption of long chain hydrocarbons (C12–C29) present in the soil. Desorption of hydrocarbons from the soil fits well with first order kinetic model. This study successfully demonstrated the reuse of spent graphite (a lubricant waste) recovered from metal forging operations for remediating the fuel oil contaminated soil.

1. Introduction

Over the last few decades, large quantities of oil has been accidently released into the environment as a result of petroleum pipeline leaks, storage tank leaks, oil tanker collisions and other accidents (Albaigés et al., 2006; Das and Kumar, 2016; Iturbe et al., 2010). Petroleum hydrocarbon contamination poses a serious threat to the fertility of the soil causing an impact on seed germination as well as plant growth (Osuji and Nwoye, 2007; Vidonish et al., 2016; Ying et al., 2013). The petroleum contaminated soil is considered as a hazardous waste due to the presence of high concentrations of poly aromatic hydrocarbons, resins, paraffin, benzene, toluene, ethylbenzene and xylenes (BTEX) and much more leading to ecological and social catastrophes (Guo et al., 2012; Pinedo et al., 2013). Hence it is important for the contaminated sites to be remediated and the strategy of remediation to be adopted depends on the degree of contamination at the site. Recently water-based graphite particle suspensions are commonly used in forging industries, because of its advantageous physical properties (Buchner et al., 2008).

Thermal remediation is a commonly sought technology for treating petroleum contaminated soils. It mainly involves three different techniques, namely, thermal desorption, incineration and MW heating (Rushton et al., 2007). A bench scale study on thermal vapour extraction of lampblack polluted soil was reported that, majority of the PAHs had been removed at 300 °C (Harmon et al., 2001). Incineration studies attempted on TPH contaminated soils were able to witness complete remediation at a very short duration of time (Anthony and Wang, 2006; Bucalá et al., 1994). Piña et al. (2002) achieved a removal efficiency of around 98% for diesel contaminated soil at 200–900 °C operating temperature by thermal desorption. In any case, these systems have been frequently shown as incapable, costly or tedious. MW heating on the other hand, has turned out to be effective for remediating different kinds of hydrocarbon contaminated soils (Czuk, 1998; Dawei et al., 2009; Mutyala et al., 2010; Yuan et al., 2006). The great attention given to MW application in the field of environmental engineering is attributed to its potential to significantly reduce the treatment time and costs. Other advantages include homogeneous heating of substance, low energy consumption jointly with short remediation times (Jones et al., 2002). Heating due to MWs occurs in the presence of certain dielectric materials that tend to absorb and thereby convert electromagnetic energy to thermal energy (Jou, 2006). The heating of the dielectric material by MW occurs due to dipolar loss which in turn depends on its

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Table 1
Literature review of contaminated soil remediation by microwave heating.

Reference	MW frequency/power	Susceptor	Time	Contaminant/Results	Remarks
(Chien, 2012)	250 MHz/2 KW	Water	3.5 h	C_{10}–C_{40} organic compounds/75.6–98.4%	Field study, Custom built MW generator
(Robinson et al., 2014)	2.45 GHz/1-5 KW	Water	35–125s	Low and high grade oil sands (Bitumen phase)/28% @ 1 KW to 53% @ 5 KW	Field study
(Liu and Yu, 2006)	2450 MHz/700 W, 300 W	GAC	15min	PCB, 2,4,5-trichlorobiphenyl/90% @ 700 W, 70% @ 300 W	Artificially contaminated.GAC concentration was 5% by wt
(Yuan et al., 2006)	2.45 GHz/750 W	MnO_2 + aqueous (50% H_2SO_4) Fe + 0.15 mL Na_2SO_4 (5.0 M) NaOH + H_2O	10mins	Hexachlorobenzene/100% Hexachlorobenzene/93.9% C_6Cl_5SH/98.6% in presence of NaOH, 44.5% in presence of H_2O	Field soil sample
(Liu et al., 2008)	2450 MHz/750 W	GAC + Iron powder + Sodium hypophospite	10mins	Capacitor oil contaminated soil (PCB's)/95–100%	Addition of iron powder and sodium hypophospite increased the removal efficiencies.
(Cravotto et al., 2007)	2450 MHz/600 W	Sodium percarbonate	15mins	2,4-Dibromophenol/80 ± 0.2%	Artificially contaminated soil
(Falciglia et al., 2016)	2450 MHz/1000 W	moisture	60mins	PAHs/79–100%	maximum temperature reached is 208 °C
(Falciglia and Vagliasindi, 2015)	2450 MHz/1000 W	moisture	30mins	Diesel fuel/90–100%	Results were favourable for sandy soils when compared to fine textured soil
(LI et al., 2009)	2450 MHz/800 W	Carbon fiber	15mins	crude oil/99%	–
Current study	2450 MHz/800 W	SG powder	60 mins	Fuel oil contaminated soil/87–91%	Field soil sample

shape, size, dielectric constant and the properties of MW equipment used such as frequency and operating power. In the presence of an electro-magnetic field, these dielectrics experience a phase-lag, as a result of which power is interspersed in the material. Liu and Yu (2006) used Granular Activated Carbon (GAC) for absorbing MWs and were able to attain high temperatures of about 500 °C at 700 W MW power. It was also reported that the addition of carbon fibre could significantly enhance the MW heating of soil at a possibly low concentration of 0.1 wt. % and a temperature of approximately 700 °C was attained within 4 min at 800 W MW power (LI et al., 2009). Various studies on MW remediation of contaminated soil using different susceptors were given in (Table 1).

The graphite recovered from spent lithium ion batteries has been widely used to treat the water contaminated by heavy metals like lead, cadmium and metals like silver (Zhao et al., 2017). There is no literature evidence of using SG as a susceptor in facilitating the remediation of fuel oil contaminated soil by MW heating. This main objective of the study is to evaluate the performance of spent graphite (a lubricant waste from auto forging industries) as a viable susceptor for the remediation of fuel oil contaminated soil by thermal desorption, which was facilitated by MW heating.

2. Materials and methods

2.1. Soil and spent graphite samples

Soil contaminated with heavy furnace oil (HFO Grade IV) due to the oil spill at Ennore port, Chennai, India was used for the decontamination studies. The basic properties of the soil are listed in (Table 2). Different susceptors like water, pure graphite (PG) - (99% purity, Fischer chemicals), activated carbon (AC- 99% purity, Fischer Chemicals), spent graphite (SG) were used for absorbing the microwave radiation. The Spent Graphite (SG) was received from an auto forging industry near Chennai, India (Fig. 1). shows the images of heavy fuel oil contaminated soil and SG.

2.2. Physio-chemical characterization of SG and soil

The particle size distribution of the soil samples were analysed by performing sieve analysis (ASTM C136 method). The pH and EC of the soil was measured using a Hanna HI9813-6 portable meter by following the ASTM D4972-13 standard test method. Moisture content present in the graphite and soil were determined by drying 1 g of graphite at 100 °C in a hot air oven for 24 h. Iron content present in the graphite flakes were determined by spectrophotometric method at a wavelength of 570 nm (American Public Health Association (APHA), 2005). Particle size of solids in dried SG was resolved utilizing dry sieving method and Laser diffraction particle size analyser (Horiba LS 300, Japan). Dried graphite solids were suspended in distilled water and kept in an ultrasonic bath (Labman scientific instruments, India) for 10 min to accomplish a uniform circulation of the particles prior to examination. The mineralogical characterizations of the solids were performed using a Scanning electron microscopy - Energy dispersive X-ray spectroscopy (SEM-EDS), (FEI Quanta FEG 200, USA). Numerous images of the dried

Table 2
Experimental matrix for MW remediation of contaminated soil.

S.No.	Description	Range	Unit
1	TPH concentration in contaminated soil	40–110	g/kg
2	Susceptors used	Activated carbon, pure graphite, spent graphite, water	–
3	Spent graphite as susceptor	1–5	Wt.(%)
4	Microwave power	300–600	W
5	Treatment time	5–60	Mins