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Short communication

Field study of in situ remediation of petroleum hydrocarbon contaminated soil on site using microwave energy

Yi-Chi Chien*

Department of Environmental Engineering and Science, Fooyin University, Kauhsiung City 831, Taiwan

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ABSTRACT

Many laboratory-scale studies strongly suggested that remediation of petroleum hydrocarbon contaminated soil by microwave heating is very effective; however, little definitive field data existed to support the laboratory-scale observations. This study aimed to evaluate the performance of a field-scale microwave heating system to remediate petroleum hydrocarbon contaminated soil. A constant microwave power of 2 kW was installed directly in the contaminated area that applied in the decontamination process for 3.5 h without water input. The C10–C40 hydrocarbons were destroyed, desorbed or co-evaporated with moisture from soil by microwave heating. The moisture may play an important role in the absorption of microwave and in the distribution of heat. The success of this study paved the way for the second and much larger field test in the remediation of petroleum hydrocarbon contaminated soil by microwave heating in place. Implemented in its full configuration for the first time at a real site, the microwave heating has demonstrated its robustness and cost-effectiveness in cleaning up petroleum hydrocarbon contaminated soil in place. Economically, the concept of the microwave energy supply to the soil would be a network of independent antennas which powered by an individual low power microwave generator. A microwave heating system with low power generators shows very flexible, low cost and imposes no restrictions on the number and arrangement of the antennas.

1. Introduction

Due to the fast development of chemical and oil-refining industry, a large amount of chemicals or petroleum products have been released into the soil environment via spills, leakage or other incidents. Oil contaminated soil is unsuitable for agricultural, residential or recreational uses that has became a major environmental issue in many industrialized countries. Furthermore, soil pollution may cause enormous economic loss, ecological disaster and destroy agricultural production [1]. A large number of studies have been reported to remedy petroleum polluted soil by using thermal decontamination [1,2], vapor extraction [3–6], surfactants or solvent flushing, chemical oxidation, steam stripping [7], biological treatment [8,9] and others. However, these technologies may have many disadvantages such as too expensive, high energy consumption and long time frame [8]. These disadvantages make them financially impossible, especially when large areas or volumes of soil are contaminated. In addition, soil structure may be damaged in the remediation process that makes the soil unsuitable for agricultural uses.

Thermal decontamination of soil is widely used in the remediation of hydrocarbon compounds contaminated soil. It has been proven to be an effective method for the soil remediation. The thermal decontamination of soil can be notably enhanced by increasing the temperature to desorb, mobilize and evaporate pollutants, to start chemical reactions forming nonhazardous products [10]. It is well known that thermal decontamination of soil may offer many advantages, including greatest efficiency above 99% in a very short remediation time and a wide range of contaminants [11]. For example, Lee et al. proposed that the remediation of petroleum-contaminated soil in a fluidized-bed exhibited 99.9% desorption efficiency in a half hour [1].

Generally, for the saving of excavation and transport costs and avoidance of second pollutions in the transport route, a tendency has been observed to remediate soil contaminant on-site instead of off-site methods [11]. Furthermore, the in situ remediation of soil on-site is extreme preferred. Generally, on-site and in situ remediation technology may provide a more viable and economical alternative to dispose contaminated soil off-site. From the point of view of soil use and environmental protection, it is also necessary to develop an in situ technology for the remediation of contaminant soil on-site.

Microwaves are a separate band of electromagnetic radiation with frequencies in the range of 100 MHz–300 GHz that are widely

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* Tel.: +886 7 783 0542; fax: +886 7 782 1221.
E-mail address: pl036@mail.fy.edu.tw

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used in industrial drying and heating operations. The unique properties of microwave heating are rapid, uniform and selective that cannot be achieved using conventional heating methods. The idea of soil heating by microwave energy is also not new. Several lab-scale investigations on the use of microwave heating in the remediation of soils contaminated by PCBs [12], chlorinated aromatics [13], PAHs, and organic solvents [14] have been reported. The first investigation about the microwave energy used in the decontamination of soil was proposed by Windgasse and Dauerman [15]. Their experiments were conducted on a small scale and had very promising results. Abramovitch et al. have investigated the in situ decomposition of PCBs, PAHs and organic solvents using microwave energy in a modified ceramic alumina bomb. Most of the chlorinated aromatics decomposed using microwave energy and graphite fibers. In addition, no PAHs were remained in soil after microwave heating [13,14]. They also found that the most efficient decompositions are attained in the presence of Cu_2O or Al powder and 10 N NaOH [16]. Removal of polychlorinated biphenyl from contaminated soil using microwave-generated steam was performed by Di and Chang. The overall removal efficiency was greater than 98% at a steam-to-soil mass ratio of 3:1. Removal efficiency was dependent on the amount of steam added, expressed as a mass ratio of steam added to soil mass [12]. Application of activated carbon in a microwave heating to treat trichloroethylene [17], phenol [18], and BTX [19] was investigated by Jou. A microwave desorption of trichloroethylene in the reactor has also been achieved by inducing steam distillation in the granulated activated charcoal [17]. While the microwave energy is provided, organic compounds and water are desorbed from granulated activated charcoal and the arching occurs. A destruction of the organic compounds is occurred to arc between granulated activated charcoal particles by microwave heating [17–19]. Phenol [18], benzene, toluene and xylene could be treated to a non-detectable level [19]. The remediation of polychlorinated biphenyl-contaminated soil by microwave heating enhanced by activated carbon was reported by Liu and Yu. They found that rates of polychlorinated biphenyl removal are highly dependent on microwave power, soil moisture content and the amount of granular activated carbon added [20]. Yuan et al. studied the remediation of hexachlorobenzene contaminated soil by microwave energy with the assistance of MnO_2 or Fe in a sealed vial. They found that the concentration of hexachlorobenzene was reduced from 55.8 mg/kg to 0.91 mg/kg with 10 min microwave irradiation [21]. Kawala and Atamañczuk have also demonstrated the pilot-scale investigation on the remediation trichloroethylene-contaminated sand. The microwave energy was supplied intermittently, each 12 irradiation min being followed by a 3 min pause for 75 h at an identical power of 600 W. After the experiment, the concentration of trichloroethylene decreased from 5000–22,300 mg/kg to 8–29 mg/kg. They reported the presence of moisture in soil allows for a complete removal of a semi-volatile compounds at a temperature lower than 100 °C. Their results suggest the microwave heating is a very effective soil remediation process for the removal of volatile and semi-volatile hydrocarbon, and it is especially effective for the polar compounds [11]. However, to the best of our knowledge, the in situ remediation of contaminant soil on-site by microwave energy has never been investigated. In the present paper, the suitability of microwave heating for optimizing soil remediation is demonstrated at the field scale. The investigation will clarify general aspects, engineering options and conclusions for microwave heating remediation tasks.

2. Experimental

The contaminated site in this study is located at an oil-refining plant in southern Taiwan. The water level in the field was about 10 m below the surface. Due to the previous use of the site, the soil was contaminated with petroleum hydrocarbon compounds from leaking storage tanks.

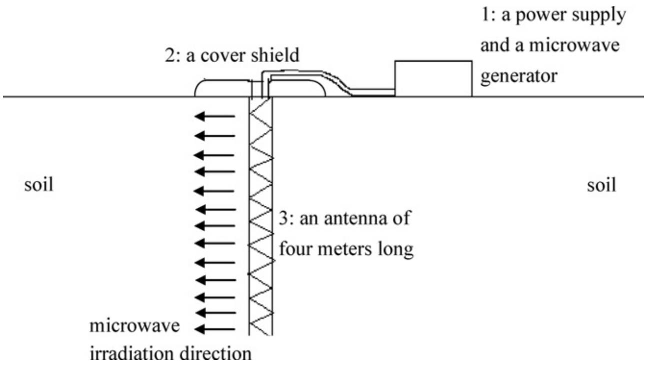

Fig. 1. The complete experimental setup of a 2 kW generator working at the frequency of 2450 MHz used in the experiment. The microwave heating system consists: (1) a power supply and a microwave generator; (2) a cover shield (a diameter of 1 m) and (3) an antenna of 4 m long.

The microwave heating system was developed as a patented process [22]. The complete experimental setup of a 2 kW generator working at the frequency of 2450 MHz used in the experiment is schematically shown in Fig. 1. The microwave heating system consists of four main elements: a power supply, a microwave generator, a cover shield with a diameter of one meter and an antenna of four meters long. The cover shield is used for the avoidance of microwave release to the atmosphere. The antenna was inserted into the contaminated soil at a depth of 4 m. The microwaves are generated by the generator, introduced via the coaxial cable and emitted through slot cut of the antenna to soil in the irradiation process for 3.5 h continuously. The soil temperatures were simultaneously recorded by two type-K thermocouples during microwave irradiation at the distances of 40 cm and 55 cm with a depth of 2 m.

Before and after the irradiation process, soil samples were collected from four depths of 0–1.2 m, 1.2–2.4 m, 2.4–3.4 m and 3.4–4.0 m below ground at different distances (40 cm, 55 cm and 100 cm) from the antenna. The samples were stored in a Teflon-sealed screw-cap vial for storage at about 0 °C to minimize loss of volatile organic compounds. All samples were analyzed in three days. In the following, all contents of hydrocarbons were related to the mass of the dry samples.

Concentrations of benzene, toluene, ethylbenzene, and xylenes were determined by gas chromatography/mass spectroscopy (GC/MS) using a method of NIEA M157.00C/M711.01C. The detection limits of benzene, toluene, ethylbenzene, and xylenes were all 0.1 mg/kg. The detection limits of C6–C9 and C10–C40 were 18.3 and 30 mg/kg, respectively. Concentrations of C6–C9 and extractable petroleum hydrocarbon (EPH) (C10–C40) were both measured by gas chromatography-flame ionization detection using a method of NIEA M157.00C/S703.61B. Hexane was used as the solvent to extract soluble organics from the soil samples. One certificate reference materials tests and one reagent blank test were analyzed for every 20 soil samples. The trip blank, and equipment blank samples were also collected and analyzed. No toluene, ethylbenzene, xylene, and TPH found in the trip blank, and equipment blank samples. A duplicate analysis was conducted to assess the reproducibility of all analysis.

3. Results and discussion

Basic properties of the test soil are listed in Tables 1 and 2. The bulk density of the soil was 1.69–2.12 g/cm^3, and the porosity was 15.35–25.92%. The porosity of tested soil was lower than

Table 1
Physicochemical and granularity properties of the test soil.

Depth (cm)	Bulk density (g/cm^3)	Particle density (g/cm^3)	Porosity (%)	Sand (%)	Clay (%)	Silt (%)
0–35	1.69	2.28	25.92	73.96	11.28	14.77
35–85	1.81	2.28	20.41	72.37	12.61	15.02
85–135	1.98	2.35	15.74	35.60	33.87	30.53
135–165	1.62	2.06	21.54	37.92	29.26	32.82
165–195	1.73	2.31	24.87	69.94	23.03	7.02
195–245	2.12	2.52	15.91	67.14	14.61	18.26
245–360	1.87	2.21	15.35	88.67	4.59	6.78

Sand (particle size: 1–0.05 mm); clay (particle size: <0.002 mm); silt (particle size: 0.05–0.002 mm).

that of typical soil porosity. It was due to the soil at the site were mixed with other soil imported from other places and compressed. Thus, the bulk density of the tested soil may be increased and the porosity of soil decreased. The moisture of the test soil sample was 7.76–22.20%. It would provide enough moisture for the formation of steam to co-evaporation of hydrocarbon contaminants in soil.

Microwave heating is a very effective soil remediation process for the removal of volatile and semi-volatile hydrocarbon, and it is especially effective for the polar compounds [11]. This method is comparably simple to realize and robust in use. A constant microwave power of 2 kW was applied in the decontamination process for 3.5 h without water input. Basically, although the larger soil contaminant concentration and heating inhomogeneities due to the larger soil volume, the principal effects observed during microwave heating process was the same in the laboratory experiments. Concentrations of benzene, toluene, ethylbenzene and xylene (BTEX), C6–C9 and C10–C40 are listed in Tables 3–8. It is apparent that the soil was strongly contaminated with C10–C40 hydrocarbons and minor contaminated with C6–C9 hydrocarbons. In addition, n.d. −0.57 mg/kg of BTEX was also determined in the soil. Most of the treated samples show significantly low concentrations of hydrocarbons than the untreated samples. A major decrease in the concentration of many hydrocarbons was observed. The removal of volatile BTEX may be due to volatilization by microwave heating. In Table 8, it is apparent that the most C10–C40 hydrocarbons were destroyed or desorbed from soil by microwave heating for 3.5 h. The highest residual concentration (28,200 mg/kg) near the bottom layer of the antenna can be explained by the highest initial concentration (21,900 mg/kg). It is believed that as the radiation time increased, the residual concentrations may be reduced. Tables 3–8 also showed the petroleum hydrocarbon in soil sample was not removed effectively in the depth of 3.4–3.6 m at 100 cm distances from the antenna. It was due to the restriction of sampling, soil was collected at different point (but very close) before and after the irradiation process, respectively. In addition, because the texture of soil was inhomogeneous, the original contaminant concentration at the two sampling points may be different. Thus, in some cases, the contaminant concentrations increased after the irradiation process. The drawback will be overcome by the microwave energy supply to the soil in a network of independent antennas or prolong the irradiation time.

On the other hand, in Tables 3–8, the remediation of contaminant may be combined with decomposition and evaporation (or co-evaporation). For the C10–C40 organic compounds, the developing steam by microwave energy caused pollutants to be co-evaporation from soil without decomposition. The moisture in damp soil is a good microwave absorber that can absorb microwave energy and the steam vaporized to catch the contaminants and remove them out of the soil. The moisture may play an important role in the absorption of microwave and in the distribution of heat. Roland et al. also reported that the formation of steam from moisture in soil being related to a volume increase by about 3 orders of magnitude, result in an additional gas stream directed outward from the soil. It is responsible for the efficient transport medium for the hydrocarbons [23]. Furthermore, the moisture content may influence the contaminant availability and soil permeability, which is the most major factor on the contaminants migration into the soil. Thus, the co-evaporation of C10–C40 with steam may be as a major mechanism in moving C10–C40 from the soil using microwave energy. Rapid expansion and evaporation of moisture in soil by microwave heating accelerated co-evaporation rates of C10–C40 molecules.

The soil temperature was about 28–30 °C that measured during the microwave heating process. This is because of soil is transparent to microwave energy. Most energy is delivered directly to hydrocarbon molecules and moisture that exhibit dielectric heating. It is benefit for saving energy in the remediation process. Dauerman et al. also reported the decontamination of 1 ton of soil by microwave heating can save 77% operation costs than that of typical incineration process [15].

On the other hand, the molecules temperature increase is due to a fast reorientation of molecular dipoles in the rapidly changing external electromagnetic field. However, much larger penetration depths in the meter-range can be realized with microwave heating, which is essential for in situ remediation procedures. With microwave heating, heat is produced within the soil volume. The attainable heating rates depend on the installed power [10]. Since the remediation of soil by microwave heating is based on the application of high-grade electrical energy, the efficiency of heat formation in the soil is a major parameter for estimating the economic aspect of this technology. The transformation of microwave energy into heat in the desired soil volume should be optimized [10]. In addition, the length of the antenna depends on the depth of the contaminated soil layer to be treated. Economically, the concept of the microwave energy supply to the soil would be a network of independent antennas which powered by an individual microwave

Table 2
Moisture content of the test soil.

Depth	Distance from the antenna					
	0.4 m (%)		0.55 m (%)		1.0 m (%)	
0.6–0.8 m	8.83[a]	8.35[b]	8.38[a]	15.67[b]	6.85[a]	7.80[b]
1.6–1.8 m	14.74[a]	14.48[b]	14.45[a]	7.76[b]	14.46[a]	14.56[b]
3.2–3.4 m	17.51[a]	15.27[b]	15.57[a]	13.78[b]	19.23[a]	22.20[b]
3.4–3.6 m	15.09[a]	14.27[b]	15.16[a]	15.46[b]	17.03[a]	17.51[b]

[a] Moisture content of the test soil before microwave irradiation.
[b] Moisture content of the test soil after microwave irradiation.

Table 3

Initial and residual concentrations of toluene in soil at different distances and depths by microwave heating.

Depth	Distance from the antenna								
	0.4 m (mg/kg)			0.55 m (mg/kg)			1.0 m (mg/kg)		
0.6–0.8 m	ND[a]	0.29[b]	–[c]	ND[a]	0.19[b]	–[c]	0.18[a]	ND[b]	100[c]
1.6–1.8 m	ND[a]	ND[b]	–[c]	ND[a]	0.29[b]	–[c]	ND[a]	ND[b]	–[c]
3.2–3.4 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	0.26[a]	0.14[b]	46.2[c]
3.4–3.6 m	0.57[a]	0.23[b]	59.6[c]	ND[a]	ND[b]	–[c]	0.29[a]	0.37[b]	–[c]

[a] Initial concentrations.

[b] Residual concentrations.

[c] Removal efficiency.

Table 4

Initial and residual concentrations of ethylbenzene in soil at different distances and depths by microwave heating.

Depth	Distance from the antenna								
	0.4 m (mg/kg)			0.55 m (mg/kg)			1.0 m (mg/kg)		
0.6–0.8 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]
1.6–1.8 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]
3.2–3.4 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	0.17[a]	ND[b]	100[c]
3.4–3.6 m	0.37[a]	ND[b]	100[c]	0.13[a]	ND[b]	100[c]	0.28[a]	0.30[b]	–[c]

[a] Initial concentrations.

[b] Residual concentrations.

[c] Removal efficiency.

Table 5

Initial and residual concentrations of *m*-xylene and *p*-xylene in soil at different distances and depths by microwave heating.

Depth	Distance from the antenna								
	0.4 m (mg/kg)			0.55 m (mg/kg)			1.0 m (mg/kg)		
0.6–0.8 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]
1.6–1.8 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]
3.2–3.4 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	0.21[a]	ND[b]	100[c]
3.4–3.6 m	0.20[a]	ND[b]	100[c]	0.14[a]	ND[b]	100[c]	0.13[a]	0.31[b]	–[c]

[a] Initial concentrations.

[b] Residual concentrations.

[c] Removal efficiency.

Table 6

Initial and residual concentrations of *o*-xylene in soil at different distances and depths by microwave heating.

Depth	Distance from the antenna								
	0.4 m (mg/kg)			0.55 m (mg/kg)			1.0 m (mg/kg)		
0.6–0.8 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]
1.6–1.8 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]
3.2–3.4 m	ND[a]	ND[b]	–[c]	ND[a]	ND[b]	–[c]	0.31[a]	ND[b]	100[c]
3.4–3.6 m	0.50[a]	ND[b]	100[c]	ND[a]	ND[b]	–[c]	0.23[a]	0.25[b]	–[c]

[a] Initial concentrations.

[b] Residual concentrations.

[c] Removal efficiency.

Table 7

Initial and residual concentrations of C6–C9 in soil at different distances and depths by microwave heating.

Depth	Distance from the antenna								
	0.4 m (mg/kg)			0.55 m (mg/kg)			1.0 m (mg/kg)		
0.6–0.8 m	ND[a]	21.9[b]	–[c]	ND[a]	47.2[b]	–[c]	ND[a]	ND[b]	–[c]
1.6–1.8 m	ND[a]	ND[b]	–[c]	ND[a]	23.9[b]	–[c]	ND[a]	ND[b]	–[c]
3.2–3.4 m	76.7[a]	19.2[b]	75.0[c]	36.2[a]	ND[b]	100[c]	1070[a]	60.5[b]	94.3[c]
3.4–3.6 m	1670[a]	63.2[b]	96.2[c]	904[a]	ND[b]	100[c]	1700[a]	1290[b]	24.1[c]

[a] Initial concentrations.

[b] Residual concentrations.

[c] Removal efficiency.

Table 8
Initial and residual concentrations of C10–C40 in soil at different distances and depths by microwave heating.

Depth	Distance from the antenna								
	0.4 m (mg/kg)			0.55 m (mg/kg)			1.0 m (mg/kg)		
0.6–0.8 m	2600[a]	4610[b]	–[c]	367[a]	2150[b]	–[c]	3520[a]	752[b]	78.6[c]
1.6–1.8 m	ND[a]	456[b]	–[c]	801[a]	9640[b]	–[c]	232[a]	ND[b]	100[c]
3.2–3.4 m	2510[a]	372[b]	85.2[c]	1660[a]	145[b]	91.3[c]	8320[a]	2030[b]	75.6[c]
3.4–3.6 m	28500[a]	3880[b]	86.4[c]	13500[a]	218[b]	98.4[c]	21900[a]	28200[b]	–[c]

[a] Initial concentrations.
[b] Residual concentrations.
[c] Removal efficiency.

generator. A custom-built high power generator (2 kW) can thus be replaced by several low-cost commercially available 750 W generators used in microwave ovens. A microwave heating system with low power generators shows very flexible and imposes no restrictions on the number and arrangement of the antennas.

4. Conclusions

The present work shows the microwave heating is a cost-effective and time-efficient technology for the remediation of soil contaminated with volatile organic compounds and petroleum hydrocarbons. The contaminant soil can be remediated by microwave heating without disturbance or excavation. The remediation is permanent and there is minimal exposure of the public and personnel to the affected contamination site. The microwave heating system is relative simple, and can be easily developed. This technique involves introducing microwave into the site to encourage volatilization of contaminants out of the soil directly. The reactor designed in this study has demonstrated its ability in effectively removal of organic compounds from contaminant soil. The results suggest the contaminated soils can be remediated safely by microwave energy even the soils are going to be used for agricultural purposes. The organic contaminants present in the soil will either be destroyed in situ or volatilized in a short remediation time without excavating soil.

Microwave heating can be considered as the flexible technique for remediation of hydrocarbon contaminated soil because different types of soil can be treated. Furthermore, the remediation process can be completed under pressure of time. The successful tests gave much insight into the engineering, physical and chemical aspects of microwave application. General conclusions on the appropriateness and competitiveness of the microwave heating as well as on preferred application fields are drawn. We believe these results encourage further research efforts to develop microwave heating system for the remediation of contaminated soil. This novel process provides a possibility for in situ remediation of hydrocarbon contaminant soil on-site in a large industrial scale.

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