

Original Research

Microwave Remediation of Diatomite-Contaminated Nitrobenzene and Soil-Contaminated Gasoline

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Received: 25 December 2018

Accepted: 26 March 2019

Abstract

Contaminated soil generally exists in industrial areas, and soil remediation is becoming more and more important. In this study, the electric field intensity and temperature distribution under different levels of microwave power were simulated. The effects of different factors on soil remediation were studied by single-factor and response surface method, and microwave radiation was compared with the remediation effect of the SVE method. The results show that the radiation time and microwave power are the main factors that affect the removal rate of pollutants. With the increase of microwave power, the electric field intensity is higher and the soil temperature increasing rate is faster. When microwave power is 700 W, radiation time is 18.48 min, moisture content is 0.78 mL/g, and the handling capacity is 20 g, the removal rate of nitrobenzene in diatomite can reach up to 84.5%. When the contaminated soils were treated for 30 minutes by microwave irradiation and SVE technology respectively, isobutylene content in the former method was only 1/5th of that in latter method. Furthermore, it is difficult to deal with a large amount of contaminated soil in a short time by utilizing SVE technology, so the microwave irradiation with characteristics of high remediation efficiency draws the attention of researchers.

Keywords: microwave, remediation, numerical simulation, response surface methodology, soil vapor extraction (SVE)

Introduction

The distribution of chemical products involves several transportation and storage modalities, during which large volumes of contaminants are released into the environment, causing hydrocarbon and benzene

series contamination of soils and groundwater – one of the most prevalent environmental concerns worldwide due to economic, environmental and human health impacts [1-3]. In the last decade, different chemical–physical and/or biological remediation technologies have been employed to remove pollutants from different soils [4]. However, these treatments may be too expensive or lengthy [5].

Recently, thermal remediation using microwave irradiation has attracted great attention in the

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environmental field representing a potential remedial alternative for contaminated soils, sludge or wastes [6]. According to the existing research results, the microwave method can greatly shorten the heating time and can selectively treat the polar pollutants, and reduce energy consumption under the condition of shortening the remediation time [7-9]. In recent years, the technology of microwave irradiation to remediating persistent organic pollutants (POPs)-contaminated soil has attracted much attention, and many scholars have studied the remediation of POPs-contaminated soils such as hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), pentachlorophenol (PCP) and polycyclic aromatic hydrocarbons (PAHs) by microwaves [10-14]. Some scholars have studied the limits of microwave (MW) heating in situ for hydrocarbon-contaminated soil remediation by modeling one-dimensional transient equations of energy [15, 16]. According to the results of numerical simulation, it has been found that the properties of the material after microwave heating depend on many factors such as geometry, size [17-20], frequency [21, 22], power input [23, 24], dielectric [25, 26] and so on. A.J. Buttress et al. used COMSOL to simulate the electric field distribution of equipment for microwave treatment of contaminated soil [27]. Some scholars at home and abroad have carried out numerical simulation research on microwave heating food, coal samples and fluid [22, 28, 29].

Due to the simultaneous thermal and non-thermal effects in the microwave radiation process, the microwave thermal effects were studied by numerical simulation in this study. At the same time, microwave remediation experiments were carried out on nitrobenzene and gasoline-contaminated soil, and the effects of microwave power, irradiation time, soil moisture and pH on soil remediation were studied. The response surface analysis method was used to obtain the most significant influencing factors and experimental optimal conditions. The results of the microwave and SVE techniques on the remediation of gasoline-contaminated soils were analyzed and compared. The advantages and disadvantages of the two methods were compared, which provided a theoretical basis for the selection of practical methods.

Materials and Methods

Model Development

Governing Equations

First and foremost, simulations were carried out using multiphysics software to verify the distribution of the electric field generated inside the oven cavity due to microwave incidence and the temperature distribution of the soil in the beaker after it has been heated.

Maxwell's equations are used to solve electromagnetic propagation in the simulation. The

governing equation of the electric field wave is given by:

$$\nabla \times (\mu_r^{-1} \nabla \times \vec{E}) - k_0^2 \left(\epsilon_r - \frac{j\sigma}{\omega \epsilon_0} \right) \vec{E} = 0 \quad (1)$$

...where ω is the angular frequency, ϵ_0 is the permittivity of vacuum (8.85×10^{-12} F/m), μ_r is the relative permeability, ϵ_r is the relative permittivity, k_0 is the wave number in free space and σ is the electrical conductivity.

The microwave power density, P (W/m^3) that is absorbed by a material can be expressed by:

$$P = \left[(\sigma + \omega \epsilon'') E^2 + \omega \mu'' H^2 \right] \quad (2)$$

...where σ , ϵ'' , ω , and μ'' are electrical conductivity, dielectric loss factor, angular velocity, and magnetic permeability, respectively. For non-magnetic materials, the magnetic part is assumed to be zero ($H = 0$).

Electric permittivity is described as a complex quantity with both real and imaginary parts, which is given by the expression:

$$\epsilon^* = \epsilon' - j\epsilon'' \quad (3)$$

...where ϵ^* is the complex permittivity (F/m), ϵ' is the real permittivity (10F/m), and ϵ'' is the imaginary permittivity (1.5F/m) [30].

Geometric Model

In this work, a 3-D finite element model (FEM) was developed to simulate the heating of soil in the ultrasonic microwave combined reaction system with a frequency of 2.45 GHz (Fig. 1). The dimensions of the microwave cavity, waveguide, beaker and soil sample

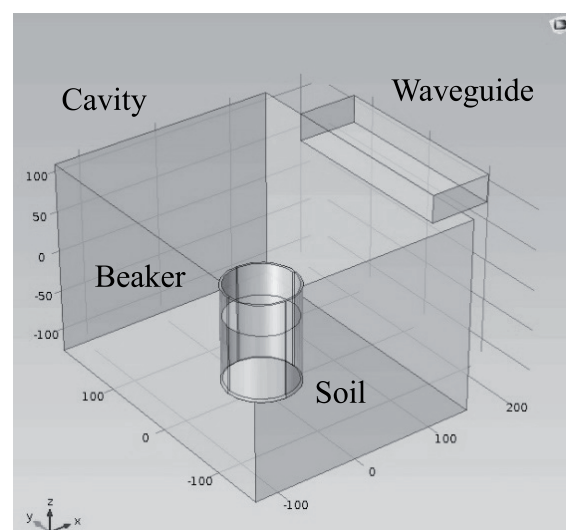


Fig. 1. Microwave cavity geometry model.

Table 1. Dimensions of microwave cavity, waveguide, beaker and soil sample.

Dimensions	Height (mm)	Length (mm)	Width (mm)	Radius (mm)
Microwave cavity	235	360	280	-
Waveguide	34	230	75	-
Beaker	122	-	-	45
Soil sample	80	-	-	42.5

were measured as shown in Table 1. The soil sample is surrounded by a medium with zero dielectric losses (air).

Physically controlled mesh was selected for meshing the whole geometry. Element size was set to finer. In order to get an accurate result, the maximum element size was refined to $1/5^{\text{th}}$ of the microwave wavelength. The total number of element was 165626 and the minimum element quality was 0.07514. The quality evaluation of mesh element is shown in Fig. 2, the results of the calculation are reliable when the value of mesh quality is greater than 0.6. In this simulation, 94% mesh elements meet this requirement. This means the quality of the mesh elements can acquire accurate results.

Model Assumptions

In order to save computational time and obtain reliable calculation results, the following assumptions are made for the model:

- The soil sample is homogeneous and isotropic.
- Variation in volume, physical, and chemical properties are not considered.

- The chemical reaction between the air and soil sample is negligible.

The impedance boundary condition was used to define the walls of waveguide and cavity. In order to obtain a temperature distribution after the soil has been heated, only the heat conduction in the soil sample is considered.

Experimental Methodology

Materials and Equipment

The experiment materials were dried soil, diatomite, methanol, nitrobenzene, sodium hydroxide, and concentrated sulfuric acid. The instruments used in the experiment include moisture analyzer (HC103), gas detector (PGM7340VOC), SVE reactor, ultrasonic microwave combined reaction system (XO-SM50), thermo fourier transform infrared spectrometer, ultrasonic oscillator (TSX-100), desktop high speed centrifuge (TG16-WS), and ultraviolet spectrophotometer (UV-2600).

Preparing Contaminated Soil

Nitrobenzene was diluted with methanol to obtain 200 ml of a nitrobenzene solution having a concentration of 1000 mg/L, which was poured into 200 g of diatomite and stirred well. After methanol is completely volatilized (naturally air-dried for 24 h), nitrobenzene-contaminated soil is obtained. 6 mL of gasoline was added to 120 mL of natural soil and uniformly mixed, and each of four beakers was poured into 20 g of soil as a sample to be treated.

Microwave Remediation of Nitrobenzene-Contaminated Soil Experiment

40 g of nitrobenzene-contaminated soil samples were taken and divided into two equal parts. Two samples were subjected to conventional heat recovery and microwave irradiation, respectively, and the concentration of nitrobenzene in the soil was measured at 70°C, 110°C, and 150°C.

Take 100 g of contaminated soil evenly divided into 5 parts for single factor experiments. Tests were performed under different conditions, with different radiation times (5 min, 10 min, 15 min, 20 min, and 25 min) and different microwave powers (200 W, 300 W, 450 W, 600 W, and 750 W), different moisture content (add 0 mL, 0.5 mL, 1.5 mL, 2.5 mL, 5 mL of deionized water respectively), and different pH (4 samples were added dilute sulfuric acid 2 mL, 4 mL, sodium hydroxide solution 2 mL, 4 mL). Weighed five contaminated soil samples (3-25 g) of varying quality and perform single factor experiments on different handling capacity under the same conditions. 0.5 g microwave-treated soil was subjected to ultrasonic extraction (30 min), followed by centrifugation for 10 minutes (5000 n/min), and

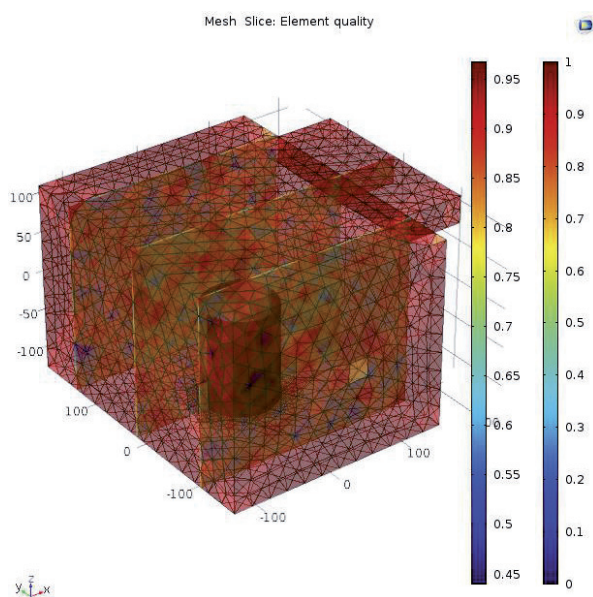


Fig. 2. Quality evaluation of mesh element.

Table 2. Factor and level of experiment by response surface methodology.

Independent variable	Value level		
	-1	0	1
Microwave power(W)	200	450	700
Radiation time(min)	5	12.5	20
Moisture content(mL/g)	0.2	0.6	1
Handling capacity(g)	5	12.5	20

the supernatant was taken up to 50 ml with methanol, and the absorbance was measured using an ultraviolet spectrophotometer that saved the results.

We took the microwave power, radiation time, moisture content, and handling capacity as optimization factors. Factor level values were determined according to the optimal range determined by single-factor experiments; the experiment design scheme is shown in Table 2.

Microwave Treatment and SVE Comparison Experiment

Three gasoline-contaminated soil samples were treated with 700 W of microwaves for 5 min, 15 min, and 30 min, respectively; another sample was subjected to SVE heat recovery. In the experiment, nitrogen was used to purge the gasoline in the soil into the buffer bottle for testing. The content of isobutylene in the soil samples of the two remediation technologies at different treatment times was recorded. A gas detector was used to measure the content of isobutylene in the treated soil and its infrared spectrum was obtained.

Results and Discussion

Numerical Simulations

Electric Field Distribution

By simulating the electric field distribution under different input power (200 W, 300 W, 400 W, 500 W, 600 W, 700 W), we found that the electric field distribution was the same when the input power was different. The electric field distribution is shown in Fig. 3(a-b). Fig. 3(c) shows that the peak value of the electric field intensity increased as the power increased.

The maximum value of the electric field strength appears at the bottom of the cavity due to the reflection of electromagnetic waves on the surface of the metal wall. The electric field is symmetrical about the XOZ plane because we believe that the physical properties of the air are uniform during the simulation, so the propagation of the electromagnetic field is not disturbed. In the XOZ plane, the electric field strength is distributed

in a zigzag pattern, because the soil sample placed in the cavity has a certain absorption and reflection effect on the electromagnetic field.

Temperature Distribution

By comparing Fig. 3b) and Fig. 4, it can be seen that the electric field intensity was consistent with the temperature distribution. In soil samples, the temperature was high in places where the electric field was strong. This is the "hot spot" effect in the microwave thermal effect. When the input power is 200W, the maximum temperature after 120s is 335.65K; when the power is increased to 700W, the maximum temperature is 469.93K. During the same microwave irradiation time, an increase in microwave power will result in an increase in the maximum temperature of the soil sample, indicating that the microwave heating efficiency increases with power. In the simulation results, the temperature distribution law does not change when the power increases, but the temperature difference in the soil sample increases, that is, the increase of power exacerbates the "hot spot" effect. This makes the excessive power not necessarily get better results in practical engineering applications.

Microwave Remediation of Nitrobenzene-Contaminated Soil Experiment

Single-Factor Experiment

As shown in Fig. 5a), both the microwave irradiation and the conventional heating remediation pollutant removal effect are greatly improved with increasing temperature. At the same temperature, the effect of microwave irradiation is obviously better than that of conventional remediation, and with the increase of temperature, the advantages are further strengthened. It can be proved that the microwave irradiation process not only achieves the purpose of removing pollutants by heating the soil, but also changes the internal structure of the soil or pollutants to achieve the purpose of remediation.

As can be seen from Fig. 5b), in the case of a microwave power of 500 W, the remediation efficiency of the soil in 15 min continues to increase. According to the simulation results, the soil temperature rises rapidly when the microwave power is 500 W. There was no significant change in the effect of soil remediation between 15 min and 25 min, and there was even a tendency to decrease. This is due to the temperature of soil increasing over time, when the nitrobenzene is degraded but the intermediate product remained in the soil. Therefore, taking into account the economic benefits and remediation effects, the best microwave irradiation time is 15 minutes.

With the increase of microwave power, the rate of change of nitrobenzene removal rate in soil gradually decreased. As with the simulation result, as the

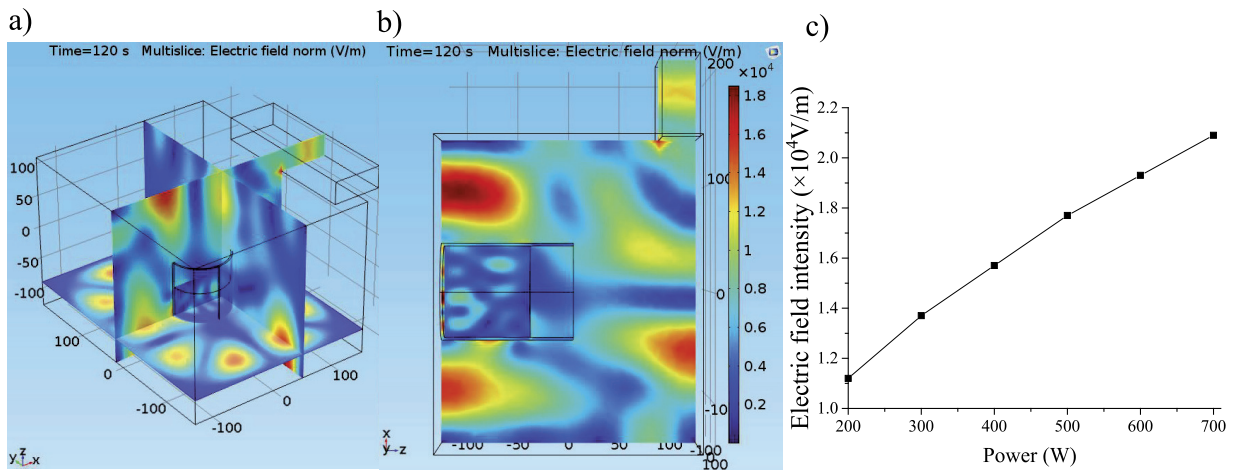


Fig. 3. a), b) Electric field distribution and c) maximum electric field strength at different powers.

microwave power increased, the temperature of the soil increased rapidly and the nitrobenzene evaporated. When the power was low, only some regions of the soil sample rapidly increased in temperature, and when the power was high, the overall temperature of the soil sample increased, which increased the removal rate of nitrobenzene. According to the experimental results, 700 W was selected as the optimal treatment condition.

When the added deionized water was greater than 5 ml, the microwave remediation efficiency began to decrease. The moisture content has a great influence on the dielectric properties of the soil. When the moisture content was low, water absorbed microwaves to warm up and evaporated. Nitrobenzene can evaporate with the volatilization of water vapor. When the moisture content is high, the evaporation of water requires a large amount of heat to be consumed. Without sufficient

energy support, the removal rate of nitrobenzene decreases. Therefore, in the microwave remediation of nitrobenzene-contaminated soil, not only to consider

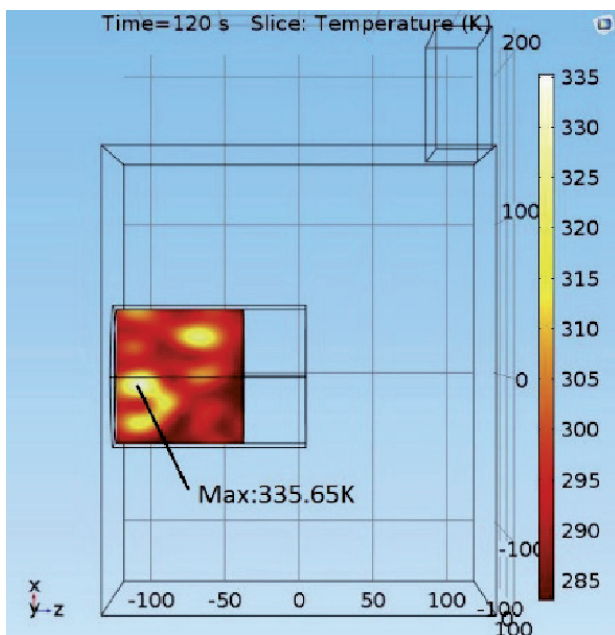


Fig. 4. Temperature distribution at 200 W microwave power.

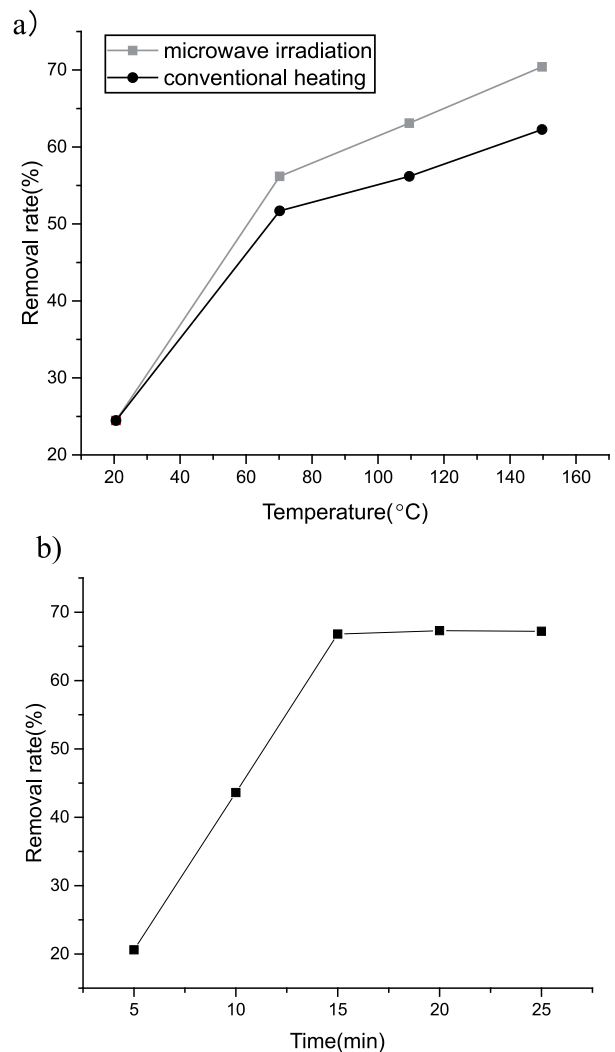


Fig. 5. Nitrobenzene removal rate with a) different temperatures and b) treatment times.

Table 3. Response surface methodology results.

Experiment number	Microwave power (W)	Radiation time (min)	Moisture content (mL/g)	Handling capacity (g)	Removal rate (%)
1	450	5.0	0.2	20.0	46.31
2	700	12.5	0.6	5.0	73.92
3	200	12.5	1.0	12.5	28.25
4	450	5.0	1.0	12.5	51.28
5	450	5.0	0.6	20.0	54.79
6	200	12.5	0.2	12.5	32.65
7	450	12.5	0.6	12.5	62.98
8	700	12.5	1.0	12.5	74.77
9	200	12.5	0.6	20.0	37.46
10	200	20.0	0.6	12.5	37.62
11	700	20.0	0.6	12.5	79.15
12	450	20.0	0.6	20.0	65.48
13	450	12.5	0.6	12.5	62.20
14	450	20.0	0.6	5.0	63.14
15	450	12.5	0.2	20.0	55.34
16	450	12.5	1.0	20.0	57.19
17	200	12.5	0.6	5.0	32.97
18	200	20.0	1.0	12.5	66.13
19	450	12.5	0.6	12.5	63.02
20	450	12.5	0.2	5.0	56.26
21	700	12.5	0.6	20.0	84.86
22	450	5.0	0.6	5.0	51.66
23	450	20.0	0.2	12.5	57.17
24	450	12.5	1.0	5.0	52.35
25	700	5.0	0.6	12.5	69.73
26	200	5.0	0.6	12.5	24.38
27	700	12.5	0.2	12.5	72.49

the role of water absorption of microwaves, but also consider the heat consumed by water evaporation factors. Therefore, about 0.6 mL/g of moisture content was the most suitable.

We added a small amount of acid or alkali in the soil to improve microwave remediation. This showed that the addition of strong acid and alkali can effectively destroy the components of nitrobenzene in the soil and the nature of the soil under the irradiation of microwaves, which had a significant effect on the removal of nitrobenzene.

Under conditions where the soil treatment volume was less than 20 g, the removal rate of nitrobenzene increased significantly with soil volume. The higher the soil bed height, the better the microwave energy

transfer efficiency. The soil can absorb more microwave energy and reach a higher temperature, thereby increasing the removal rate of nitrobenzene. However, when the amount of soil treatment is too large, making it difficult for the internal nitrobenzene to evaporate, the rate becomes smaller. It can be seen that this experiment should control the amount of soil treatment within 20 g.

Response Surface Optimization

The response surface methodology was designed based on the previous experimental results and methods (the experiment design and results are shown in Table 3). The analysis of model variance and the

Table 4. Variance analysis results.

Source	Sum of squares	d _f	Mean square	F value	P-value Prob(P)>F
Model	59.37-66.10	1.54	6476.74	64.62	<0.0001
A- Microwave power	20.12-13.48	0.77	5702.44	796.59	<0.0001
B- Radiation time	4.20-7.56	0.77	414.66	57.92	<0.0001
C- Moisture content	-0.87-2.50	0.77	7.92	1.11	0.3135
D- Handling capacity	0.39-3.75	0.77	51.34	7.17	0.0201
AB	-3.87-1.96	1.34	3.65	0.51	0.4890
AC	-1.24-4.58	1.34	11.16	1.56	0.2357
AD	-1.30-4.53	1.34	10.40	1.45	0.2513
BC	-1.92-3.91	1.34	3.98	0.56	0.4702
BD	-3.11-2.72	1.34	0.16	0.022	0.8851
CD	-1.47-4.35	1.34	8.29	1.16	0.3029
A ²	-8.08-3.03	1.16	164.77	23.02	0.0004
B ²	-5.76-0.71	1.16	55.80	7.79	0.0163
C ²	-7.84-2.79	1.16	150.71	21.05	0.0006
D ²	-3.44-1.61	1.16	4.44	0.62	0.4464

significance test are important ways to measure the rationality and predictability of model design. The variance analysis and significance test of the model regression equation were performed, the results are shown in Table 4.

The effect of the model fitting experimental data is significant, and the missing error is not significant. The F value of the model is 64.62. The model shows a significant effect, and the value of the unrealized item F is 39.99, indicating that the probability of the missed model is only 2.46% (the general requirement is less than 5%) and the missing model term is not significant. This shows that the model has a good degree of fit and the experiment error is small.

In the reaction surface of the removal rate, the four single-factor reactions were significant, and the interaction between microwave power and moisture content, microwave power and handling capacity was significant. The order of influence is: microwave power>radiation time>handling capacity>moisture content. In the second term, only the handling capacity is not significant, and the other two items are very significant. The quasi-elimination term was not significant, indicating that the linear effect of other influencing factors on the removal rate of nitrobenzene in the experimental reaction was not significant, and neglecting the influence of other factors did not affect the accuracy of the experiment.

The effect of each factor and its interaction on the removal rate of nitrobenzene was evaluated based on the response surface of the quadratic fit model and contour lines. The results were shown in Fig. 6. Fig. 6a) shows

the significant interactive effects of microwave power and soil moisture on nitrobenzene removal. It can be seen that the microwave power and the removal rate of the edge surface are relatively steep, indicating that it has a significant influence on the removal rate. With

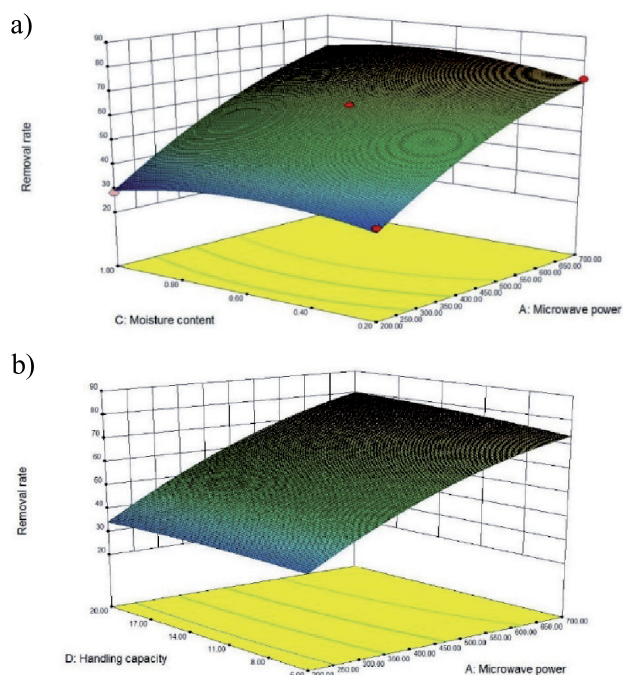


Fig. 6. a) Response surface of microwave power and moisture content and b) response surface of microwave power and handling capacity.

the increase of microwave power, the removal rate of nitrobenzene removal increased significantly. Water acts as a good absorbing medium, and it can help the microwave absorption when the moisture content is constant. However, when the moisture content is too high, a large amount of microwave energy is used for the evaporation and absorption of moisture, thereby reducing the treatment efficiency. Fig. 6b) shows the significant interactive effect of microwave power and throughput on nitrobenzene removal when the radiation time is 12.5 minutes and the moisture content is 0.6 mL/g. The interaction between the moisture content and the radiation time is close to significant. Under the conditions of a certain time, the removal rate of nitrobenzene increases and then decreases with the increase of soil moisture content, but when the radiation time is higher, the inflection point at which the removal rate begins to decline obviously moves backward. Therefore, the longer the radiation time, the better. If the radiation time is too long, the soil temperature will be too high, thus limiting the removal of nitrobenzene.

By multiple regression of the removal rate of nitrobenzene in 27 groups of experiments, a quadratic multinomial regression equation for the removal rate for four variables was finally obtained:

$$Y = -20.43 + 0.15A + 2.29B + 24.23C + 0.05D - 0.04AB + 0.02AC - 0.04AD + 0.33BC - 0.03BD + 0.48CD - 0.05A^2 - 0.06B^2 - 33.22C^2 - 0.02D^2 \quad (4)$$

...where Y is the nitrobenzene removal rate, %; A is microwave power, W; B is the radiation time, min; C is the soil moisture content, mL/g; and D is the handling capacity, g.

The optimized processing conditions were microwave power 700 W, radiation time 18.48 min, soil moisture content 0.78 mL/g, and soil handling capacity 20 g. At this point, the nitrobenzene removal rate will reach 84.5%. After the correction, the actual remediation experiment was performed. There was no significant difference between the nitrobenzene removal rate and the model predictions. It can be seen that the model better reflects the relationships among microwave power, radiation time, soil moisture content, and treatment volume in the restoration of nitrobenzene-contaminated soil. When the working conditions of these factors are known, the remediation effect can be predicted by the regression equation of the model. This study provides a certain theoretical basis for the practical application of engineering.

Experiment Analysis of Microwave Processing and SVE Comparison

The experimental results are shown in Table 5. Because of the low temperature (20°C) and low pumping rate in the heat-strengthening SVE technology, the remediation rate was slow, and the isobutylene content showed a steady decline during the remediation process. Under the influence of higher microwave power, the gasoline in the soil in the first 10 minutes of treatment was quickly dissolved. Compared with the SVE technology, the process of pumping to generate a vacuum first leads to slow progress in the early stage of processing. Because microwave technology directly acts on the soil, the microwave energy rapidly destroys the molecular composition of the gasoline and shortens the remediation period. The microwave allows the soil to heat up to more than 100°C in a relatively short period of time. Therefore, compared with gas phase extraction, microwave technology has a faster and deeper remediation rate, but the energy loss is higher.

Fourier-infrared spectroscopy scans of contaminated soils with microwaves were performed at different times. Representative spectra are shown in Fig. 7a). It can be found that all the oil soils have strong absorption at wavenumbers of 1383 cm⁻¹, 1542 cm⁻¹, and 2368 cm⁻¹, and there are multiple peaks near 3700-3800 cm⁻¹. There is a broad absorption peak around wave number 3200, which is mainly attributed to unsaturated carbon, namely C-H or O-H, N-H stretching vibration absorption. Because the 1380 cm⁻¹ peak is sensitive to the structure, it is useful for identifying the methyl group. Therefore, the degradation of gasoline can be judged by the intensity change of the absorption peak at 1380 cm⁻¹ in the infrared spectrum. As the microwave time increases, the peak area at about 1383 cm⁻¹ decreases, indicating that the CH surface bending vibration of the alkane-based tert-butyl group is reduced. The decrease in the peak area near 1540 cm⁻¹ indicates that the asymmetric bending vibration in the methyl CH plane of the alkane is reduced. It can be seen that the content of alkane light components in soil samples has declined.

The infrared spectra of the two methods were compared (Fig. 7b). We found that most of the absorption peaks in the soil after microwave treatment and SVE treatment were the same. However, the wave numbers and intensities of most peaks have changed. Some of the chemical components and functional groups in the pollutant gasoline after microwave treatment have changed, especially in the 3500-4000 cm⁻¹ area where the original chemical

Table 5. Gasoline contaminated soil remediation results.

Isobutylene content (mg/m ³)	5 min	10 min	15 min	20 min	30 min
SVE (80 m ³ /h)	25122	17300	12690	10010	5883
Microwave (700 W)	12217	5196	2048	1477	1021

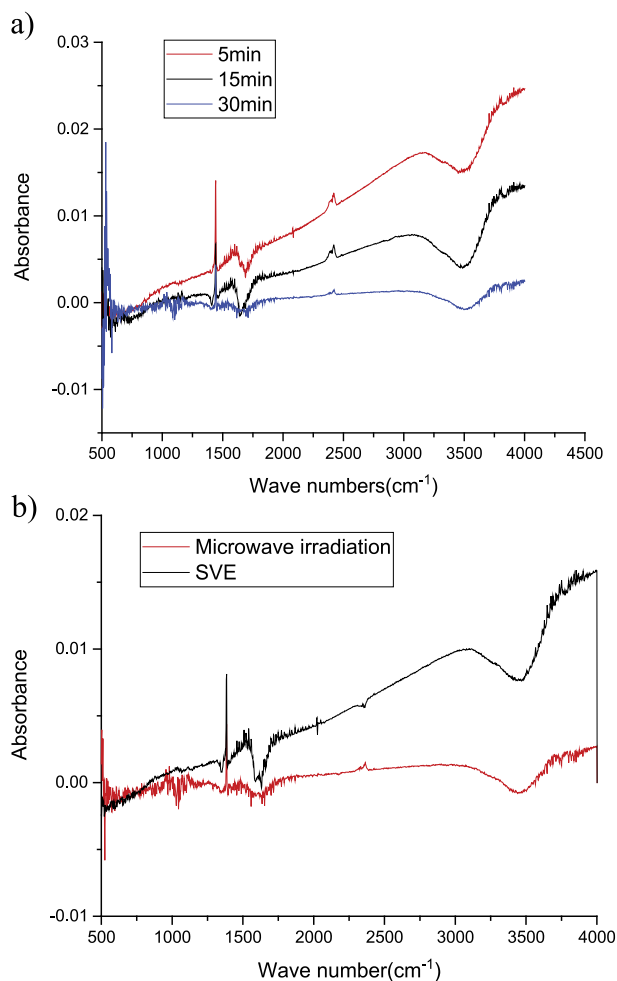


Fig. 7. Comparison of infrared spectra a) different microwave times and b) thermally enhanced SVE and microwave remediation.

components were largely degraded. The infrared spectrum of the soil after microwave remediation did not produce new peaks compared with that before remediation. It was proved that the soil remedied by microwaves did not produce new pollutants, and the original pollutants were greatly degraded. At the same treatment time, the effect of microwave irradiation was better than SVE. Therefore, the microwave irradiation method is more rapid and convenient, and the remediation efficiency is high.

Conclusions

In this study, through numerical simulation, comparison of experiments, single factor experiments, and response surface methodology, the following conclusions were obtained: Thermal effects are an important aspect of microwave remediation. When the input power increases, the electric field strength and temperature maximum increase, but the distribution of the two does not change. The handling capacity, radiation

time, moisture content and microwave power are all factors that can affect soil remediation, the order of influence is: microwave power>radiation time>handling capacity>moisture content. The regression equation obtained by the response surface method is well fitted, and can be used to predict the remediation effect that can be achieved under known working conditions. After the contaminated soil was also treated for 30 minutes, the gasoline content in the soil after microwave irradiation was about 20% after the SVE treatment. Conventional heating, microwave irradiation and SVE can reduce the concentration of pollutants in the soil, and the treatment effect is sorted: microwave>SVE>conventional heating. SVE disadvantage is that the treatment process is too long and it is difficult to deal with a large amount of contaminated soil in a short time so the microwave irradiation remediation efficiency is higher.

Acknowledgements

This work was funded by the Academic Human Resources Development in Institutions of Higher Learning under the Jurisdiction of Beijing Municipality (PHR201107213). The authors are indebted to the anonymous reviewers for their valuable comments.

Conflict of Interest

The authors declare no conflict of interest.

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