



Electrokinetics-enhanced biodegradation of heavy polycyclic aromatic hydrocarbons in soil around iron and steel industries

Fengmei Li^a, Shuhai Guo^{a,*}, Niels Hartog^b

^a Institute of Applied Ecology, Chinese Academy of Sciences, Shenyang 110016, China

^b KWR Watercycle Research Institute, 3433 PE Nieuwegein, The Netherlands

ARTICLE INFO

Article history:

Received 19 April 2012

Received in revised form 9 August 2012

Accepted 15 August 2012

Available online xxx

Keywords:

Electrokinetics

Heavy polycyclic aromatic hydrocarbons

Biodegradation

ABSTRACT

Bioremediation is a safe and cost-effective technology for the removal of polycyclic aromatic hydrocarbons (PAHs) contaminated soils, but its remediation rate is usually very slow at soils contaminated with heavy PAHs in high concentrations. This paper describes the feasibility of using electrokinetics to enhance the degradation of heavy PAHs in soil around iron and steel industries. Three bench-scale experiments were conducted for 90 days using historically polluted soil with a total PAHs content of 220.01 mg/kg dry soil. All of the experiments were inoculated with PAHs degrading bacteria, but experiments II and III were performed using constant polarity and alternating polarity electrokinetic conditions, respectively. Results were compared with those from the control experiment (experiment I), which did not receive any electrokinetic treatment. The results demonstrated that the electrokinetic process could enhance the biodegradation extent of total PAHs and heavy PAHs in the soil. The final degradation extents of total PAHs were 9.5% and 13.5% higher in experiments II and III, respectively, as compared to experiment I. Under the electrokinetic and bacteria conditions, the relative enhancement in the degradation of four- to six-ring PAHs compared to the control experiment was much stronger and increased with increasing ring number. The final degradation extents of four- to six-ring PAHs increased by 7.9–8.6%, 11.0–18.4% and 17.2–25.6% in experiments II and III compared to experiment I, respectively. The results also showed that the electrokinetic operation mode could affect not only the degradation extent of total PAHs but also bacterial counts and soil moisture of different regions in soils. The use of alternating polarity electrokinetics was favorable to the bacterial growth and kept the soil properties uniform. In addition, there was a positive correlation between the degradation extent of PAHs, bacterial counts and moisture content by Pearson correlation analysis under electrokinetics. The results of this work demonstrate that the use of electrokinetics can significantly enhance the degradation of PAHs by influencing soil conditions. Therefore, the use of electrokinetic technology may provide a useful tool for enhancing the bioremediation of heavy PAHs in soil.

© 2012 Elsevier Ltd. All rights reserved.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of potentially carcinogenic organic substances composed of more than two fused benzene rings [1,2]. Natural and anthropogenic processes generate PAHs during the incomplete combustion of all types of organic matter [3]. Coal- and coke-burning are the major sources of PAHs in the environment, causing soils around iron and steel industries to become heavily contaminated with PAHs [4], typically with high concentrations of heavy PAHs (>3 aromatic rings). Low molecular weight PAHs, with two or three fused aromatic rings, have a relatively higher water solubility and thus degrade more easily, while high molecular weight PAHs, with four to six fused

aromatic rings, are quite hydrophobic and more difficult to break down [5]. As heavy PAHs are more recalcitrant and may persist in the environment for a long time, the remediation of soils around iron and steel industries poses a great challenge. Bioremediation is typically regarded as an attractive technology for removing PAHs from soils because it is cost-effective and environmentally acceptable [6]. However, many factors hinder the biodegradation of PAHs in soils, such as insufficient bioavailability of the contaminant and nutrient [7]. In recent years there has been increasing interest in employing electrokinetic remediation to overcome the problems associated with bioremediation [8–11].

Electrokinetic remediation is an in situ technology that consists of the controlled application of a low intensity direct electric current through the contaminated soil between appropriately distributed electrodes. The technique, which relies on three process – electromigration (movement of charges), electroosmosis (water), and electrophoresis (charged particles) – has been

* Corresponding author. Tel.: +86 024 83970449; fax: +86 024 83970448.
E-mail addresses: shuhaiguo@yahoo.com, fmli2010@hotmail.com (S. Guo).

successfully applied to remove heavy metals from the soil [12–14]. In recent years, there has been increasing interest in the treatment of soil contaminated with organic pollutants. Several studies have indeed demonstrated improved removal of organic pollutants such as gasoline hydrocarbons, aromatic compounds, herbicides or trichloroethylene in electric fields applied to soil [15–17]. During the electrokinetic treatment of a soil, nutrients and microorganisms are spread throughout the soil [18,19] and the organic pollutants can be transported to the area harboring microbial populations able to degrade the pollutants [20–22]. The electric current also induces redox reactions on the electrode surfaces, resulting in the destruction of organic compounds [23,24]. In addition, the electrical current can cause heating of the soil, which can be beneficial for contaminant remediation in cold climate areas [10]. Biodegradation may also be affected indirectly by the alteration of soil properties such as pH and moisture content caused by the applied electrical field [25]. So, several mechanisms contribute to the potential of electrokinetics to enhance the bioremediation of organic contaminants. While some researchers have successfully applied electrokinetics to enhance the biodegradation of various organic compounds [21,25–27], studies on PAHs have mainly focused on the degradation of individual PAHs or light PAHs [28–30]. Furthermore, these studies have mainly been based on experiments using artificially contaminated soils [31]; mixed and heavy PAHs in naturally contaminated soils have received less attention.

The main objectives of this study were to experimentally investigate the enhancement effect of electrokinetics, in particular the differences under conditions of constant and alternating polarity electrical fields, on the biodegradation of total PAHs and heavy PAHs in contaminated soils and to determine the possible mechanism(s) involved. We used historically polluted soil around Benxi Iron and Steel Group Corporation in which heavy PAHs make up the majority of the total PAHs contamination. Benxi is an old industrial city in northeastern China that mainly produces iron and steel. The city has a serious PAH pollution problem because of an inadequate industrial structure, as well as special climatic, geographical and environmental conditions [4]. The outcomes of this study are aimed at providing essential reference information for future remediation efforts of heavy PAHs in soils surrounding iron and steel industries.

2. Experimental

2.1. Soil

The soil used in this study was a sandy loam, obtained from the topsoil layer (0–10 cm) of woodland around Benxi Iron and Steel Group Corporation, China. In order to keep the test consistent, the sample was prepared by air drying for 2 weeks, and sieved (2 mm mesh). The resulting soil sample was thoroughly mixed and its initial characteristics were analyzed according to standard methods for soil analysis [32,33]. The characteristics were as follows: 20% moisture; pH 8.36; 1.77% organic matter; 0.073% total nitrogen; 0.046% total phosphorus; a particle size distribution of 64.9% sand, 21.9% fine, 13.3% clay; and a total PAH concentration of 220.01 mg/kg dry soil (sum of 11 EPA PAHs). The concentration and composition of individual PAHs are listed in Table 1. Heavy (four- to six-ring) PAHs accounted for 92.19% of total PAHs in the study. The light PAHs phenanthrene accounted for the remaining 7.81%.

2.2. Bacteria

The PAHs-degrading bacteria were isolated from contaminated soil samples around Benxi Iron and Steel Group Corporation. PAHs were also extracted from this soil, which were then

Table 1
Concentration and composition of individual PAHs.

Polycyclic aromatic hydrocarbons	Rings	Content (mg/kg)	Percentage (%)
Phenanthrene (Phe)	3	17.19 ± 0.034	7.81
Fluoranthene (Flu)	4	36.49 ± 0.225	16.59
Pyrene (Pyr)	4	29.50 ± 0.541	13.41
Benzo(a)anthracene (BaA)	4	9.41 ± 0.336	4.28
Chrysene (Chr)	4	18.93 ± 0.185	8.61
Benzo(b)fluoranthene (BbF)	5	29.52 ± 0.346	13.42
Benzo(k)fluoranthene (BkF)	5	11.91 ± 0.432	5.41
Benzo(a)pyrene (BaP)	5	21.92 ± 0.234	9.96
Indeno(1,2,3-cd)pyrene (IcdP)	5	3.00 ± 0.196	1.36
Dibenzo(a,h)anthracene (DahA)	6	21.66 ± 0.098	9.85
Benzo(g,h,i)perylene (BghiP)	6	20.47 ± 0.076	9.30

added to the mineral media to provide the sole carbon source (500 mg/L). The mineral media contained KH_2PO_4 (0.9 g/L), K_2HPO_4 (0.1 g/L), NH_4NO_3 (0.1 g/L), $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ (0.1 g/L), CaCl_2 (0.08 g/L), $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (0.01 g/L), and 1 mL/L of microelements stock to solution with a pH value of 7.0 [23]. All the chemicals used were of analytical grade. The bacterial cells were grown in the mineral media on a shaker at 28 °C and 160 rpm and collected by centrifugation when they were in the exponential growth phase. After washing twice with sterilized water, they were resuspended in the mineral media to obtain a highly concentrated bacterial suspension for the experiments.

2.3. Experimental setup

Fig. 1 shows a schematic diagram of the experimental test setup used. It consisted of a soil cell, two pairs of electrodes, an electrode control system and a power supply. The soil cell was made of Perspex with inner dimensions of 24 cm length, 12 cm width and 10 cm height. Column-shaped stainless steel electrodes with a length of 12 cm and a diameter of 0.5 cm were used to generate the electric field. The lateral distance between the two electrode pairs was 6 cm. The electrode autocontrol system (Siemens) was capable of alternating the polarity of the electric field, thus allowing testing of different operation modes during the experiments. The power supply provided a constant direct current (DC) electrical potential difference of 24 V for the experiments (1 V/cm). During operation the experimental set-up was covered by a lid to prevent evaporation.

2.4. Testing procedure

The bacterial suspension was mixed directly into the soil while preparing the soil specimen at an initial inoculation in the order of 5.21×10^7 cfu (colony-forming units)/g dry soil. The soil was then rehydrated to moisture content about 20% (w/w) with sterilized mineral media. The wet soil was tamped into the soil cell in layers with a pressure of 0.1 kg/cm² to a height of 8 cm [34] (Fig. 1). The

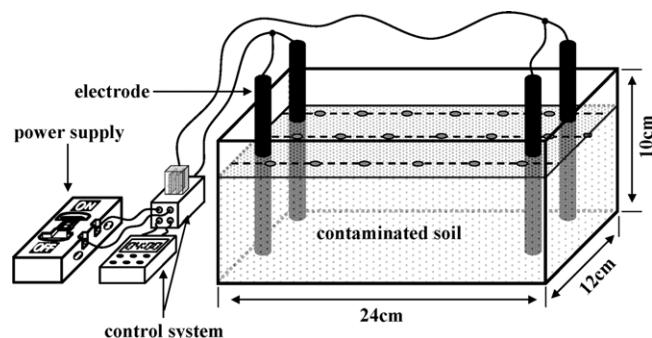


Fig. 1. Schematic of the experimental setup (●, sampling location).

electrodes were immersed directly into the soil and connected to the power supply. This procedure was repeated for all three experiments. In experiment I, no electrokinetic treatment was applied (control). In experiment II, the electrical field was provided using constant polarities and in experiment III the electrical field was provided using polarities alternating at the electrodes every 12 h. Each experiment was conducted at room temperature ($25 \pm 1^\circ\text{C}$), which was controlled by air conditioning. Three parallel sampling lines along the length of the soil cell, each with six equally spaced sampling points, provided a total of eighteen soil samples per sampling round at 0 day, 7 days, 14 days, 28 days, 60 days and 90 days (Fig. 1). Samples (10 mm diameter \times 8 cm height) were collected by a metal tubing (10 mm inner diameter). For each sampling round, the samples at the same distance from the three sampling lines were thoroughly mixed together to form one composite sample before analysis.

2.5. Analytical procedure

The concentration of PAHs, bacterial concentration, soil pH, temperature and moisture content were analyzed, all analytical determinations were performed in triplicate, and the results were calculated as means. All data obtained in the study are presented as mean \pm standard deviation (SD). Statistical analysis was performed with SPSS for Windows Ver. 11.5.

Bacteria concentration was determined by preparing serial dilutions of soil suspensions in sterilized water, plating onto LB-agar plates, and incubating at 30°C for 3 days. Bacterial counts were calculated per gram of air-dried soil and log-transformed ($\log_{10} x$) to improve the homogeneity of the variance of the data. Moisture content was measured by heating to 105°C until constant weight was achieved and soil pH was measured by pH meter under a soil-to-water ratio of 1:2.5 [35]. The soil temperature was monitored and taken down by digital thermometer every day.

The total PAHs was extracted according to the EPA Standard Method 3550C (USEPA, 1996) [36]. The concentrations of 16 PAHs (US EPA priority PAHs) were determined by high performance liquid chromatography (HPLC, Waters) equipped with a variable wavelength fluorescence detector (FLD, waters 2475) and a Waters PAHs Column (250 mm \times 4.6 mm i.d., $5 \mu\text{m}$ particle size). Prior to injection, the extraction of PAHs was filtered through a $0.22 \mu\text{m}$ Teflon filter. The injection volume was set at $10.0 \mu\text{l}$ and the column temperature was 25.0°C . The gradient elution program used consisted of 60% water and 40% acetonitrile for 2 min, then programmed to 100% acetonitrile in 12 min at a flow rate of 1.0 ml/min.

3. Results and discussion

3.1. Changes in soil pH

The changes of soil pH in the three experiments are presented in Fig. 2. The average values of soil pH did not show significant changes during the experiment period (Fig. 2A). The change extents of soil pH were between 0.1% and 1.4% in three experiments. Fig. 2B shows the changes of soil pH with distance from the anode at 90 days. The results showed that the soil pH remained broadly constant in experiments I (control) and III (alternating polarity electrokinetics). But the soil pH had a little change at the anode and the cathode in experiment II (constant polarity electrokinetics). The pH of the soils decreased to 8.13 near the anode while increased to 8.40 near the cathode (Fig. 2B), and this was attribute to the electrolysis reaction. The electrolysis of water results in the formation of H^+ at the anode (low pH region) and OH^- at the cathode (high pH region) under constant polarity electrokinetics [37–39]. However, the H^+ and OH^- generated at electrodes may be neutralized automatically under alternating polarity electrokinetics. Gee et al. proposed that soil buffer capacity increased as the amount of calcium carbonates in the soil increased [40]. The test soils had a pH of 8.36 and a high buffering capacity because of the high carbonate mineral content [41]. Thus, test soils kept the weak alkaline all the time under electric fields.

3.2. Changes in moisture content

Applying an electric field may induce changes in soil pH, moisture content and soil temperature, further influencing the growth and distribution of soil microorganisms [42–44]. During the experimental period, the soil temperature presented little change and was consistent with room temperature ($25 \pm 1^\circ\text{C}$). Although the tests showed that the electric field did not influence soil pH and temperature, it did affect soil moisture content and distribution (Figs. 3 and 4). Over the duration of study, the soil moisture content of experiments II and III (with electrokinetics) decreased by 7.0% and 6.6%, about one time greater than in the control experiment (I) (3.8%) (Fig. 3). Under constant polarity electrokinetics, soil moisture content decreased at the anode and increased at the cathode, an indication of pore liquid transport due to electro-osmosis [38]. After running for 14 days, the soil moisture content near the anode dropped by 5.1% while near the cathode it increased by 4.5% in experiment II (Fig. 4A). Fig. 4B shows that under alternating polarity electrokinetics soil moisture content distribution remained even. In experiment III, the soil moisture content stayed within the range

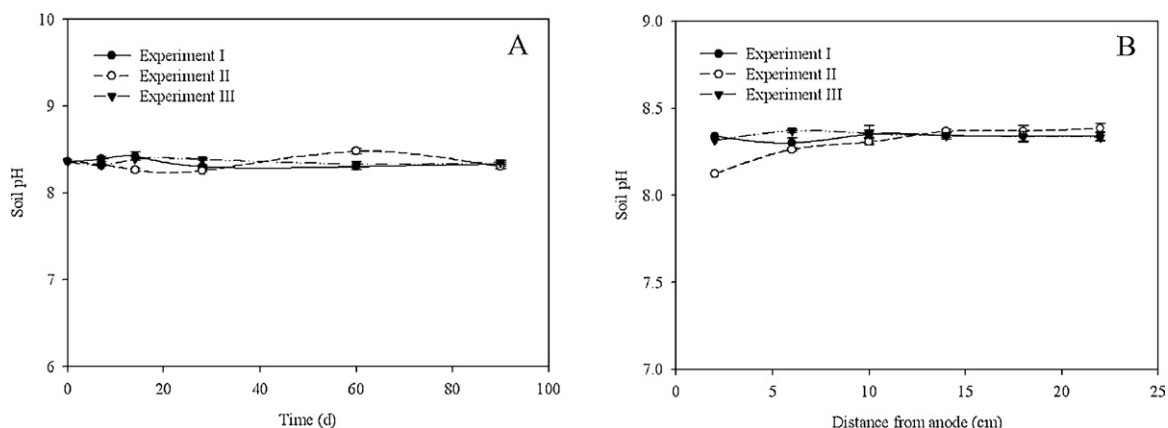


Fig. 2. Changes in soil pH in the three experiments. (A) During the experimental period and (B) at 90 days.

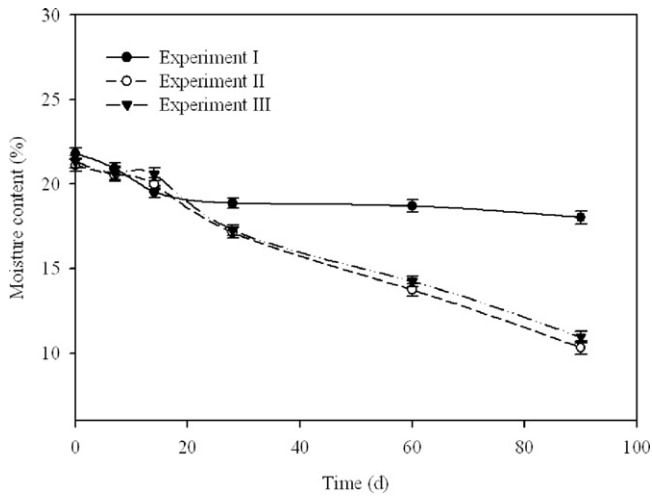


Fig. 3. Changes in soil moisture content in the three experiments.

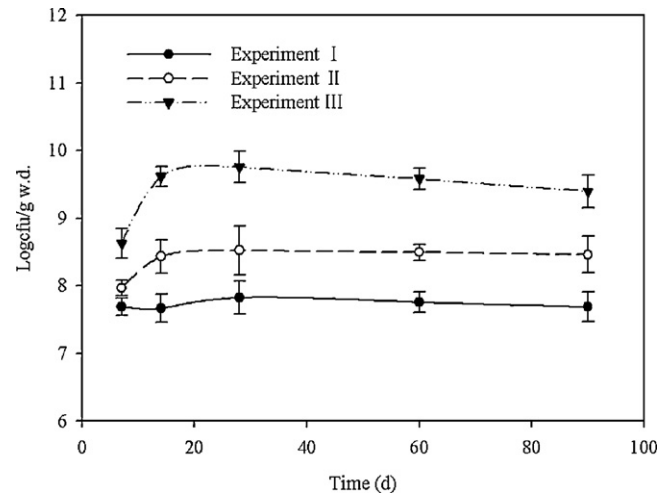


Fig. 5. Changes in bacterial counts as the log of colony forming units (cfu) in the three experiments.

18.4–22.6% within 14 days, indicating that alternating polarity electrokinetics could overcome pore liquid single-direction transport, thus causing soil moisture content to be subject to little change (Fig. 4B). In addition, alternating polarity electrokinetics may also have helped stabilize homogenous distribution and availability of ionic nutrients [45,46]. From the results of these two experiments, over the period 28–90 days the moisture content have the same tendency as the changes during the period 0–14 days. The result is identical to the findings of Luo et al. [39]. Therefore, alternating polarity electrokinetics may be favorable for in situ bioremediation.

3.3. Changes in culturable bacterial counts

The counts of culturable bacteria (colony-forming unit, cfu) in soils changed during the tests, as shown in Fig. 5. Bacterial counts remained constant in the control experiment (I) over the experimental period (i.e. 7.67–7.82 log₁₀ cfu/g dry soil). In experiments II and III, bacteria counts increased rapidly within the first 28 days of testing and then started to decrease gradually. The bacterial counts of experiments II and III (with electrokinetics) increased by an order of magnitude of 0.26–2.58 compared to the control experiment (I). In experiment III (alternating polarity electrokinetics), the counts of bacteria were higher by 9.3% compared to experiment II (constant polarity electrokinetics). These findings show that the activity

of PAHs-degrading bacteria is stimulated when exposed to appropriate electrokinetics and an alternating polarity approach is better for bacterial growth. This observed influence of electrokinetic treatment on microbial activity and community is in line with the results of Kim et al. and Lear et al. [47,9].

The bacterial counts also varied spatially under electrokinetic conditions, as shown in Fig. 6. In the experiment II with constant polarity electrokinetics, the bacterial counts gradually increased from anode to cathode (Fig. 6A). Peak bacterial counts were observed at 18 cm from the anode with counts increasing from 7.71 to 10.31 log₁₀ cfu/g dry soil, while near the anode counts dropped to about 6.24 log₁₀ cfu/g dry soil. This result suggests that, under constant polarity electrokinetics, proximity to the cathode as opposed to the anode is more beneficial for bacterial growth. Under alternating polarity electrokinetics, bacterial counts in experiment III increased from 7.71 to 10.93 log₁₀ cfu/g dried soil and were higher in the middle region rather than near the electrodes (Fig. 6B). The test soils had a pH of 8.36 and a high acid buffering capacity because of the high carbonate mineral content [41]. During the tests, the soil average pH did not show any apparent changes and stayed weakly alkaline in all experiments (Fig. 2). Thus, the changes of the soil moisture content and ionic nutrients in water may be the factors for influencing bacterial activity and counts (Figs. 4 and 6) [48].

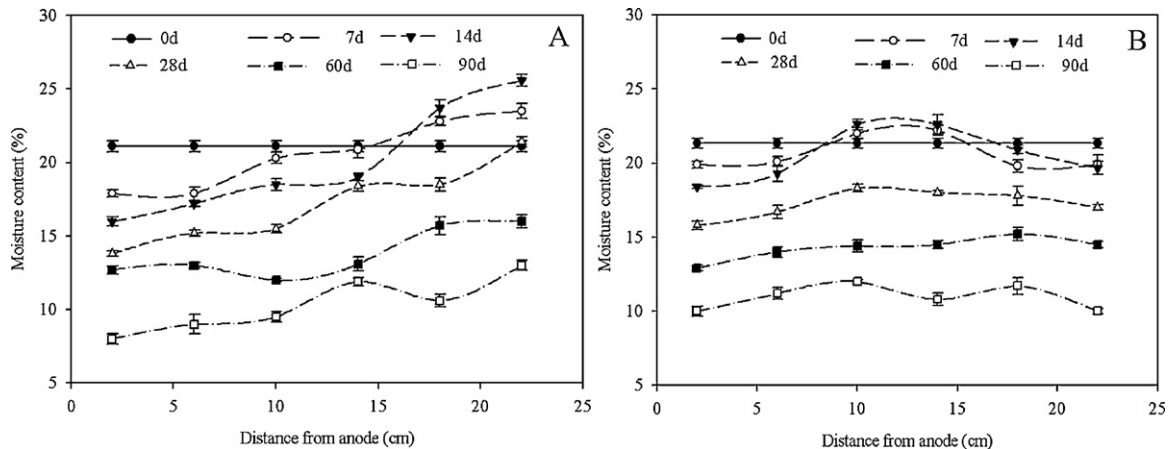


Fig. 4. Changes in soil moisture content under different electrokinetic operation modes. (A) Experiment II and (B) experiment III.

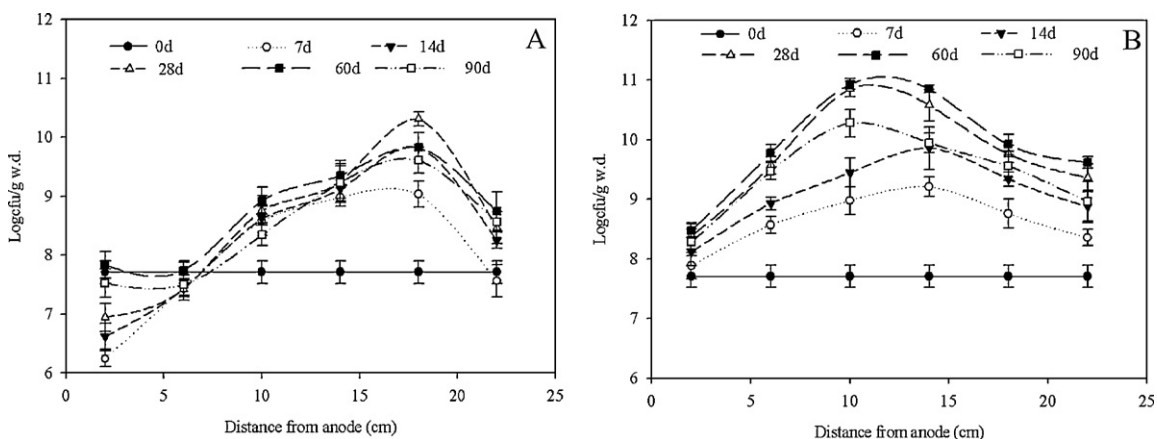


Fig. 6. Changes in bacterial counts under different electrokinetic operation modes. (A) Experiment II and (B) experiment III.

3.4. Total PAHs degradation

Fig. 7 presents the average degradation extent of total PAHs for all samples (18 samples) in the three experiments. As is evident, all three experiments were similar insofar as their degradation extent of total PAHs of different regions in soils. For experiment II, which was fastest in the initial stages and then stalled toward the end of each experiment at 90 days. On the other hand, results differed in that the degradation extent of total PAHs was significantly higher in experiments II and III (electrokinetics and bacteria) than in the control experiment (I) (bacteria only). Under continuous operation for 90 days, the degradation extent of total PAHs in experiments II and III reached 24.3% and 28.3%, respectively, which was about 9.5% and 13.5% higher than in experiment I (14.8%). In addition, the total PAHs degradation extent in experiment III was higher than that in experiment II, indicating that alternating polarity electrokinetics enhanced the biodegradation of PAHs more effectively than with constant polarity electrokinetics. Electric fields have been applied in soil to increase contact opportunities among bacteria, nutrients and pollutants [24]. When an alternating polarity electrokinetics is imposed upon soil, it can change the movement direction, and hence may produce more opportunities for the bacteria, nutrients and contaminants in soils to contact and interact with each other [29]. This result is similar to those found by Luo et al. and Fan et al. when they applied an alternating direct current to phenol-contaminated soil [34,39].

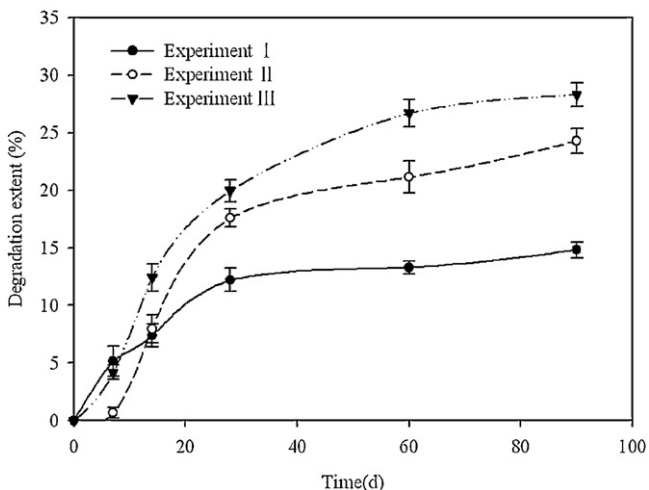


Fig. 7. Progression of the total PAHs degradation extents during the experimental period.

Fig. 8 shows a comparison of PAHs degradation extents in relation to distance from the anode at different times throughout the course of experiments II and III. The results show that the electrokinetic operation mode could affect the degradation extent of total PAHs of different regions in soils. For experiment II, which used constant polarity electrokinetics, the degradation of PAHs was limited near the anode, but gradually increased and peaked at a distance of 18 cm, before decreasing slightly toward the cathode (Fig. 8A). However, under the alternating conditions of experiment III, the degradation extent of PAHs was basically equal from anode to cathode, albeit slightly lower near to both electrodes (Fig. 8B). Compared to Figs. 4, 6 and 8, there was the same tendency among soil moisture content, culturable bacterial counts and degradation extent of PAHs. These results indicated that the changes of soil moisture content and bacterial counts affected the degradation extents of PAHs. These observations are similar to those of Harbottle et al. who studied the use of alternating polarity electrokinetics to enhance the biodegradation of pentachlorophenol in unsaturated soil [27].

3.5. Degradation of heavy PAHs

Fig. 9 shows the differences in degradation extents for three- to six-ring PAHs in the three experiments. Compared to the control experiment (I), the degradation extents were higher in the electrokinetic experiments (II and III) for all ring numbers. Furthermore, for four- to six-ring PAHs, degradation extents were more enhanced at 90 days, indicating that electrokinetics could promote the biodegradation of heavy PAHs. For the three-ring PAHs, the degradation extents were higher in experiments II and III than experiment I from 14 days until the conclusion of the experiments at 90 days, with values being almost the same in all three experiments at 90 days (Fig. 9A). However, for four- to six-ring PAHs, the degradation extents increased significantly from 14 days up until 90 days in experiments II and III, and the degradation extent increased as the ring number increased (Fig. 9B–D). It may be due to varying resonance properties (be induced by electrokinetics) of different size PAHs and degradation microorganisms, and that further research into these particular effects is required. The four- to six-ring PAHs degradation extents increased by 7.9–8.6%, 11.0–18.4% and 17.2–25.6% in experiments II and III compared to experiment I, respectively, which were far higher than that for three-ring PAHs (2.2%) at 90 days (Fig. 9B–D). These results indicate that electrokinetics could promote the biodegradation of heavy PAHs and electro-bioremediation is a potentially useful approach for the treatment of soil contaminated with heavy PAHs. The results

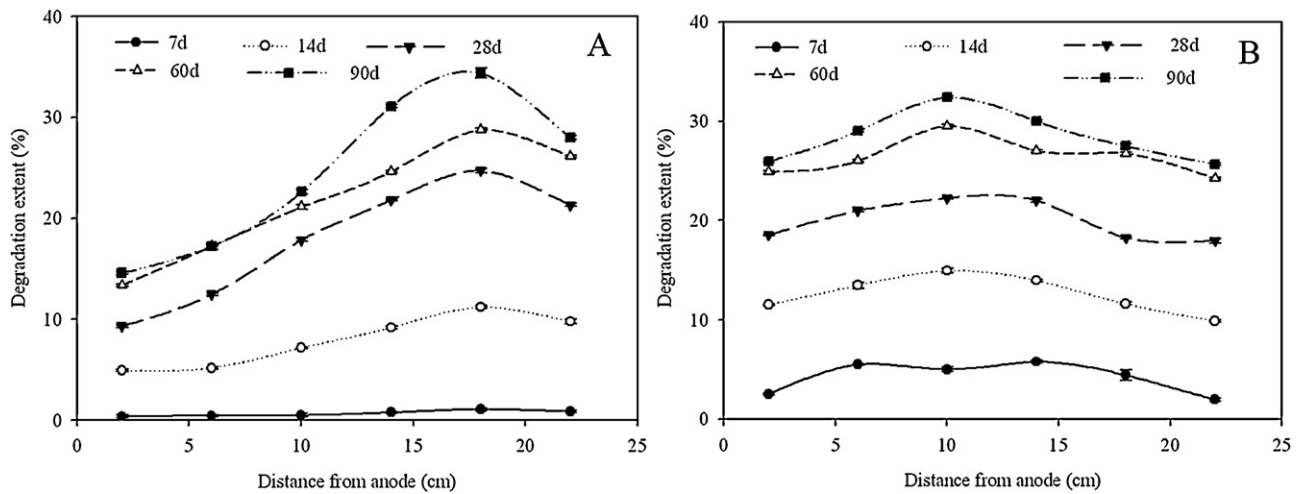


Fig. 8. Changes in total PAHs degradation extents under different modes of electrokinetic operation. (A) Experiment II and (B) experiment III.

also show that alternating polarity electrokinetics is more favorable for PAHs degradation than constant polarity electrokinetics.

3.6. Relationships among degradation extent, bacterial counts and soil moisture content

The present study has shown that the electrokinetic operation mode could affect soil moisture content (Figs. 3 and 4), bacterial counts (Figs. 5 and 6) and the degradation extent (Figs. 7 and 8). Moreover, the distribution change profiles of the above three factors have the same tendency under electrokinetics. In order to

further study the relationships, Pearson correlation analysis among degradation extent, bacterial counts and moisture content was performed, as shown in Table 2. The results show that degradation extent and bacterial counts, as well as degradation extent and moisture content, have significant positive correlations, reaching 0.919 and 0.787, respectively (significant at the 0.01 level). Furthermore, correlation between bacterial counts and moisture content also 0.643 (significant at the 0.05 level). Electrokinetics inevitably impacts upon soil conditions [42], such as pH and soil moisture content, which are critical for soil microbial community composition and processes [37]. In this study, electrokinetics resulted

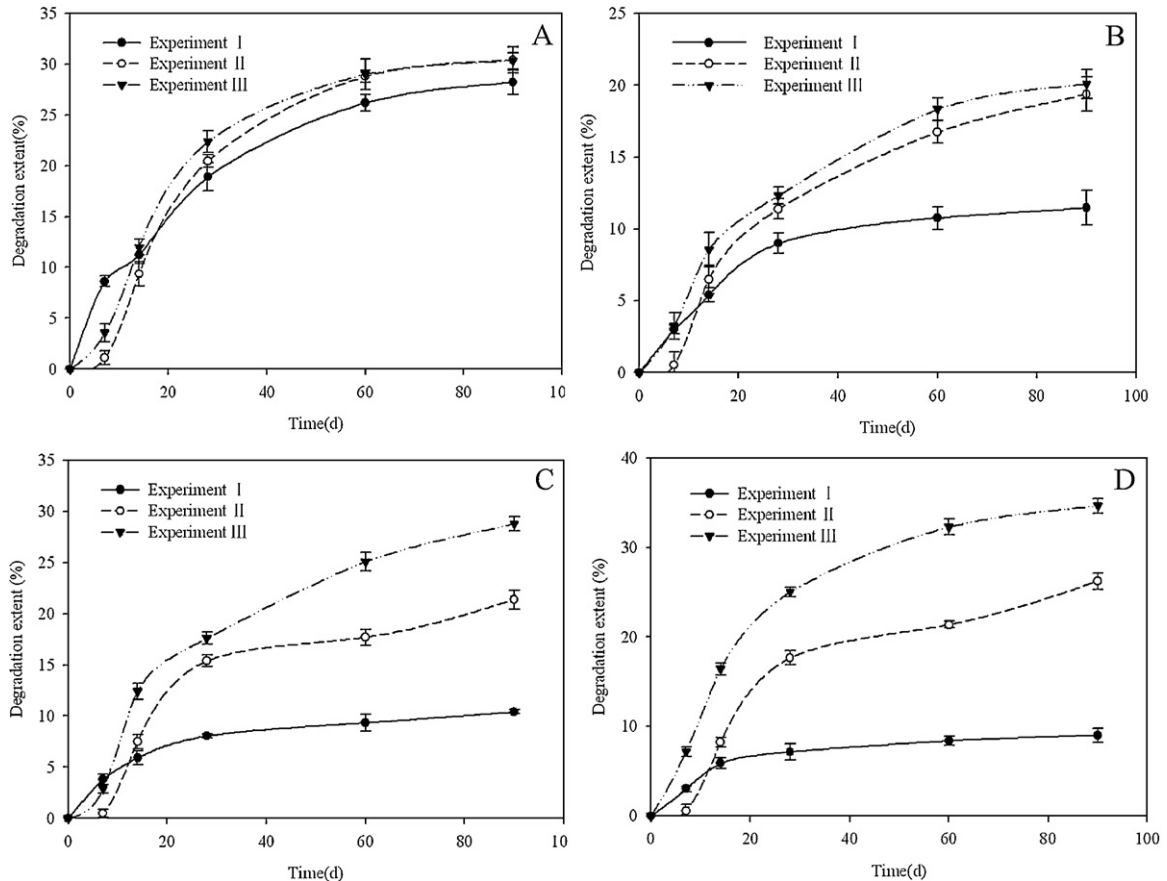


Fig. 9. Degradation extents of three- to six-ring PAHs under different operation modes. (A) Three rings; (B) four rings; (C) five rings; (D) six rings.

Table 2
Pearson correlation analysis among degradation extent, bacteria counts and moisture content.

	Degradation extent (%)	Bacteria counts (log cfu/g w.d.)	Moisture content (%)
Degradation extent (%)	1	0.919**	0.787**
Bacteria counts (log cfu/g w.d.)	0.919*	1	0.643*
Moisture content (%)	0.787**	0.643*	1

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

in the movement of soil moisture content which transported the nutrients (e.g. K^+ and NH_4^+) [44,45,49], thus affecting the soil bacterial counts and activity, which in turn influenced the degradation extent of PAHs.

4. Conclusions

This study has shown that electrokinetics may enhance the biodegradation of heavy PAHs. Moreover, alternating polarity electrokinetics has been found to be more efficient for the biodegradation of heavy PAHs than constant polarity electrokinetics. Under alternating polarity electrokinetics, the properties of soil are less impacted upon and adapt to the growth of microorganisms. Therefore, electro-bioremediation is a potential approach for the treatment of soil contaminated with heavy PAHs.

Acknowledgments

We gratefully acknowledge the constructive comments of the three reviewers and their suggestions for improving the paper. The funding for this work was provided by the project supported by the National Natural Science Foundation of China (21047006) and the Knowledge Innovation Project Key-Direction Project Sub-project of Chinese Academy of Sciences (No. KZCX2-EW-407).

References

- [1] K.L. White, *Journal of Environmental Science and Health* 4 (1986) 163.
- [2] L. Flowers, S.H. Rieth, V.J. Cogliano, G.L. Foureman, R. Hertzberg, E.L. Hofmann, D.L. Murphy, S. Nesnow, R.S. Schoeny, *Polycyclic Aromatic Compounds* 22 (2002) 811.
- [3] B.R.T. Simoneit, *Atmospheric Environment* 33 (1984) 5071.
- [4] F.M. Li, S.H. Guo, B. Wu, H.F. Ye, *Chinese Geographical Science* 21 (2011) 185.
- [5] S. Jonsson, Y. Persson, S. Frankki, B.V. Bavel, S. Lundstedt, P. Haglund, Ms. Tysklind, *Journal of Hazardous Materials* 149 (2007) 86.
- [6] E.E. Diplock, D.P. Mardlin, K.S. Killham, G.I. Paton, *Environmental Pollution* 157 (2009) 1831.
- [7] A.R. Johnsen, L.Y. Wick, H. Harms, *Environmental Pollution* 133 (2005) 71.
- [8] H. Harms, L.Y. Wick, *Engineering in Life Sciences* 6 (2006) 252.
- [9] G. Lear, M.J. Harbottle, G. Sills, C.J. Knowles, K.T. Semple, I.P. Thompson, *Environmental Pollution* 146 (2007) 139.
- [10] S. Suni, E. Malinen, J. Kosonen, H. Silvennoinen, M. Romantschuk, *Journal of Environmental Science and Health A* 42 (2007) 277.
- [11] L. Shi, S.S. Müller, N. Loffhagen, H. Harms, L.Y. Wick, *Microbial Biotechnology* 1 (2008) 53.
- [12] S. Pamukcu, J.K. Wittle, *Environmental Progress* 11 (1992) 241.
- [13] K.R. Reddy, S. Chinthamreddy, *Waste Management* 19 (1999) 269.
- [14] J.L. Chen, S.F. Yang, C.C. Wu, S. Ton, *Separation and Purification Technology* 79 (2011) 157.
- [15] M.M. Page, C.L. Page, *Journal of Environment Engineering* (2002) 208.
- [16] S.A. Jackman, G. Maini, A.K. Sharman, F. Sunderland, C.J. Knowles, *Biotechnology and Bioengineering* 74 (2001) 40.
- [17] H. Gomes, C. Dias-Ferreira, A.B. Ribeiro, *Chemosphere* 87 (2012) 1077.
- [18] L.Y. Wick, P.M. Mattle, P. Wattiau, H. Harms, *Environmental Science and Technology* 38 (2004) 596.
- [19] S. Suni, M. Romantschuk, *FEMS Microbiology Ecology* 49 (2004) 51.
- [20] C. Comminellis, *Electrochimica Acta* 39 (1994) 1857.
- [21] S.A. Jackman, G. Maini, A.K. Sharman, G. Sunderland, C.J. Knowles, *Biotechnology and Bioengineering* 74 (2001) 40.
- [22] L. Shi, S. Müller, H. Harms, L.Y. Wick, *Environmental Geochemistry and Health* 30 (2008) 177.
- [23] R.A. Torres, W. Torres, P. Peringer, C. Pulgarin, *Chemosphere* 50 (2003) 97.
- [24] Q.S. Luo, X.H. Zhang, H. Wang, Y. Qian, *Chemosphere* 59 (2005) 1289.
- [25] L.Y. Wick, L. Shi, H. Harms, *Electrochimica Acta* 52 (2007) 3441.
- [26] K.R. Reddy, R.E. Saichek, *Journal of Environmental Science and Health A* 39 (2004) 1189.
- [27] M.J. Harbottle, G. Lear, G.C. Sills, I.P. Thompson, *Journal of Environment Management* 90 (2009) 1893.
- [28] R.E. Saichek, K.R. Reddy, *Chemosphere* 51 (2003) 273.
- [29] A.N. José-Luis, B.M. Marisa, P.B. Rosa, J.O.C. José, *Environmental Pollution* 142 (2006) 326.
- [30] P.T. Duong, S.R. Amatya, S. Mika, *Journal of Environment Engineering* 136 (2010) 739.
- [31] M.T. Alcántara, J. Gómez, M. Pazos, M.A. Sanromán, *Journal of Hazardous Materials* 179 (2010) 1156.
- [32] A. Klute, *Methods of Soil Analysis. Physical and Mineralogical Methods*, Soil Science Society of America, Madison, WI, 1986.
- [33] A.L. Page, R.H. Miller, D.R. Keeny, *Methods of Soil Analysis. Chemical and Microbiological Properties*, Soil Science Society of America, Madison, WI, 1982.
- [34] X.Y. Fan, H. Wang, Q.S. Luo, J.W. Ma, X.H. Zhang, *Journal of Hazardous Materials* 148 (2007) 29.
- [35] R.K. Lu, *Analytic Methods for Soil Agricultural Chemistry*, China Agriculture Science and Technology Press, Beijing, 2000, p. 13.
- [36] USEPA Method 3550C: Ultrasonic Extraction, United State Environmental Protection Agency, Washington, DC, 1996.
- [37] G. Lear, M.J. Harbottle, C.J. van der Gast, S.A. Jackma, C.J. Knowles, G. Sills, I.P. Thompson, *Soil Biology and Biochemistry* 36 (2004) 1751.
- [38] J. Virkutyte, M. Sillanpää, P. Latostenmaa, *Science of the Total Environment* 289 (2002) 97.
- [39] Q.S. Luo, X.H. Zhang, H. Wang, Y. Qian, *Journal of Hazardous Materials B* 121 (2005) 187.
- [40] C. Gee, M.H. Ramsey, I. Thornton, *Applied Geochemistry* 16 (2001) 1193.
- [41] F. Cao, Study on effective utilization of mineral resources of Benxi area, Master Thesis, Northeastern University, 2005.
- [42] Y.B. Acar, A.N. Alshawabkeh, *Environmental Science and Technology* 27 (1993) 2638.
- [43] Y.B. Acar, A.N. Alshawabkeh, *Journal of Geotechnical Engineering* 122 (1996) 173.
- [44] Q.S. Luo, X.H. Zhang, H. Wang, Y. Qian, *Techniques and Equipment for Environmental Pollution Control* 5 (2004) 40.
- [45] C.A.B. Schmidt, M.C. Barbosa, M.S.S. Almeida, *Journal of Hazardous Materials* 143 (2007) 655.
- [46] S.T. Lohner, D. Katzoreck, A. Tiehm, *Journal of Environmental Science and Health A* 43 (2008) 913.
- [47] S.H. Kim, H.Y. Han, Y.J. Lee, C.W. Kim, J.W. Yang, *Science of the Total Environment* 408 (2010) 3162.
- [48] X.J. Chen, Z.M. Shen, Y.M. Lei, S.S. Zheng, B.X. Ju, W.H. Wang, *Soil Science* 171 (2006) 638.
- [49] Y.B. Acar, M.F. Rabbi, E.E. Ozsu, *Journal of Geotechnical and Geoenvironmental Engineering* 123 (1997) 239.