

Remediation of Chrysene from Contaminated Soil by Enhanced Electrokinetics

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Polycyclic aromatic hydrocarbons like chrysene in soil is difficult to remove because of insolubility in water well adsorbed in soil plus their persistence. They are well known for the health hazardous elements. Enhanced electrokinetic method could be used to desorb and destroy the pollutants. The main objective of this research was to investigate the technical feasibility of AC/Ultrasonic electrokinetic method in remediation of chrysene from contaminated soil. Three lab scale experiments including electrokinetic alone (EK), ultrasonic electrokinetic (EK-US) and AC-electrokinetic (EK-AC) were carried out to remove chrysene from artificially contaminated kaolin. There was increase in removal of chrysene by 3-4 % by enhanced electrokinetics using ultrasound and AC electricity. The technical feasibility of AC electrokinetic method in remediation of chrysene from the contaminated soil was investigated. The results suggest that the utilization of AC and US enhanced the electrokinetic removal of chrysene.

Keywords: AC current; electrokinetics; organic pollutants; ultrasound

1. INTRODUCTION

Electrochemical remediation is defined as a method that removes contaminants using a direct current to either mobilize or decompose them in soils, sediments and water. This method is applicable for both organic and inorganic pollutants. However, persistent organic pollutants such as polycyclic aromatic hydrocarbons (PAHs) in soil are difficult to be removed by electrokinetic treatment because the pollutants adsorb strongly in the soil and have long half lives. Therefore, electrochemical remediation alone is not efficient in remediation of chrysene- contaminated soil. There are many ways to enhance the electrochemical technique. These include chemical and biological methods and application of ultrasound radiation [1-3]. The use of chemicals raises operational costs and may cause environmental hazards, while biological methods are slow. Ultrasound may be one of the best

technologies to enhance the electrochemical treatment [4]. The application of alternative current (AC) in the electrochemical technology may also increase the efficiency of remediation. AC electrochemical method uses electric field to generate forces that act on fluids or particles (including those made of dielectric or biological materials) and cause them to move in different ways [5-7]. Induced charge electroosmosis refers to electroosmotic fluids flow that occurs on polarizable surfaces as a result of induction of surface charges and subsequent formation of double layer [8, 9].

The objective of the present study was to study the removal of chrysene, a persistent highly insoluble four fused benzene ring hydrocarbon, by combined AC electrochemical method and ultrasonically assisted EK.

2. EXPERIMENTAL PART

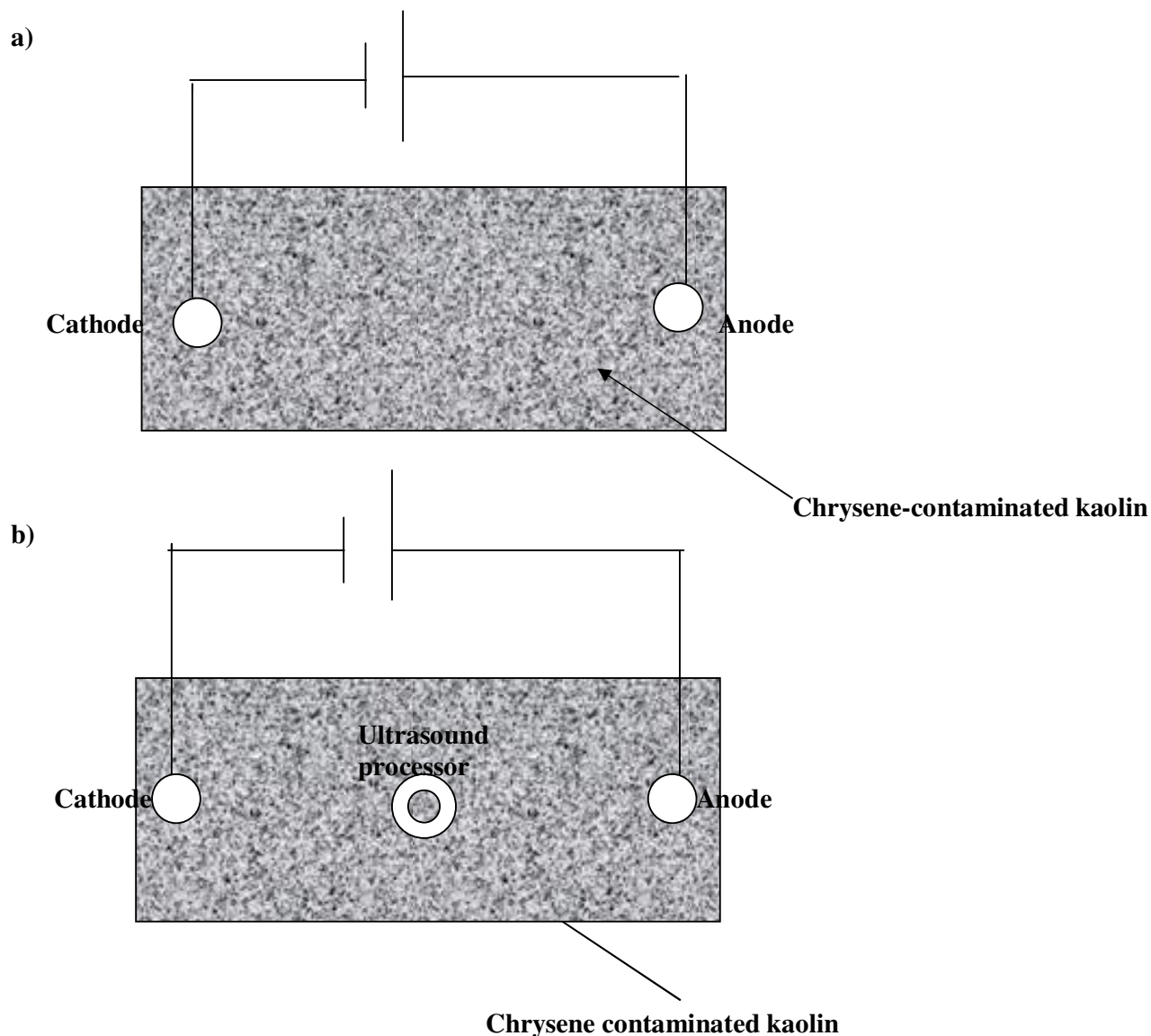
Chrysene was purchased from Acrös organics and hexane was supplied by Merck. All chemicals were of analytical grade. The ultrasonic processor used in these experiments was UP100H with an operating frequency of 30 kHz, power of 100 W and 10 mm of horn diameter (Hielscher – Ultrasound Technology Company). The power of these ultrasonic processors could be controlled in the amplitude range of 20-100%. Graphite electrodes of 10 cm long and 10 mm diameter were connected to the direct current dual power supply. Kaolin was used as a model of low permeability clayey soil. The physico-chemical properties of kaolin, determined based on the methods prescribed by Soil Science: Methods and Applications [10], are presented in Table 1.

Table 1. Characteristics of kaolin

Properties	Values
kaolin	100 %
colour	white
pH	5.0
Specific density	0.51
Moisture	1.11 %
Total organic carbon	Nearly 0
Carbonate content	5.5 %
Electrical conductivity	189 μ S
Cation exchange capacity	3 m mol/100g
Particle size distribution	
sand	3.9 %
silt	20.2 %
clay	75.9 %

Three different experiments of EK, EK-US, EK-AC were carried out at ambient temperature in an open pan without separating chambers for electrodes simultaneously. The dimensions of pan made of HDPE were 20 cm \times 14 cm \times 8 cm (Figs. 1a-1c). For contamination of soil, chrysene was weighed for 100 mg/kg soil. They were dissolved in hexane and then mixed with soil. The soil and solvent were mixed well with stainless steel spoon to make homogenous distribution of compounds in the soil.

Then, the slurries were kept in flume wood for about 7 or more days to assure total evaporation of solvent [2, 3, 11]. Before starting each experiment, the concentration of pollutants in the soils was measured. Approximately 500 g of kaolin contaminated with chrysene was transferred into each pan. Electrodes were inserted into soil at end of the pan. There was one pan with any treatment for reference. These laboratory experiments were designed for on site implementation. The tap water was used to make slurry of the soil. During the experiments, the ratio of water and soil was maintained at about 2:1. The hydraulic transport of soil in slurries was typically 40% by weight [12]. During the experiments, the voltages were kept constant at 30 V (the voltage gradient was 1.5 V/cm), and changes in currents were recorded periodically. The kaolin-water mixed slurry in EK-US tests was subjected to ultrasonic waves at the same condition of 5 hours on the first day and 1 hour per day for the remaining of experimental (10 days). In the case of EK-AC, the AC electric field was applied from the beginning of the experiments nearby the cathode and anode as shown in Fig. 1c.



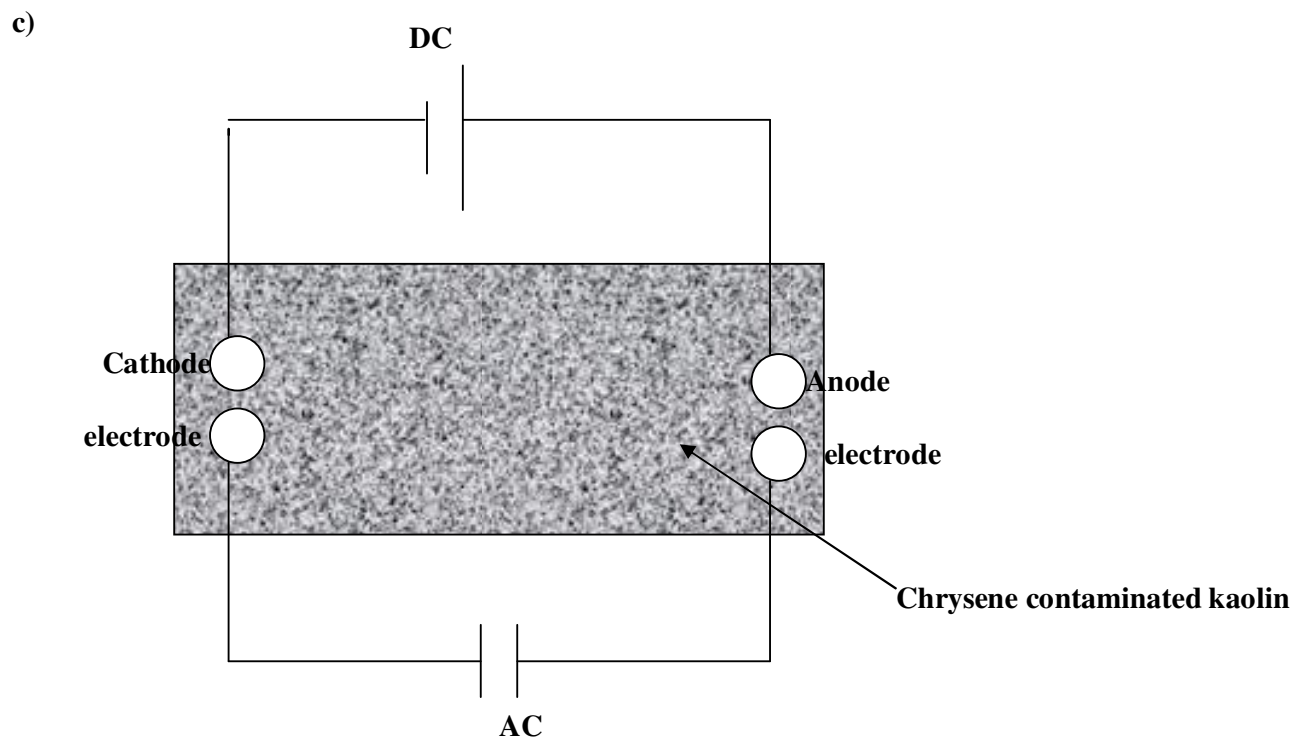


Figure 1. Sketch diagram of configurations on a) EK experiment b) EK-US experiment c) EK-AC

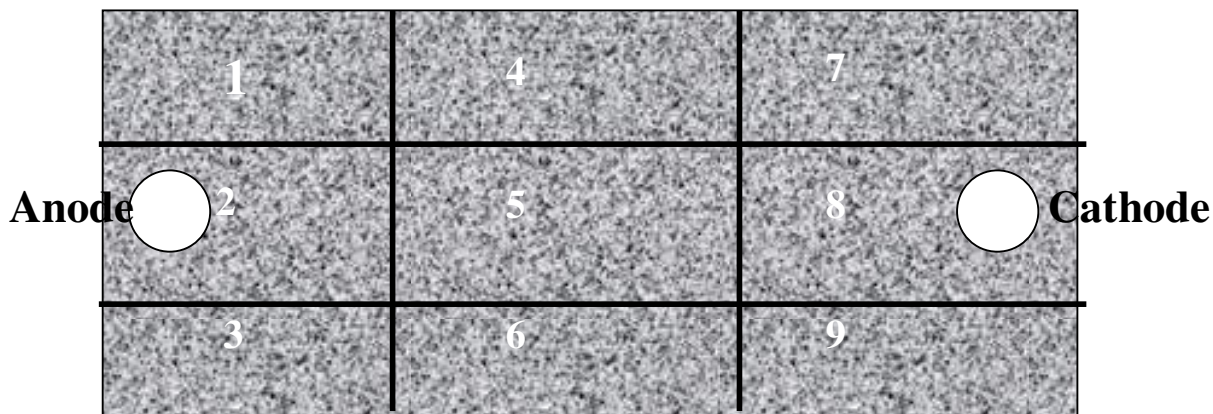


Figure 2. Division of soil for analysis

3. EXTRACTION AND CHEMICAL ANALYSIS

At the end of the experiments, the power supply was switched off. The final soil mass was divided into 9 sections from anode to cathode (Fig 2). These samples were dried in an oven at 80 °C over 24 hours. The dried kaolin was then pulverized for chemical analysis. Samples were analyzed in

duplicates. One gram of sample was mixed with 5 mL hexane in glass tube and was transferred into ultrasonication bath for 30 min to collect the organic compounds extracted from the soil mediums into hexane solvent. The glass tubes were centrifuged at 4000 rpm for 20 min. [13]. The supernatants were then transferred into 2 mL glass tubes. TA Finnigan Trace 2000 GC equipped with a Finnigan Polaris Q mass spectrometer was used to determine the remaining concentrations of the chrysene in the extracts. This method was taken instead of the traditional approach, Soxhlet extraction because it gave nearly identical results that came from traditional method and fast and less organic solvent required [14,15]. The pH of the soil samples were also measured periodically and all sections of final soil mass at the end of the experiments.

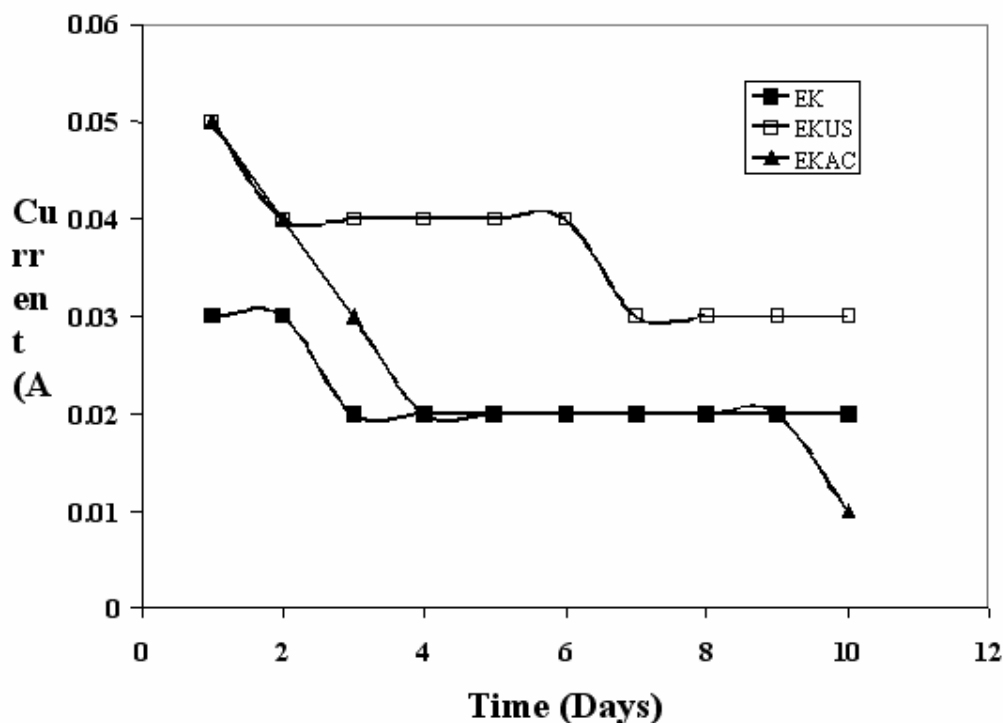


Figure 3. Current changes during the experiments

4. RESULTS AND DISCUSSION

Fig. 3 shows the current as a function of EK treatment time throughout the experiments. The starting current was 0.03 A for the EK. After 3 days, it decreased to 0.02 A and was maintained constant to the end of the experiment. For the EK-US, the initial current was 0.05 A. After 2 days, it changed to 0.04 A and continued up to 6th day. However, it reduced to 0.03 on the 7th day. The starting current was 0.05 A in the EK-AC. It decreased by 0.01A per day up to 4th day and became constant afterwards.

During the experiments the current depends on the concentration of ions present in the soil matrix [16, 17]. The decrement of the current was attributed to the loss of free ionic species in the soils

by mobilisation to respective electrode or due to neutralisation with a protons and hydroxides formed by electrolysis of water [18]. The fluctuation of current during the experiments was affected by the changes in pH due to electrolysis [19]. For the EK-US, the current decreased after 7th day due to the loss of water by ultrasonication in the system. For EK-AC, the proton and hydroxide ions might have formed tern by tern in the same electrodes. This may attribute to the decrease stepwise up to 4th day. After 4th day, both neutralization and solubility rate became constant.

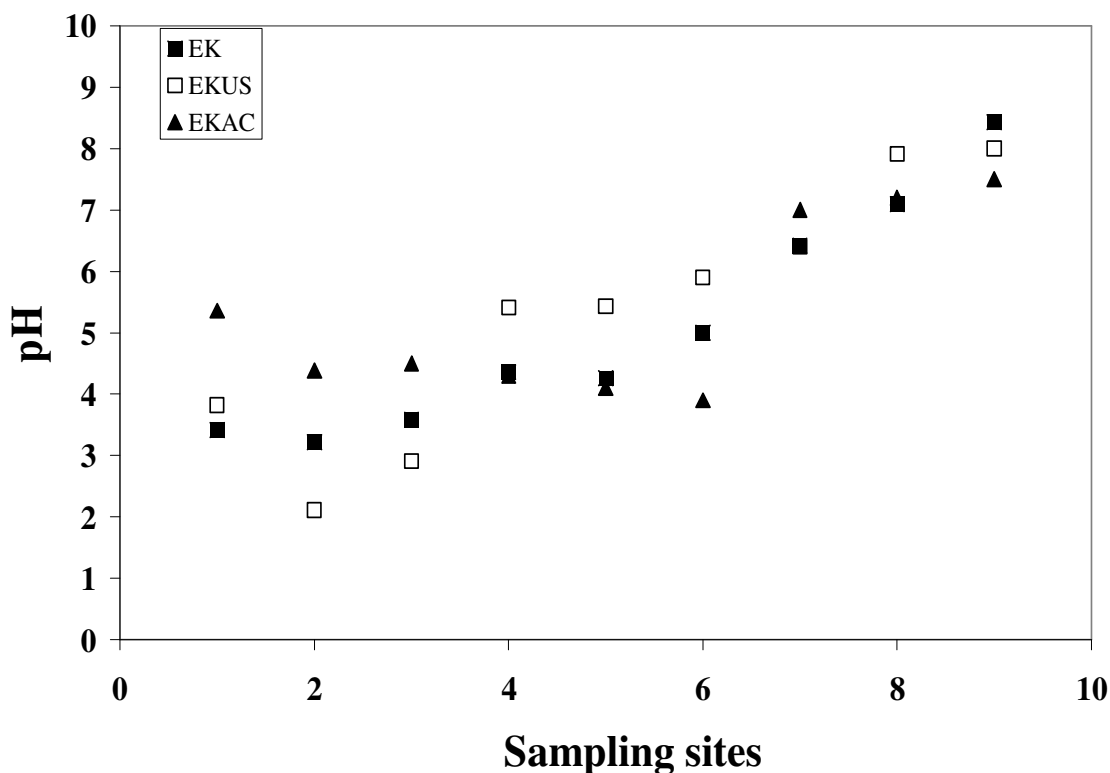


Figure 4. pH variation in the soil profile

Fig 4 shows pH variations in the soil profiles after complete experiments. The pH of the soil close to the anode was acidic (ranging from 4.4 to 2.1). It was significantly lower than that of other soil parts. It was attributed to continuous formation of H^+ ions in soil by the anode. At the same time, pH was higher nearby cathode due to the formation of OH^- ions. The pH was higher at the anode and lower at cathode for EK-AC. The reason may be same as given above.

The removal of chrysene was relatively high close to the anode and the cathode in all cases (Fig. 5). The chrysene moved towards cathode by electroosmosis with water from the anode side and electrophoresis. However, in the pan, the water pushed the soil mass toward the anode. As a result the concentration of chrysene also decreased by dilution. The middle is the region that was least affected by cathode and anode. Therefore, the chrysene accumulated at the centre. The removal performance was comparable because the ultrasound processor was placed at the centre and due to the moisture

content, the ultrasonication worked better nearby the cathode. In general, the overall removal efficiencies of EK, EK-US and EK-AC were 50 %, 54 % and 53 %, respectively.

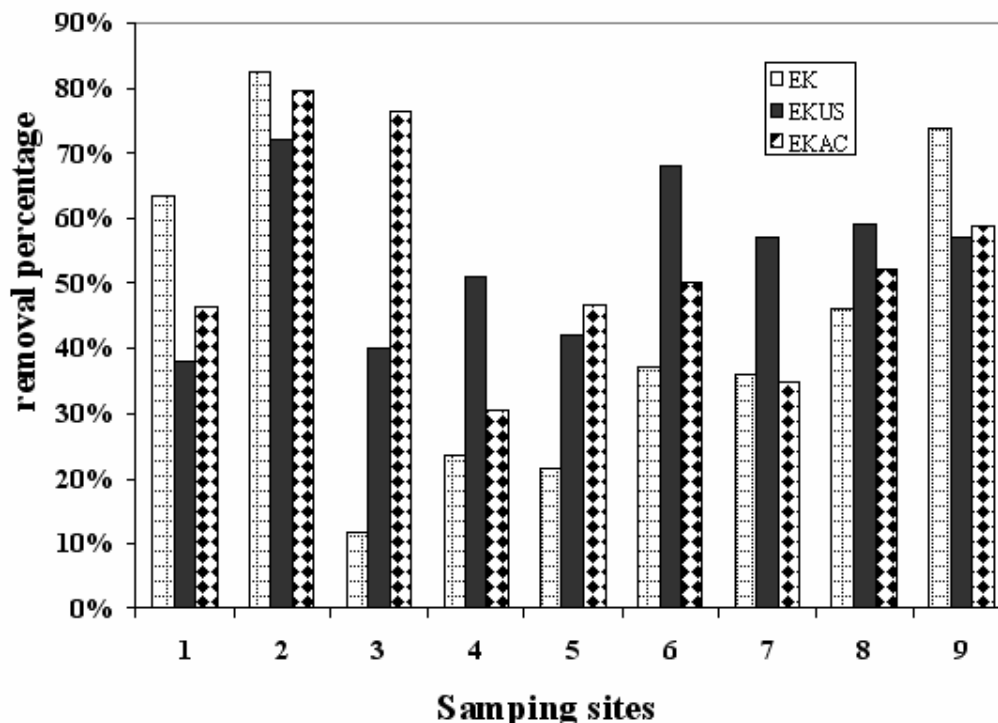


Figure 5. Removal of chrysene from kaolin contaminated soil profile

5. CONCLUSIONS

The technical feasibility of AC electrokinetic method in remediation of chrysene from the contaminated soil was investigated. Thus, we can conclude that the use of the ultrasound and alternative current increased the treatment performance, but the impact was relatively mild. There was increase in removal of chrysene by 3-4 % by enhanced electrokinetics using ultrasound and AC electricity.

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