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Research article

Efficient Removal of Copper and Cadmium from Contaminated Soil Utilizing Electrokinetic Process

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Keywords: soil decontamination, heavy metal, removal efficiency, economy, operating parameters, electrokinetic process

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Abstract

Present manuscript demonstrates the efficient removal of copper and cadmium from contaminated soil utilizing electrokinetic process. The results of systematic bench-scale study on improved extraction of copper and cadmium shows the suitability of process to decontaminate low permeable soil. The influence of operating parameters such as electrode material, soil pH, current density, soil temperature, inter-electrode spacing and initial soil moisture content on the decontamination performance is investigated. Study revealed that, removal of copper and cadmium reached to 89.4% and 62.7% respectively while using titanium electrodes. Removal efficiency improved with the increase in current density, soil temperature and initial soil moisture content. About 74.5% of copper removal achieved within 20 hours at soil temperature of 35°C and current density was kept at 9.3 mA/cm^2. During this study, soil pH decreased from 6.7 to 2.4 near anode and reached up to 12.3 near cathode resulting in augmented metal removal. An improvement of 33% in removal efficiency observed when moisture content increased from 45% to 73%. Rate of metal removal increased as the inter-electrode spacing decreased to 15 cm, which demonstrated the highest copper removal at a current density of 9.3 mA/cm^2. The maximum energy consumption found to be 5.2 kWh/m^3 of soil volume demonstrated the economy of process, which can be scaled up at cleanup sites.

1. INTRODUCTION

Increasing degradation of soil with variety of contaminants including heavy metals is causing significant perturbation to the ecology.[1] In many parts of the world, especially in chemical and power producing industry, soil contamination is increasing at an alarming rate.[2] Heavy metal concentrations more than the permissible limits will generally result in severe negative effects on ecological components in the environment and human health.[3] Due to the higher toxicity level and hazards associated with the metal contamination the Maximum Allowable Concentration Limits (MACL) in soil and in the discharge effluent from industry are set to very low values (table 1).

Researchers have developed technologies in order to remediate such contaminated soils, however, most of them demonstrated significantly lower remediation efficiencies specially when dealing with low permeable soils.[8] Remediation of soil with electrokinetic technique found to be a practical, economical and efficient decontamination technique.[9] The electrokinetic technology is a combination of three processes namely; electroosmosis, electrophoresis and electrochemical processes.[10] With the embarkment of new millennium research on the application of electrokinetic process for the decontamination of soil congregate the attention of many researchers. Some researchers tried to improve the extraction efficiency of process by utilizing various additives including benzoic acid cometabolite,[11] Zinc oxide (ZnO)-*Citrus sinensis* nano-additive[12] and Ethylenediaminetetraacetic acid (EDTA).[13]

A number of researchers explored this area and a large number of laboratory as well as pilot scale studies have been done that describe the contaminant transport in low permeable media.[8,14,15] However, there is a need to explore various operating parameters on which the efficiency and economy of the process depend. Current research is struggling to overcome the mass transfer limitations and reduce the cost and time of treatment.[10]

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TABLE 1. MAXIMUM PERMISSIBLE CONCENTRATION LIMITS OF SOME METAL IN SOIL AND IN THE DISCHARGE EFFLUENT FROM INDUSTRY.

Metals	MACL of Metals in Soil Used for Land Applications (mg/kg)			
	^WHO	*US-EPA	**China	!Hungarian
Cd	0.003	1.6	0.3-0.6	1
Cr	0.1	120	150-300	75
Cu	--	100	--	75
Pb	0.1	60	80	100
Hg	0.08	0.5	0.3-1.0	0.5
Se	--	1.6	--	1
Ni	0.05	32	40-60	40
Zn	--	220	--	200
Ar	--	14	--	15

^Chiroma et al.[4]; *US-EPA[5]; **He et al.[6]; !Mayar[7]

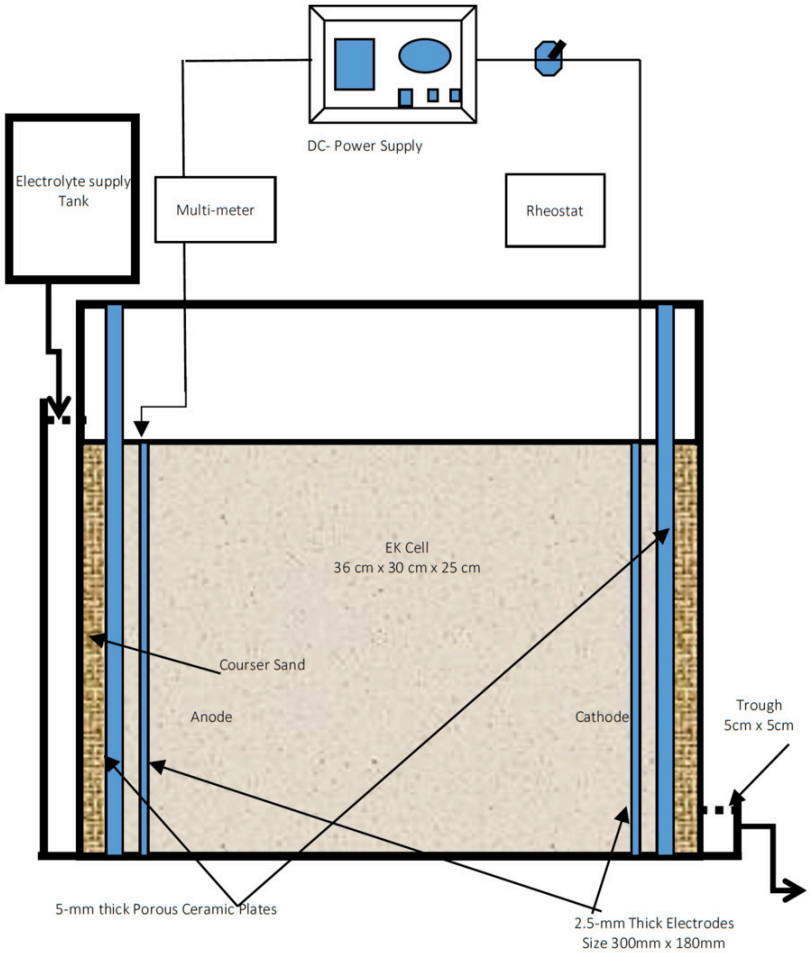

Fig. 1. Schematic of EK soil decontamination setup

2. MATERIALS AND METHODS

A laboratory scale rectangular electrokinetic setup including electrodes and other accessories was prepared in the environmental engineering laboratory of Civil Engineering department at Jubail University College, Saudi Arabia.

The electrokinetic plexiglas cell is a rectangular box having dimensions of 36 cm x 30 cm x 25 cm as shown in figure 1. Cell having one anode and one cathode with thickness of about 2.5-mm having dimension of 28 cm x 18 cm. Electrodes are connected to a constant DC power supply source (GW Instek, SPD-3606 DC power supply) along with a rheostat (DDR/DNR/DSR series, MFPR, Hong Kong) and a

TABLE 2. XRD MINERALOGICAL ANALYSIS RESULTS OF SOIL SAMPLE.

Mineral	Percentage
Quartz	73.4
Calcite	9.3
Kaolinite	1.6
Albite	3.8
Muscovite	2.7
Thuringite	1.6
Cristobolite	1.4
Montmorilonite	6.2

multimeter (8846A, Fluke, USA) to adjust and monitor the voltage and current. Spacing between the electrodes were kept 15 cm in all experimental runs except during the study of inter-electrode separation. In order to provide drainage through cell-porous media, two 5-mm thick porous ceramic plates were provided at the end of the cell adjacent to a coarser sand layer which also prevent the flow of soil and loss of cell mass (figure 1). A separate electrolyte tank has been used to provide a controlled flow of 0.5 ml/hr except during study of initial soil moisture content. All the analysis performed was based on standard procedures.[16]

3. RESULTS AND DISCUSSION

XRD mineralogical analysis results of soil sample are presented in table 2. Results indicate the presence of major part of quartz and calcite. The clay fraction is mainly consisting of montmorillonite, muscovite and kaolinite. Therefore, it gives the partially swelling type characteristic to the soil sample. Calcite present in the soil (9.3%) provided buffering during electrokinetic remediation runs. Soil samples were also analyzed from standard geotechnical and physicochemical analysis during electrokinetic remediation study and results of analysis are shown in table 3. Results of soil metal analysis are presented in table 4 which is showing high concentration of copper and cadmium.

3.1 EFFECT OF ELECTRODE MATERIAL

The compatibility of electrode materials has great importance in electrokinetic process to improve and optimize the process efficiency and economy of the treatment process. Therefore, in order to select the suitable material for present study materials namely titanium, aluminum, nickel and 304 stainless steel were tested to find a suitable candidate electrode material.

Temporal variation of copper and cadmium at the middle of soil sample was monitored while applied current density was 9.3 mA/cm^2. Selected electrodes were tested and removal efficiency of metals were noted. Results are presented in figure 2. It can be seen that the removal efficiency of all electrode materials are higher in case of copper removal as compared to cadmium. This may be attributed to the higher mass number of cadmium as compared to copper which is a significant factor in the mass transport through low permeable soils.[17]

Among all four types of studied electrode materials titanium and nickel demonstrated higher removal efficiency. However, highest removal efficiency was achieved in case of titanium electrode. Removal efficiency of titanium for copper and cadmium removal reached up to 89.4 and 62.7% respectively at the end of 100 hours run. It can be seen from figure 2, the removal efficiency for copper reached up to 74.5% within initial 20 hours of experimental run. Therefore, electrokinetic process with titanium electrode is capable of removing significant amount of metal from contaminated soil during initial 20 hours. As titanium electrode showed highest removal efficiency, remaining experimental runs were performed with titanium electrodes. The removal efficiency achieved in this study is higher than reported in literature.[18,19]

3.2 VARIATION OF SOIL pH WITH TIME

Variation in soil pH after 24 and 80 hours of experimental run at different points between electrodes is presented in figure 3. At the beginning of experimental run, pH of soil was determined and found to be 6.7. It can be seen from figure 3 that the soil pH in the vicinity of anode is decreasing with time while an increasing trend observed around cathode.

Curves of figure shows that acid front moves across the soil from anode towards cathode and a base front moves from cathode to anode. However, acid front moves faster than the base front. This could be due to the mobility of H^+ which exceeds the mobility of OH^-. This shift in the curve is also reported by some researchers and attributed to the direction of electro-osmotic flow which is generally towards the cathode.[20]

As the decontamination of soil depends on metal ion solubility with either a low or a high pH throughout the soil and avoiding the conditions resulting in precipitation. Thus control of pH is of major importance in soil decontamination.

3.3 EFFECT OF CURRENT DENSITY

During this study applied DC current was varied in a controlled manner to achieve current density ranging from 2.5

TABLE 3. GEOTECHNICAL AND PHYSICOCHEMICAL CHARACTERISTICS OF SOIL SAMPLE.

Parameter	Value
pH	6.7 ±0.5
Moisture content	15.7-32.5%
Buffering capacity	0.1±0.01 mM/g
Density	1.18±0.01 g/cm3
Porosity	48.7±1.2%
Sp. Gravity	2.47±0.2
Organic content	0.13±0.2%
Electrical conductivity	67.4±1.5µS/cm
Hydraulic conductivity	10^{-4} – 10^{-3} cm/sec
Liquid limit	25.8±0.5%
Plastic limit	16.2±0.8%
Plastic index	9.7±1.2

TABLE 4. RESULTS OF METAL ANALYSIS SHOWING BACKGROUND METAL CONCENTRATION PRESENT IN SOIL SAMPLE.

Metal	Concentration (µg/g)
Na	4165.2±8.5
K	1306.3±5.5
Mn	752.9±2.5
Fe	67.4±1.5
Zn	11.6±1.2
Cr	22.4±1.5
Ni	0.7±0.01
Pb	0.2±0.01
Cu	4975.6±8.5
Co	0.3±0.01
Cd	926.3±1.5
V	16.3±1.2

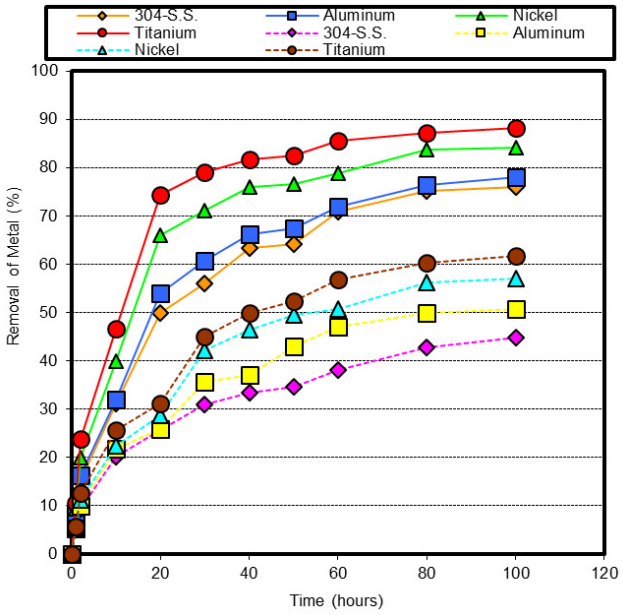

Fig 2. Temporal removal of copper and cadmium with different electrode materials (— continues lines) and cadmium (- - - dotted lines)

mA/cm^2 to 12.2 mA/cm^2 during four experimental runs (2.5 mA/cm^2, 5.5 mA/cm^2, 9.3 mA/cm^2 and 12.2 mA/cm^2). Temporal variation in the copper and cadmium removal when different current densities were applied are presented in Figure 4.

The metal removal found to be augmented with the increase in the applied current density however; significant increase in cadmium removal (24.3%) found as compared to copper (8.6%) when current density increased from 2.5 mA/cm^2 to 12.2 mA/cm^2. It was found that at current density of 9.3 mA/cm^2 copper concentration reached below 83 µg/g from 4975.6 µg/g at the end of experimental run (100 hours). However, further increase in applied current density does not improve the removal efficiency of process. Thus, current density 9.3 mA/cm^2 is a reasonable value for an effective copper and cadmium removal.

3.4 EFFECT OF SOIL TEMPERATURE

Effect of soil temperature on copper removal efficiency was investigated in two experimental runs by keeping temperature of soil matrix around 20°C and 35°C. An isotherm water-circulating bath (WCB Circulation water bath) was used and temperature of soil was maintained at desired value by keeping the electrochemical cell in the bath. Effect of soil temperature on the copper removal efficiency at 20°C and 35°C is shown in figure 5.

Removal efficiency of the process reached to about 75% when the temperature was 35°C as compared to it was about 22% when the temperature was 20°C therefore, an increase of about 52% in the process efficiency was observed. However, with time this difference diminished and at the

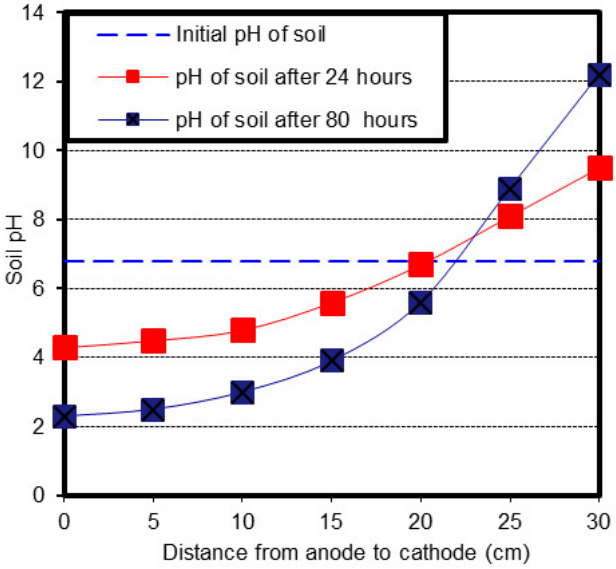

Fig 3. Variation of soil pH at different points between electrodes at 24 and 80 hours run

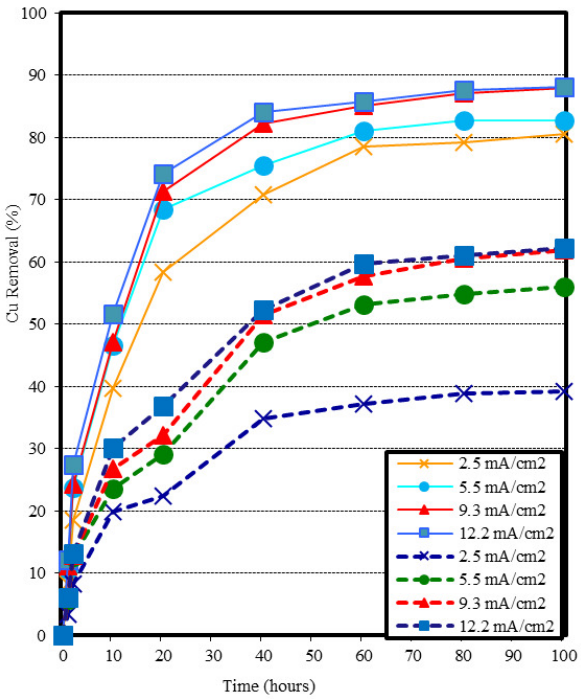

Fig 4. Effect of current density on the removal of copper and cadmium (— continues lines) and cadmium (- - - dotted lines)

end of 100 hours of run it reached to about 16%. Therefore, results suggested that the effect of soil temperature is very important during first few hours and it may significantly improve the removal efficiency of process. Therefore, further study is warranted to investigate the effects of soil drying which may result in soil shrinkage, cracks development and creation of irregular flow paths, which may ultimately cause termination of osmotic flow.

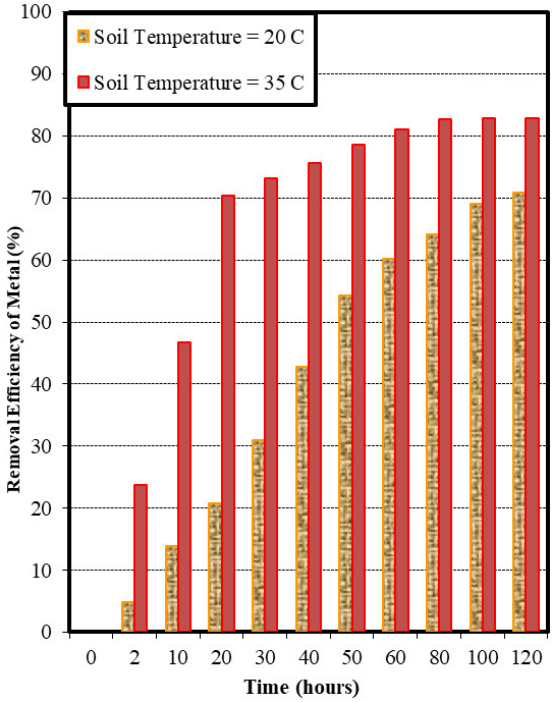

Fig. 5. Removal efficiency of EK for copper process at 20 C and 35 C

3.5 EFFECT OF INTER ELECTRODES SEPARATION

Inter-electrode separation is one of the important operating parameters of electrokinetic remediation method because the increase in the separation between electrodes will result in economical treatment and reduce the capital cost of treatment. However, it may result in reduction in the treatment efficiency of the process. Furthermore, increasing the inter-electrode separation will result in longer treatment time.[21]

Results presented in figure 6 show that the copper removal efficiency is increasing with the reduction in the inter-electrode separation. Maximum removal of copper achieved at separation of 15 cm. It was observed that by increasing the inter-electrode separation from 15 cm to 30 cm there is a decrease in removal efficiency from 89.5% to 68.7%. However, if separation decreased from 15 cm to 10 cm no improvement in removal efficiency was observed and at 5 cm separation trend shows a nonlinear behavior. The nonlinear behavior may be attributed to the transitions during buildup of double layer forces that are more effective near certain inter-electrode separation. Thus, copper removal efficiency is reduced beyond a separation of 15 cm.

3.6 EFFECT OF INITIAL SOIL MOISTURE CONTENT

Moisture content in the soil matrix could be one of the important parameters, which may enhance the electrokinetic process efficiency.[22]

Results presented in figure 7 show that with the increase in soil moisture content the cooper removal is increasing especially during initial 20 hours of run. An increase in moisture content from 45% to 73% (29% increase) enhance

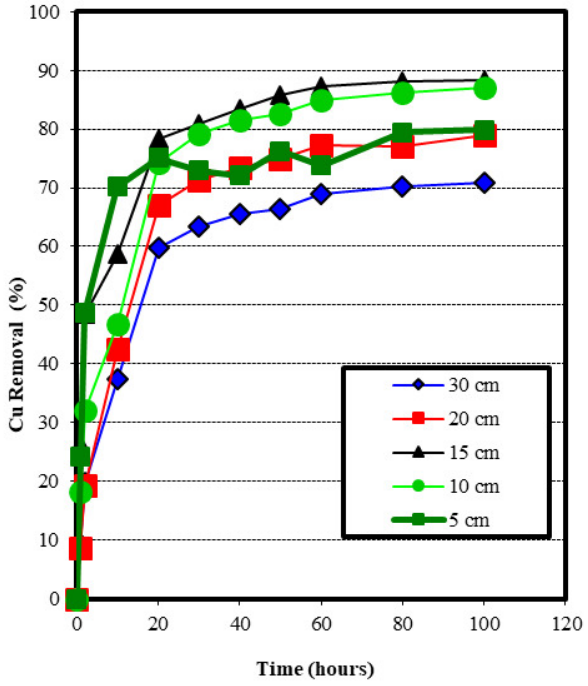

Fig. 6. Effect of inte-electrode separation on copper removal

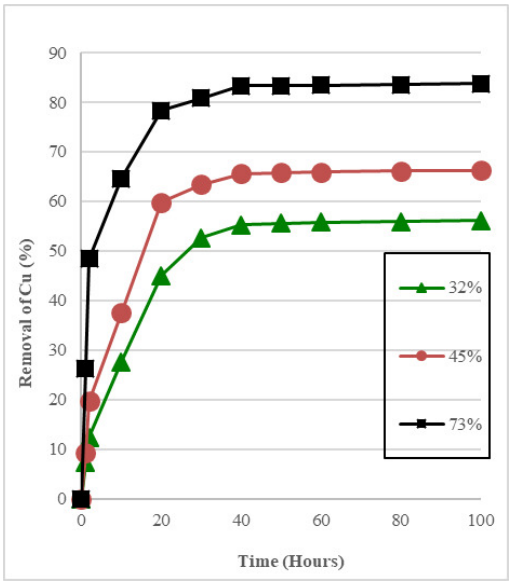

Fig. 7. Effect soil moisture content on copper removal

the cooper removal up to 33% (at 20 hour). However, removal of copper declined after 40 hours of run and became almost constant which may be attributed to the drying of pore space and cracks formation in the soil matrix.

Overall, it was observed that the initial moisture content affects the electrokinetic process but it does not significantly influence the overall migration and removal of copper, and the results indicate that the soil moisture content initially affect migration and removal of copper until there is sufficient moisture present in the soil matrix.

4. SUMMARY AND CONCLUSION

Electrokinetic treatment process has been used successfully at many sites such as Jaslovske Bohunice and Chernobyl nuclear power plant and many superfund sites for the removal of metals and other contaminants.[23-26] A detailed laboratory scale study was performed to investigate the efficiency of the process in order to remove copper and cadmium from soil samples. Influence of operational parameters such as electrode material, soil pH, current density, soil temperature, inter-electrode separation and soil moisture content on the treatment performance was investigated.

Following are the conclusions drawn on the basis of the results obtained from study.

- Titanium found to be electrochemically most stable material among tested materials and showed minimum dissolution during experimental run. At the end of 100 hours run removal efficiency of titanium for copper and cadmium found to be 89.4% and 62.7% respectively.
- Low pH found to be favorable for copper removal from soil because of the more desorption of sorbed metals from soil surface occurred in the presence of H_3O^+.
- Metal removal is positively related to the current density. Results show that current density of 9.3 mA/cm^2 is capable of bringing the copper concentration below 83 µg/g from 4975.6 µg/g within 100 hours of experimental run.
- Effect of soil temperature on the copper removal efficiency was monitored and found that during initial 20 hours run removal efficiency of process improved by 52% while soil temperature increased from 20 to 35°C. It seems that the increase in temperature enhanced the mobility of copper in the moist soil especially during first few hours.
- Rate of copper removal increased as the inter-electrode separation decreased. Maximum removal of copper achieved with a separation of 15 cm. A reduction of inter electrode separation from 30 to 15 cm may increase copper removal efficiency from 68.7% to 89.5%. But after an optimum separation electrostatic double layer formed due to the cations swarm near the soil particle surface which may dominate and suppressed the copper removal.
- A nonlinear behavior in removal efficiency observed at a separation of 5 cm and may be attributed to the transitions in overwhelming double layer forces becoming effective near certain inter-electrode separation.
- An increase of 29% moisture content improve the removal of copper up to 33% during initial 20 hours of run. However, it does not significantly enhance the overall migration of metal ions.

Application of electrokinetic process is quite tempting since it offers an elucidation to remediate sites that would otherwise be cost-prohibitive. Scale-up considerations has been evaluated by researchers and found better results with large scale devices using lesser energy and additives.[27,28] There is a great potential for remediation of actual contaminated sites. However, there still exists a need for well-planned and well-instrumented application of technology in the field. Site characteristics and inherent complexity with the electrokinetic process need to be investigated. Finally, optimization of operational parameters is imperative for each site to be decontaminated to obtain maximum achievable efficiency of the electrokinetic process.

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