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ELECTROKINETIC REMEDIATION OF CADMIUM CONTAMINATED SOIL IN FIELD CONDITION

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Abstract

Electrokinetic is an effective technique for remediation of soils contaminated with heavy metals and sediments. This technique has a higher capability than other prevalent methods, mainly in soils with low hydraulic conductivity. Electrokinetic removal of heavy metals from soils has been investigated in many studies, but most of them are laboratory studies and have not been studied in field, especially in soils with high amount of lime. In this study, the remediation of a clay loam soil contaminated with cadmium by the electrokinetic method was investigated at three depths of 0, 15 and 30 cm in four plots (4 m^2) under field conditions on a farm located outside of the Marand city, East Azarbayjan province, NW of Iran. The experiments were carried out by applying a voltage gradient of 1 V/cm for the three periods of two, four and six days. The contaminated soil had a high concentration of the heavy metal cadmium (24.6, 21.7 and 17.7 mg/kg at the three depths of 0, 15 and 30 cm, respectively). The results indicated that the removal efficiency of Cd from soil increased remarkably with increasing the duration of the electrical current application. The maximum removal efficiency of Cd for the three depths of 0, 15 and 30 cm was equal to 20.3%, 22.4% and 24.2%, respectively. The mean removal efficiency at the three depths of 0, 15 and 30 cm after six days of applying the electric current increased by 10.9, 9.3 and 10.9 units, respectively, relative to the two-day application. In addition, the pH changes in the soil showed an ascending trend from the anode (pH=5.5) to the cathode (pH=9.2) in the experimental plots.

Key words: cadmium, electrokinetic removal, field condition, heavy metals, soil

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1. Introduction

In recent years, the technology of electrokinetic remediation as an *in-situ* technique has received much attention, and many efforts have been made to apply it in new, innovative and cost-effective methods (Cameselle and Gouveia, 2018; Lee and Yang, 2000). Environmental researchers have given various names to this technology, including electrokinetic remediation, electro-reclamation, electro-restoration and electro-chemical decontamination. In general, electrokinetic requires the installation of electrodes below the surface of the contaminated area. After the electrodes are installed, a little electric potential is applied among the positive (anode) and the negative (cathode) electrodes. Due to the electrical potential between the two electrodes in the soil, different physio-chemical reactions take place and the contaminants are transported under different mechanisms in the soil and groundwater (Fu et al., 2017; Reddy and Chinthamreddy, 1999; Zhu et al., 2015). The applied electric current (or electric potential difference) results in electrolysis reactions in the vicinity of the electrodes, the acid-base distribution affected by chemical, electric and hydraulic potentials differences, absorption and desorption, deposition and dissolution reactions, pore fluid and ion transport, and precipitation (Acar et al.,

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1996; Gomes et al., 2012). During electrokinetic processes, the electrolysis of water occurs at the electrodes, production hydrogen (H^+) ions (solution with low pH) and oxygen gas at the anode electrode and hydroxyl (OH^-) ions (solution with high pH) and hydrogen gas at the cathode electrode, using Eqs. (1-2) (Popov et al., 2008; Reddy et al., 2004):

$$2H_2O \rightarrow 4H^+ + O_2\uparrow + 4e^-, E^\circ = -1.229 \quad \text{mV} \quad \text{(anode)} \tag{1}$$

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2\uparrow, \quad E^\circ = -0.828 \quad \text{mV} \quad \text{(cathode)} \tag{2}$$

The H^+ ions produced at the anode move across the soil towards the cathode by ion migration, diffusion and pore fluid advection and pore fluid flow. On the other hand, the reduction reaction at the cathode region dissociates water to form H_2 and OH^- during electrolytical dissociation. Therefore, it causes increase in the pH value beside the cathode. The OH^- and H^+ ions produced by the electrolytic dissociation move through the pore fluid within soil particles towards either the anode or the cathode electrodes (Pamukcu et al., 1992; Reed et al., 1995). Both electrolysis reactions and soil pH changes at the electrodes play a significant role in the electrokinetic remediation (Giannis et al., 2005). Also, by applying electric fields to the contaminated soil, the displacement of charged ions (such as heavy metal ions) occurs. The negatively charged ions move through the soil towards the anode and the positively charged ions moves towards the cathode (Chowdhury et al., 2017; Fu et al., 2017; Virkutyte et al., 2002). The heavy metal ions migrated towards the electrodes can be removed from the soil with various methods. However, according to numerous studies, a sharp increase in pH reduces the efficiency of this process in removing heavy metals, because heavy metals, once enter the alkaline environment (resulting from OH^- produced during the electrolysis process around the cathode), are adsorbed onto the soil particles, or precipitate in the form of hydroxide, etc. Acidic conditions can lead to desorption, dissolution, displacement of heavy metal ions, and increased efficiency of electrokinetic remediation (Chung and Kang, 1999; Zhou et al., 2005a). The control of soil pH using different methods is the usual choice to enhance the efficiency of heavy metals' removal in the electrokinetic process (Baek et al., 2009). Enhanced electrokinetic remediation by addition of enhancing agents such as inorganic acids (e.g., HNO_3, H_2SO_4 and HCl) (Cameselle and Pena, 2016; Villen-Guzman et al., 2014; Xu et al., 2021), organic acids (e.g., acetic acid, citric acid, and oxalic acid) (Cameselle and Pena, 2016; Villen-Guzman et al., 2014; Xu et al., 2021; Zhang et al., 2018), and complexing agents (e.g., EDTA) (Beyrami, 2021; Cameselle and Pena, 2016; Gu et al., 2018; Lee and Kim, 2010; Xu et al., 2021) are used to improve the dissolution of heavy metals to promote the removal efficiency. In many studies, the high efficiency of the electrokinetic method compared to other methods such as phytoremediation, bioremediation, etc. in the heavy metals removal has been mentioned (Chang et al., 2018; Lee and Yang, 2000; Reddy et al., 1999; Sruthy et al., 2014). However, it is imperative to examination its field efficiency in the removal of heavy metals. The aim of this study is to investigate the efficiency of the electrokinetic method in refining a clay loam soil contaminated with the heavy metal cadmium (Cd) under field conditions on a farm outside of Marand city, East Azarbayjan province, NW of Iran in 2018.

2. Material and methods

2.1. Soil properties and experimental treatments

The soil of a part of a farm outside of Marand city (with geographical coordinates of 38° 25.16′ 87″ north and 45° 47.30′ 30″ east) with an area of 4 m^2 (4 m × 1 m) as the experimental plot was contaminated with the Cd using a 50 mg/kg nitrate-cadmium solution. According to previous studies, due to the high efficiency of the electrokinetic method in removing heavy metals in near-saturated soils, the soil was constantly kept saturated by irrigation. A direct electric current supply was used to apply the current with a voltage gradient of 1 V/cm (Fu et al., 2017).

2.2. Description of the electrokinetic experiment

The soil was contaminated with the Cadmium nitrate ($Cd(NO_3)_2$) solution by irrigation. The contaminated soil then was alternately irrigated with pure water for 10 days (Nogueira et al., 2007; Sah and Chen, 1998) so that the balance for the absorption of Cd by soil particles can be reached. The amount of water used for irrigation was calculated according to the previous soil moisture and field capacity moisture (Shahmohammadi-Kalalagh et al., 2017) and the soil was contaminated to the depth of 30 cm. During the experiment, soil moisture was kept close to the *FC* to prevent an increase in path resistance for electrical current.

In this study, graphite electrodes were used for anode and cathode, because metal electrodes cause problems and errors in measuring and experiment due to metal corrosion in long-term (Heister et al., 2005; Turer and Genc, 2005). Numerous researchers have reported the effect of the electrodes material on the removal efficiency of heavy metals in their studies. Therefore, choosing the right electrode material is important, especially when using the electrokinetic technologies. Ideally, graphite electrodes are supposed to be inert and allow electron transfer without entering the reaction, but the electrode material usually bonds with the species in solution and oxide formation as well as adsorption of species occurs on the electrodes (Saichek and Reddy, 2003; Yan et al., 2018; Zhang et al., 2105).

Electric current (voltage gradient of 1 V/cm) was applied using a direct current supply and graphite rod electrodes (30 cm long and 3 cm diameter) in 1 m^2 plots. Fig. 1 shows a schematic of the electrokinetic setup used to remove heavy metals. A small pump next to the cathode electrode was used to remove the cathode solution containing the Cd deposit (Shahmohammadi-Kalalagh et al., 2017).

To determine the initial concentration of total Cd in the contaminated soil for electrokinetic remediation, sampling was performed at the specified points between the electrodes and at specified depths (0, 15 and 30 cm) at the begining of the test. Soil sampling was carry out to a depth of 31 cm using an auger. The measured Cd concentrations at the three depths 0-2, 14-16 and 29-31 cm were considered as the concentrations of Cd at the depths 0, 15 and 30 cm, respectively. Fig. 2 shows the sampling points for determining the concentration of Cd in the experimental plots. Voltage gradient (1 V/cm) was applied to the plots over a six-day period. Sampling was performed during the experiment to determine the total Cd concentration at different distances from the electrode and at different depths on the second, fourth, and sixth days of the onset of applying the electric current. At the end of the test, the total concentration of Cd in the samples was measured using atomic absorption spectrophotometry method (Sposito et al., 1982). Also, pH and EC of the plots were measured. The residual Cd concentration in the soil samples was obtained according to the method of Sposito et al. (1982), modified by Turer and Genc (2005), after extraction based on the following steps. First, 5 g of air-dried soil was passed through a 0.5 mm sieve. This amount of soil was poured into a 100 ml beaker. Then, 25 mL of 4 M HNO_3^- was slowly added and was placed on a water bath at a temperature of 80 °C for 16 hours.

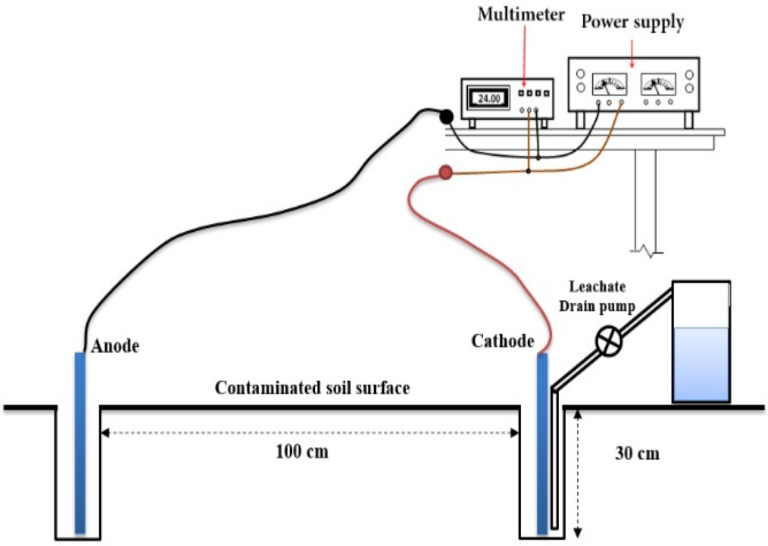

Fig. 1. Schematic of electrokinetic Set-up in the field

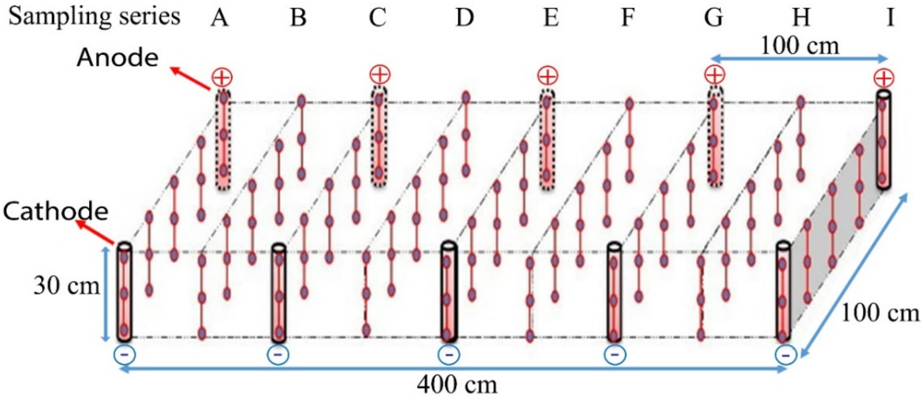

Fig. 2. Sampling series and points in the experimental plots
(the series A, C, E, G and I correspond to the position of the electrodes)

Table 1. The soil properties and cadmium concentrations

Parameters	*pH*	*OM (%)*	*CEC (cmolc/ kg)*	*Clay (%)*	*ECC (%)*	*Depth (cm)*	*Cd metals in three depths (mg/kg)*
Content	7.8	1.86	21.2	30	13	0	24.6

After that, Cd concentration in the filtered solution was measured with an atomic absorption spectrophotometer. Finally, the electrokinetic removal efficiency was calculated using the initial concentration of Cd in the contaminated soil and the obtained final concentration of Cd. A digital multimeter was used in the electrokinetic setup to measure the electrical properties (potential difference and current intensity) during the experiment. The electric potential difference between the cathode and anode electrodes as well as the electrical current intensity was measured every day. At the end of the electrokinetic remediation experiment, the soil pH was measured in thesaturated soil-paste extract

3. Results and discussion

The soil properties are shown in Table 1. The soil texture was clay loam. Due to the concentration of Cd added to it at various depths, the soil had a concentration higher than the critical range and was highly-contaminated.

3.1. Changes in soil pH during process development

Fig. 3 shows the changes in the soil pH in the plots at relative distances from the anode in the five series of samplings at the location of the electrodes after applying the electric current. According to the Figure, the soil pH on the anode side had the lowest value. It then increased rapidly and reached the initial value. Then a constant trend was observed until it increased rapidly again in the vicinity of the cathode. This increase and decrease in the soil pH was due to the high current intensity and, consequently, the increase in the production intensity of proton and hydroxyl ions in the anode and cathode caused by the electrolysis of soil water beside the electrodes, respectively (Cherifi et al., 2016; Zhu et al., 2016). Due to the distance from the source of hydroxyl and proton ions caused by the electrolysis of water, there was no considerable increase or decrease in the soil pH at the beginning and end of sampling series between the electrodes (B, D, F and H). Reddy et al. (2002) and Yuan and Chiang (2008) have attributed these changes in the soil pH to the electrolysis of water. In fact, the migration of the H^+ and OH^- ions caused by the electrolysis of water to the opposite-charged electrodes results in acidic and alkaline fronts on the anode and cathode sides, respectively. This increase and decrease have also been reported by Li et al. (2010), Kim et al. (2011), Ryu et al. (2010) and Yuan et al. (2017). Also, Giannis et al. (2008) have stated that the changes in soil pH during electrokinetic remediation is effective in removing heavy metals.

3.2. Changes in the electrical current intensity

Fig. 4 shows the changes in the current intensity during the experiment in the five electrodes series. At the beginning of the test, due to the generation of H^+ on the anode side by electrolysis of water, and the dissolution of ions, the highest current intensity was observed. Other reasons for the high value of current intensity were the development of the acid front in the soil column towards the cathode, and the increase of soluble ions in the soil. However, as time elapsed and the ions moved to the opposite-charged electrode by ion migration phenomenon (ion discharge), the electric conductivity of the path decreased (increases the resistance) and the current intensity decreased (Xu et al., 2017; Yuan et al., 2017).

Some authors (Al-Hamdan and Reddy, 2008; Cameselle et al., 2020; Guedes et al., 2019) described the increase in electrical current intensity in the early hours of the experiment to the production of H^+ ions by electrolysis and the transferring of these ions with electromigration to the cathode, which caused most contaminants by creating low pH in ion forms. Another reason for the decrease in the current intensity over time was the transport of soil moisture from the anode to the cathode by the electro-osmosis phenomenon, which increased the resistance of the path on the anode side. The initial soil water content affects the current intensity in the soil. As the soil water content increases, the current intensity increases. Increasing the soil water content not only increases the hydration of ions and the amount of free ions in the soil solution, but also increases the diffuse double layer thickness (Reddy et al., 2002).

Since in the present study it was tried to keep the soil saturated with daily irrigation, so the main factor in decreasing the electric current intensity was the ion migration phenomenon. The current intensity decreased gradually in all the five series and reached, on average, from about 470 mA at the start of the test to about 15 mA at the end of the sixth day. The changes in the electric current curves have also been reported (Al-Hamdan and Reddy, 2008; Altaee et al., 2008; Beyrami, 2021; Cameselle and Pena, 2016; Yuan and Chiang, 2008; Zhu et al., 2015).

3.3. Residual Cd concentration in soil

Table 2 gives the mean percentage values of the removal of Cd at different depths after two, four, and six days of electrokinetic remediation. According to this table, the efficiency of the removal of Cd increased with increasing the duration of applying the electric current in all the series, though this trend became descending gradually.

By applying the electric current for four days, the mean removal efficiency at the three depths of 0, 15 and 30 cm increased by 5.3, 4.8 and 4.9 units, respectively, relative to the two-day application. However, after six days of applying the electric current, the removal efficiency increased by 4.1, 3.1 and 4.1 units relative to the four-day application at the aforementioned depths. The reason for these changes can be attributed to the decrease in the current intensity with the increase in the duration of the electrokinetic decontamination, which is also shown in Fig. 4.

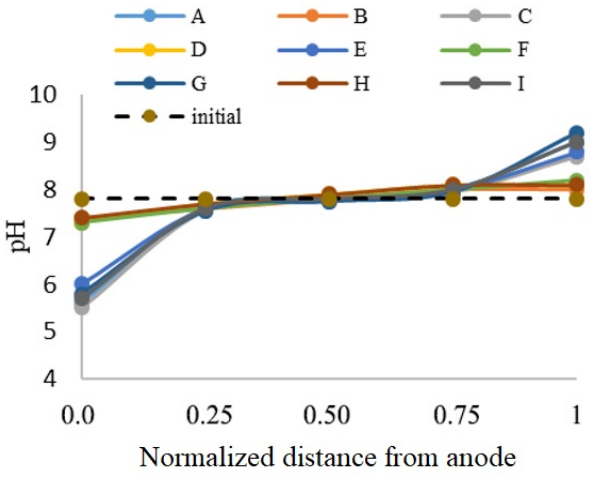

Fig. 3. The pH distribution in the soil after electrokinetic treatment

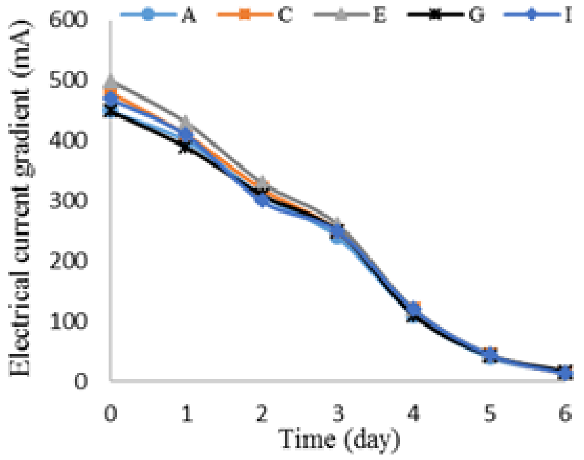

Fig. 4. The changes in the electrical current intensity during the experiment at the electrode series A, C, E, G and I

Due to the alkaline conditions on the cathode side, followed by the immobilization of cadmium and other ions, the intensity of the electric current decreased. The alkaline condition (high pH) in the cathode and part of the soil near the cathode as well as increasing the electrical resistance in the soil solution was responsible for reducing the intensity of the electric current. The increase of electrical resistance matched the decrease of the current intensity and decreasing in the Cd removal (Cameselle et al., 2020). Kim et al. (2011), Li et al. (2010) and Reddy et al. (2002) have found similar results in their studies.

Figures 5 and 6 show the residual Cd concentrations in the experimental plots in the A and B sampling series, respectively, for a two-, four-, and six-day time duration. Since the trend of the changes in Cd concentration was approximately similar in the series at the location of the electrodes, only the curves of A are shown in Fig. 5. Also, for the series amongst the electrodes, only the B series curves are shown in Fig. 6. According to these Figures, different initial concentrations were observed at different depths. This may be due to the high adsorption of Cd in the top layers when adding a Cd solution to contaminate the soil. The adsorption capacity and mobility of heavy metals in the soil have an important effect on their removal efficiency. In this experiment, less cadmium was removed due to higher adsorption capacity and immobility of cadmium. In these Figures, the distance between the initial and residual concentration curves represents the amount of the removal of Cd. The total cadmium removal amount from soil increased with increasing the duration of the electric current application, but the amount of increase in removal efficiency decreased gradually.

The reason is due to factors affecting the reduction of electric current intensity. This indicates that the operation for a long time increased the power consumption with no increment in metal removal (Cameselle et al., 2020). Other researchers, such as Yang et al., (2020), reported successful results in the removal of heavy metals.

In the vicinity of the anode the highest amount of removal occurred, while in the vicinity of the cathode Cd concentration increased. Decreasing the soil pH on the anode side due to the H^+ produced by electrolysis can increase the desorption of heavy metals (Xu et al., 2017) and, conversely, increasing the soil pH caused by OH^- can precipitate heavy metals (Giannis et al., 2008; Villen-Guzman et al., 2015). Cherifi et al. (2016) and Zhou et al. (2005b) have obtained similar results regarding the effect of the pH changes in the proximity of cathode and anode on the heavy metals removal.

Table 2. The mean values of the removal of Cd at different time duration at 3 depths

Time duration (day)	*Sampling depth (cm)*	*Mean Cd removal efficiency (%)*
2	0	9.4
	15	13.1
	30	13.3
4	0	14.7
	15	17.9
	30	18.2
6	0	20.3
	15	22.4
	30	24.2

As can be seen in Figs. 5 and 6, there is a significant difference between the curves of the removal of heavy metals from the series at the location of the electrodes (A) and the series located between the electrodes (B). The total values of the removal efficiency in these series are also different. In the series between the electrodes (B) due to the lack of H^+ production (high pH) at the beginning of the plot, the removal efficiency of Cd is not as high as its value in the series at the location of the electrodes. In fact, the highest amount of removal in these series (B) is related to the intermediate points of the plot that a lower pH was observed. In other words, there is a clear connection between the pH profile and the removal efficiencies. Therefore, in the series between the electrodes the removal efficiency of Cd in the anode side is not as high as its value in the series at the location of the electrodes. The changes in the removal of heavy metals at various distances from the electrodes have also been stated by Ryu et al. (2010) and Kim et al. (2011). The next important matter, which can be seen in Figs. 5 and 6, is the increase in the heavy metals removal due to the increase in the time duration of the electric current application.

Also, the efficiency of pollutant removal using the electrokinetic remediation depends on many factors. Numerous researchers in field and laboratory studies have stated that the variables affecting the removal of heavy metals and other contaminants from the soil include as follows: the chemical processes in electrodes (Giannis et al., 2005), the amount of soil moisture (Shin et al., 2017), soil structure (Virkutyte et al., 2002), soil pH and pH slope (Zhou et al., 2005a), type and concentration of ions in the soil solution (Virkutyte et al., 2002), the applied electrical current intensity and voltage gradient (Cameselle et al., 2020), the type of electrolyte solution used (Yan et al., 2018) and other environmental conditions.

Decreasing soil moisture reduces the cross-sectional area of the path as well as increases the path tortuosity for the ion movement; so by increasing the path resistance, it reduces the removal efficiency of heavy metals (Shin et al., 2017). The pH of the catholyte and anolyte reservoirs might affect the pH of the soil, which ultimately affects the removal efficiency of heavy metals (Zhou et al., 2005a). The intensity of ion transfer was higher in the high voltage gradient and high electrical current intensity. Cameselle et al. (2020) described that the increase in electric current intensity in the early hours of the experiment was due to the production of H^+ ions by electrolysis and their transfer to the cathode with electromigration, which caused most contaminants to be in ion forms due to low pH.

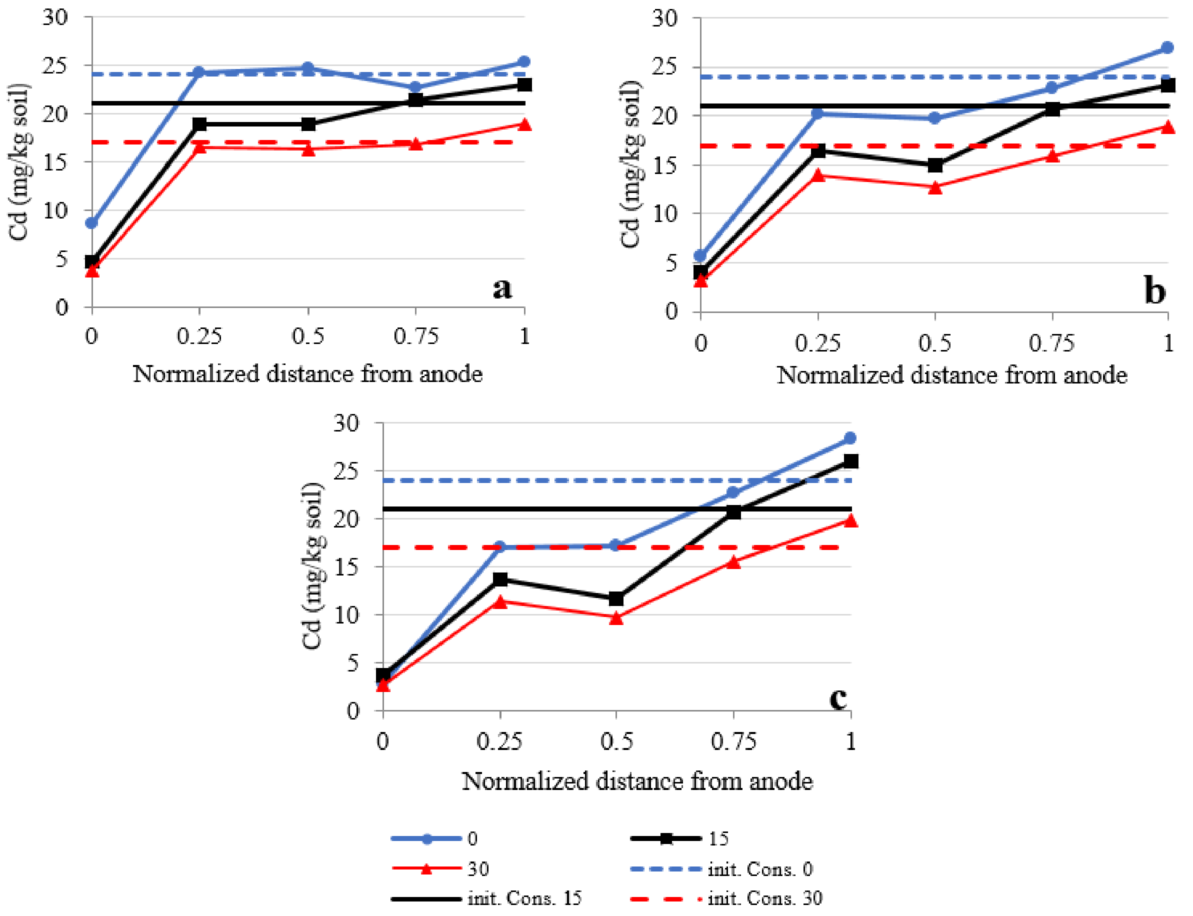

Fig. 5. The distribution of cadmium in the soil plot (the sampling series A) after 2 (a), 4 (b) and 6 (c) days of electrokinetic remediation

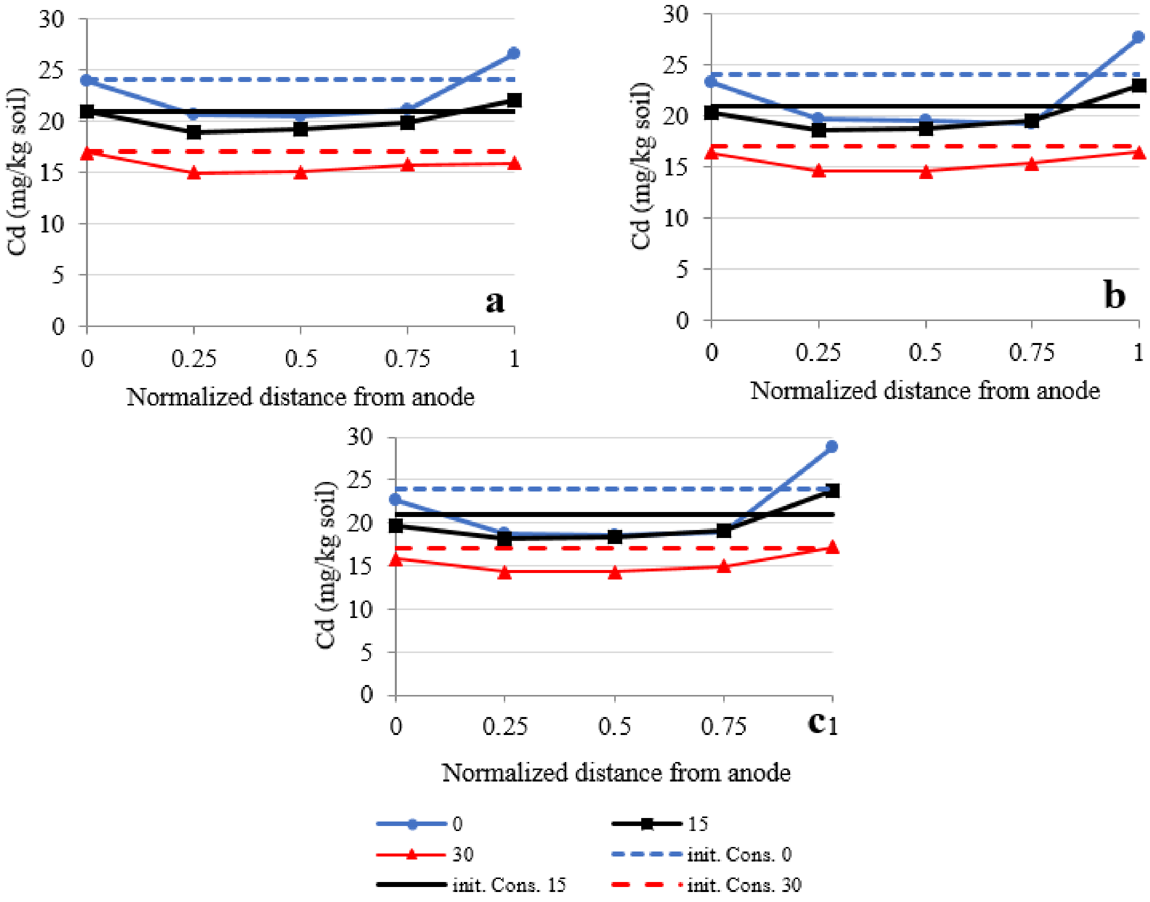

Fig. 6. The distribution of cadmium in the soil plot (the sampling series B) after 2 (a), 4 (b) and 6 (c) days of electrokinetic remediatiom

The high amount of equivalent calcium carbonate in the soil (13%) can be another reason for the decrease in the cadmium removal efficiency of this soil. Altin and Degirmenci (2005) expressed the effects of substances such as gypsum and lime on reducing the effectiveness of electrokinetic remediation of heavy metals.

4. Conclusions

In the construction stage, the recycled boilers using relatively low amount of virgin materials showed much lower environmental impacts than the new one. In the operation stage, electrokinetic remediation technique requires much less time for removal particularly in the soil with lower permeability comparing to the conventional leaching method.

The originality of the work is that the treatment process has been applied in a field and removal efficiency was analyzed depending on soil depth. Results indicated that the electrokinetic remediation technique has a proper efficiency in removing heavy metals in field. In this research, the electrokinetic technique resulted in 17.3% Cd removal from the soil. The maximum removal efficiency of Cd for the three depths of 0, 15 and 30 cm was equal to 20.3%, 22.4% and 24.2%, respectively. In this experiment, the highest amount of removal occurred in the vicinity of the anode, and in the vicinity of the cathode, Cd concentration increased due to cadmium immobilization for the alkaline environment in the cathode side with high pH. Furthermore, increasing the duration of applying the electric current increased Cd removal from the soil, but this process had a descending trend due to the decrease in the intensity of the electric current caused by the increase in the resistance of the path.

The electrokinetic remediation method was more effective at lower depths than upper layers because of the higher water content in lower depths. High lime content (13%) and high absorption power of the examined soil appear prevented removal efficiency. The use of acidic materials can increase the removal of heavy metals due to the increased solubility of heavy metals compounds. Also, results indicated that increasing the duration time would enhance the Heavy metal removal efficiency, but for a long time increased the power consumption with no increment in metal removal.

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