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USE OF CLEAN ENERGY TO REMEDIATE CONTAMINATED SOIL

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Abstract: The aim of this laboratory study is to investigate the feasibility of using solar energy in combination with electro-kinetic remediation without enhancement conditions to remove cadmium from artificially contaminated soil. Two tests were performed to investigate the effect of applying continuous and constant voltage from DC power supply, and non-continuous and constant DC voltage from solar panel. The experiment was carried out in Baghdad city. The DC volte was generated by two Solar panel each one has a maximum voltage of (17 V). The experiments were commented in the mid of May 2016 (average length of daytime 14 h 3 min). The results showed that the use of solar energy in the present study to remove cadmium from contaminated soil is not successful because of the low removal efficiencies and the need for more time to substitute the stoppage of night times.

Keywords: *Soil pollution, Cadmium removal, Electro-kinetic, Solar panel, Soil remediation*.

إستعمال الطاقة النظيفة في معالجة التربة الملوثة

الخلاصة: تهدف هذه الدراسة الى اختبار استخدام الطاقة الشمسية مع الطريقة الكهروحركية بدون استخدام ظروف تحسين للطريقة مختبريا في معالجة التربة الملوثة صناعيا بالكادميوم. تم اجراء تجربتين لاختبار تأثير تطبيق تيار ثابت ومستمر من مجهز للتيار المستمر, وتيار مستمر غير ثابت مجهز بواسطة لوح من الخلايا الشمسية. تم اجراء التجارب في مدينة بغداد بانتاج تيار مستمر من خلال زوج من الخلايا الشمسية كل واحدة لها فولتية قصوى مقدارها (17 فولت). تم اجراء التجارب في وسط شهر ايار عام 2016 بمعدل طول نهار 14 ساعة و3 دقائق. اظهرت النتائج ان استخدام الطاقة الشمسية في الدراسة الحالية لازالة الكادميوم من التربة الملوثة غير ناجحه بسبب قلة كفائة الازالة وحاجتها الى وقت اطول لتعويض التوقف اثناء الليل.

1. Introduction

In many areas around the world, soil has been severely contaminated due to inappropriate waste disposal practices, polluting industrial activities, land application of pesticides, etc. The contaminated soil in turn becomes a pollution source that threatens the environment by releasing toxic substances to the surroundings. This threat poses a challenge to scientists to develop effective and economic soil remediation technologies to inhibit the release of pollutants from the contaminated soils, extract the contaminants from soils, and destroy the contaminants in the soils [1, 2].

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Heavy metals in soil can be present as soluble compounds like ions and metal complexes or in exchangeable forms. These metals are generally rendered immobile by associating with different soil fractions and compartments such as carbonate, oxide, hydroxide, organic matter and residual materials [3]. The presence of heavy metal ions in the environment has been a matter of major concern due to their toxicity to human life. Unlike organic pollutants, the majority of which are susceptible to biological degradation, heavy metal ions will not degrade in to harmless end products [4].

The solubility of metals in soil is influenced by the chemistry of the soil and groundwater. Factors such as pH, Redox potential (Eh), ion exchange capacity, and complexation/chelation with organic matter directly affect metal solubility [5]. The toxicity of heavy metals can be listed in order of decreasing toxicity as Hg > Cd > Cu > Zn > Ni > Pb > Cr >Al >Co, although this is only approximate as the vulnerability of species to individual metals varies. Toxicity also varies according to environmental conditions that control the chemical speciation of the metals [6]. Accumulation of heavy metal in soils is poisonous to humans and animals. Cadmium in soils comes from natural and anthropogenic sources. Cadmium is relatively mobile in surface water and ground-water systems and exists primarily as hydrated ions or as complexes with humic acids and other organic ligands.

Under acidic conditions, cadmium may also form complexes with chloride and sulfate. Cadmium is removed from natural waters by precipitation and sorption to mineral surfaces, especially oxide minerals at higher pH values (pH >6). Removal by these mechanisms increases as pH increases. Sorption is also influenced by the cation exchange capacity (CEC) of clays, carbonate minerals, and organic matter present in soils and sediments. Under reducing conditions, precipitation as CdS controls the mobility of cadmium [5].

One of the emerging technologies to clean up heavy metal contaminated soils is the electro-kinetic remediation technique, which has been proven to be a very effective tool to remediate contaminated low hydraulic permeability fine grained soils [7-12]. Electro-kinetic remediation applies an electric field across a volume of soil to transport contaminants to a localized area where they can be effectively extracted [13-17]. The object of the present study is to investigate the use of solar energy in combination with electro-kinetic remediation without enhanced conditions.

2. Experimental Details

2.1 Materials

Cadmium was chosen to represent heavy metal contaminants. Cadmium was used in the form of $Cd(NO_3)_2.2H_2O$ (manufactured by E. MERCK, Denmark). To prepare a soil sample with cadmium concentration equal to 1500 mg/kg and initial moisture content equal to 30% by weight, 12.116 g of $Cd(NO_3)_2.2H_2O$ was dissolve in one liter of distilled water and 300 ml of this solution is added to 1 kg of dry soil. The soil was cleaned and sieved to achieve satisfactory uniformity. Table 1 summarizes the composition and properties of soil

Table 1. Composition and properties of the soil sample.

Property	Value
Particle size distribution (ASTM D 422)	
Sand (%)	59
Silt (%)	30
Clay (%)	11
Soil texture	Sandy loam
Hydraulic conductivity (cm s^{-1})	2.4×10^{-4}
Cation exchange capacity (meq /100 g)	6.12
pH	8.6
Organic content (%)	0.17
Electrical conductivity EC (µS/cm)	1280
Porosity (n)	0.43

2.2 Electro-kinetic Test Setup

Fig. 1 shows the schematic diagram of the electro-kinetic test setup used in this study. The system consists of an electro-kinetic cell, two electrode compartments, a power supply, and a multi-meter. The glass electro-kinetic cell has an inner dimension of 35 cm x 6 cm x 10 cm high. The actual length of the soil specimen in this cell equal to 20 cm, each electrode compartment consisted of a valve to control the flow in the cell. Perforated plastic plates and filter paper were used to separate the soil at two ends of soil. Anode and cathode electrode was graphite rode of 2 cm diameter. DC power source was used to generate a constant voltage to the electrodes, and DC voltage from solar panel used to investigate the feasibility of solar energy to remove cadmium from artificially contaminated soil. Multi-meter was used to monitor the voltage and measure the current flow through the soil sample during the test.

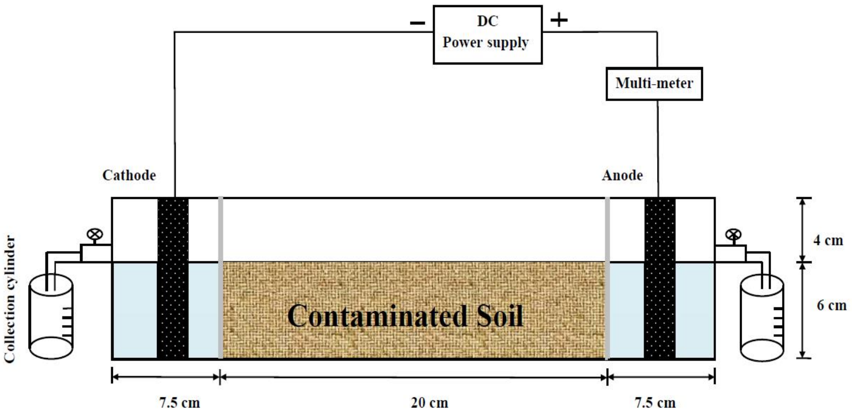

Figure 1. Schematic representation of electro-kinetic cell.

2.3 Experimental Design

Two tests were performed to investigate the effect of apply continuous and constant voltage from DC power supply, and non-continuous and constant DC voltage from solar panel. Purging solutions where used on the two electrode compartments to increase the remedial efficiency, nitric acid HNO_3 mixed with distilled water to reach to (pH= 4) and used as a purging solution. For the first test (EK-1), 24V potential was applied in the electro-kinetic cell from DC power supply, and for the second test (EK-2), two Solar panel each one have maximum voltage of 17 V was applied in the electro-kinetic cell and the other conditions are kept as in (EK-1).

2.4 Testing Procedure

The soil samples were artificially contaminated with cadmium ions in two electro-kinetic tests. The required amount of the $Cd(NO_3)_2.2H_2O$ that would yield the desired concentration was weighed and then dissolved individually in distilled water. The contaminant solution was then added to 1kg of the dry soil and mixed thoroughly with a plastic spatula in polyethylene container. A total of 300 ml of distilled water (30% moisture content) was used to represent typical field moisture conditions. The contaminated soil was then placed in the electro-kinetic cell in layers and compacted uniformly using wooden rod. The exact weight of the soil used in the cell was determined and the soil was equilibrated for 24 hrs. The filter papers were inserted at the ends of the soil sample.

The electrode compartments were filled with purging solutions (distilled water with pH=4). The elevation of water level in the electrode compartment was kept constant to prevent a hydraulic gradient from forming across the specimen. The electro-kinetic cell was then connected to the power supply and a constant DC voltage gradient was applied to the system. The electric current across the soil sample as well as the water flow, pH, and electrical conductivity in both the anode and cathode compartments were measured at different time periods throughout the duration of the experiment.

At the end of each test, the soil specimen was extruded from the cell. The soil specimen was sectioned into five parts and each part was weighed and subsequently preserved in a glass container. From each soil section, 5 g of dry soil was taken and mixed with 12.5 ml distilled water. The mixture was shaken thoroughly by hand for several minutes and the solids were then allowed to settle for one hour after that pH of the soil was measured [18].

2.5 Chemical Analysis

Cadmium in different soil sections were extracted by performed acid digestion in accordance with the Danish Standard DS 259:2003 (30 min at 200 kPa (120 $^{\circ}$C)) by adding 20 ml 1:1 HNO_3 to 1g dry matter and treating the sample in autoclave. The liquid was separated from the solid particles by vacuum through a 0.45 µm filter and diluted to 100 ml. The cadmium concentration was determined by atomic absorption spectrophotometer [18].

3. Results and discussion

3.1 Experiments one (EK-1).

The experiment (EK-1) was conducted as a baseline for the second experiments, it was run with nitric acid HNO_3 mixed with distilled water to reach to (pH= 4) was used as a purging solution in both the anode and cathode compartments. 24 DC volte from DC power supply was applied, the duration of (EK-1) was four days. Fig. 2 show the variation of pH of the fluid at anode (anolyte) and cathode (catholyte) recorded during the experiment. Due to the applied electric potential, the electrolysis of water produced H^+ at the anode and OH^- at the cathode. As a result, pH of anolyte (anode solution)

Decreased, while the pH of catholyte (cathode solution) increased. Compared with the initial pH values, pH changed significantly in the anode and cathode compartments in the test. The results show that considerable electrolysis reactions occurred at electrodes in the test, and the measured pH values are consistent with the values reported in the literature [7]. Due to high pH at cathode compartment the ionic species precipitated as hydroxides forms and limit the remediation efficiency. For this reason the cathode compartment should be lowered pH to avoid the generation of hydroxide and its transport into the soil [19]. The initial electrical conductivity (EC) value of the purging solution used in both the anode and cathode compartments equals to 142 μS/cm (Fig. 3). The EC values of the anolyte increased and finally decrease. However, the EC values of the catholyte increased to 1.78 mS/cm. duo to migrating of negative ionic species to the cathode. Relatively high EC values at the catholyte indicated higher ionic concentration in cathode electrode solution.

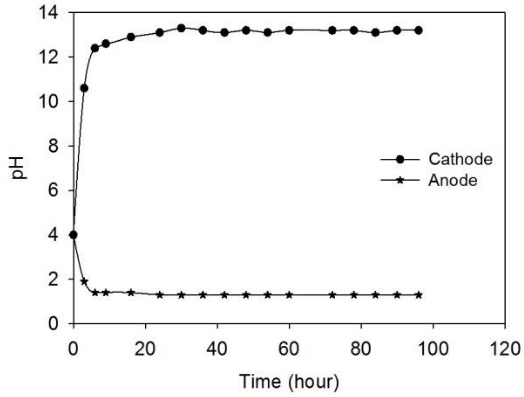

Figure 2. pH variation in the anode and cathode compartments versus the time.

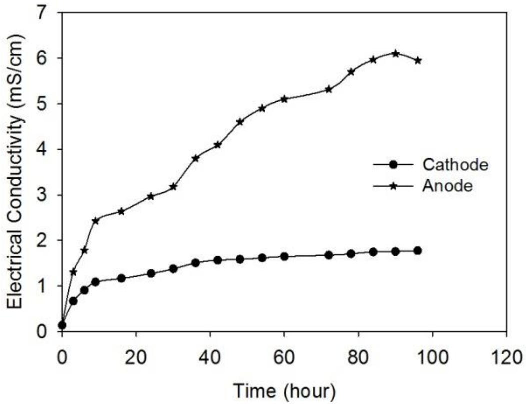

Figure 3. EC variation in the anode and cathode compartments versus the time.

Fig. 4 shows the profile of electric current during the tests the current initially was increases to 24.8 mA after 24 hour, current was decreased and finally stabilized to 8.8-8.7 mA. Direction of fluid flow was from the anode to the cathode reservoirs (zeta potential ζ <0) [20].

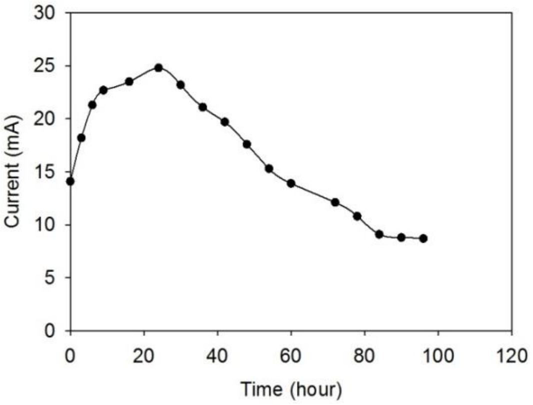

Figure 4. Electric current variation versus the time.

Fig. 5 shows the plot for cumulative volume of effluent versus time. The effluent flow increased with time and finally stopped because of the clogging of soil pores that happen from deposition in the soil of compounds such as metal hydroxides which may be formed by reaction between hydroxyl ions near the cathode and heavy metal contaminants and eventual cessation of flow [20]. Furthermore, the calcium precipitation close to the cathode can also clog the soil pores; restraining further transport of heavy metal ions for removal [21].

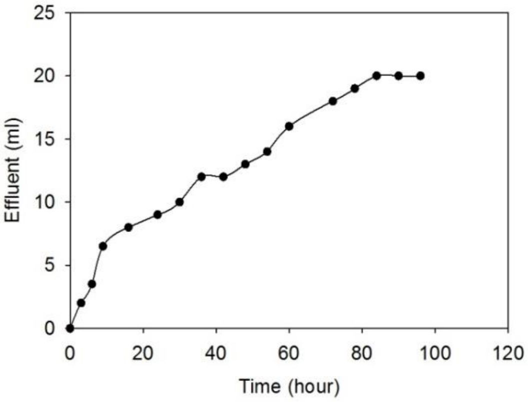

Figure 5. Electro-osmotic flow variation versus the time.

Fig. 6 shows the Cd concentration vs. normal distance from the anode with respect to starting values in the soil (1500 mg kg). As shown, the Cd migrated from the anode and accumulated toward the cathode and the removal efficiency was 20.8 %, which was calculated based on the residual contaminant masses in the soil.

Fig. 7 shows the pH changes vs. normal distance from the anode of the treated soil. The generated hydrogen and hydroxyl ions are transported through specimen in opposite directions by electrical migration, diffusion and advection. The advance of base front is slower than the advance of the acid front because of the counteracting electro-osmotic flow and also because the ionic mobility of H^+ is about 1.76 times that of OH^- [11].

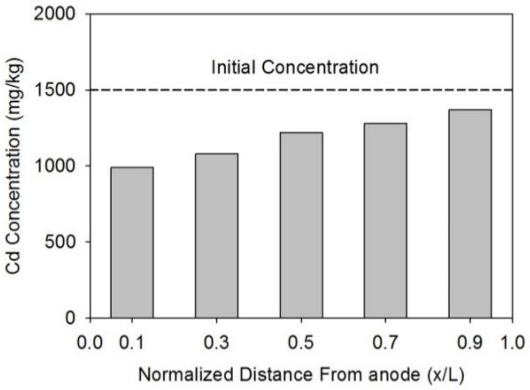

Figure 6. Cadmium concentration in soil after (EK-1).

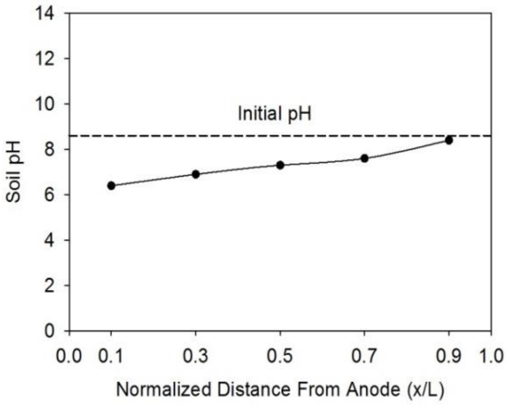

Figure 7. pH in soil after (EK-1).

When the acid and base fronts meet, water is formed resulting in a sharp change in pH, which affects the solubility of contaminants and their adsorption to the soil. An added complication is the effect of pH on the soil minerals. Soil constituents, including clay minerals, may dissolve because of the changing composition of the pore solution. An increase in the concentrations of certain ions of, e.g., Mg, Al, and Fe is often seen as a result of increased acidity and others of, e.g., Si and Al with increased alkalinity. Al ions are commonly detected in pore solutions of soils which have undergone electro remediation. Changes in the pH profiles in the soil had a great influence on contaminant migration. Tests conducted in the present study showed that pH has a significant impact on Cd migration in the soil. Due to the low pH at the anode, the Cd concentration decreased. The high pH indicated that some OH^- ions were reaching the soil and possibly interfering with the mobility of cadmium ions. The high pH environment was favorable to make $Cd(OH)_2$ which precipitate near the cathode sections and decrease the soil conductivity. According to this fact, the higher Cd concentrations were found near the cathode sections [20]. A certain amount was found in the anode compartment. It was also suggested that cadmium found in the anode compartment may be due to its capability of forming complexes with different legends present in soils [22, 10]. The drawbacks of electro-kinetic process were because of the amount of calcite or carbonate ($CaCO_3$). The presence of carbonate in soils causes an increase in buffering capacity; the alkaline pH of soil rich in carbonate does not significantly vary in electro-kinetic contaminant remediation. Consequently, heavy metal contamination is retained in precipitated hydroxide/carbonate forms. As a result, the efficiency of electro-kinetic remediation decreases [21].

3.2 Experiments two (EK-2).

The experiment (EK-2) was conducted at the same condition of experiment (EK-1) but by applying DC volte from two Solar panels, each one has a maximum voltage of 17 V. The latitude and longitude of Baghdad city (Iraq) where the experiments were carried out are 33.33333 and 44.43333, respectively.

The solar panel (Fig. 8) was put at an angle of 50º to the horizontal. The experiments were commented in the mid of May 2016 with average length of daytime of 14 h 3 min. Fig. 9 shows the variation of pH of the fluid at anode (anolyte) and cathode (catholyte) recorded during the experiment. Compared to the initial pH values, pH changed in the anode and cathode compartments in the test. The results show that electrolysis reactions occurred at electrodes in the test.

Because the applied of DC volts from solar panel during the day time (14 h 3 min), pH was decreased in anode compartment and increased at cathode compartment, however at the night time pH was slightly increased in anode compartment and decreased at cathode compartment [7]. That can be explain by the DC voltage from solar panel at the night time was equal to zero.

Figure 8. Solar panel used in (EK-2).

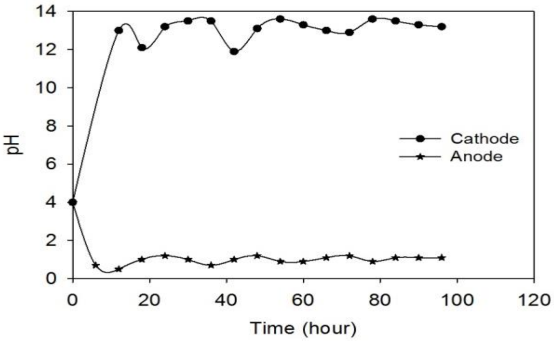

Figure 9. pH variation in the anode and cathode compartments versus the time.

Fig. 10 shows the variation of electrical conductivity in the anode and cathode compartments versus the time. The EC values of the anolyte and catholyte increased and decreased depending on DC volts of solar panel at day time and night time, duo to migrating of negative ionic species to the cathode. Relatively high EC values at the catholyte indicated higher ionic concentration in cathode electrode solution.

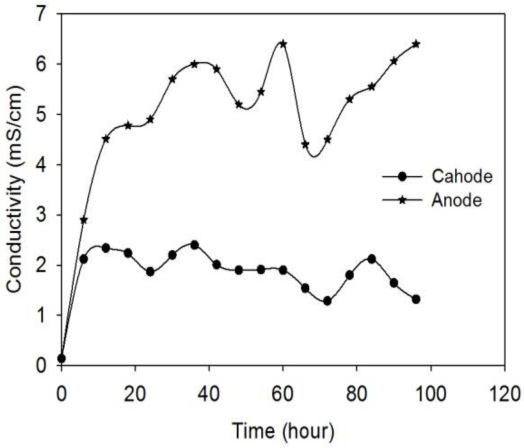

Figure 10. EC variation in the anode and cathode compartments versus the time.

Electric current during the tests shows in Fig. 11 the profile of the current initially was increases to 35.5 mA after 6 hour. Electric current was decreasing and increasing depending on DC volts of solar panel at day time and night time. Current finally reached to 21.4 mA.

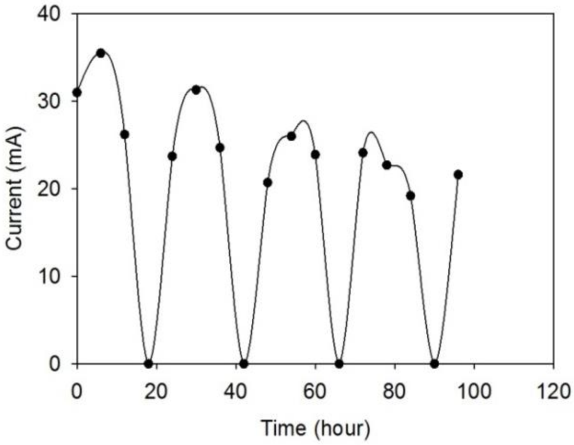

Figure 11. Electric current variation versus the time.

Fig. 12 shows the generated voltage of solar panel at day time and night time. The peak voltage during at daytime was 27 to 36 V, while 0 V was reported during the night. The peak voltage was reported after about 6 hour from test beginning (the test was start at 6 am). Fig. 13 shows the plot of cumulative volume of effluent versus time. Direction of fluid flow was from the anode to the cathode reservoirs; the peak effluent has reached to 28 ml and stopped.

Fig. 14 shows the Cd concentration versus normal distance from the anode with respect to the starting values in soil (1500 mg kg). As shown, the Cd migrated from the anode and accumulated toward the cathode. The removal efficiencies were 4.53 %.

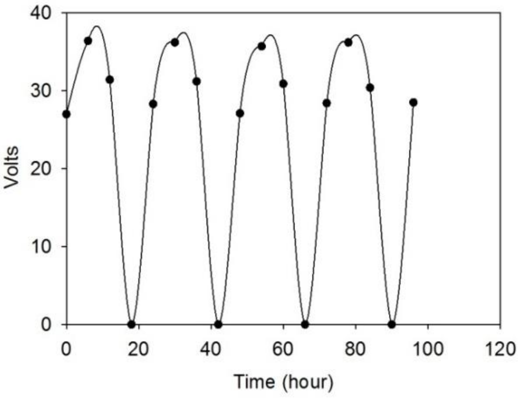

Figure 12. DC volts from solar panel variation versus the time.

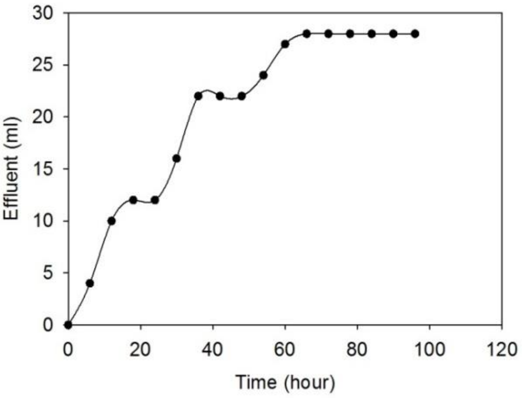

Figure 13. Electro-osmotic flow variation versus the time.

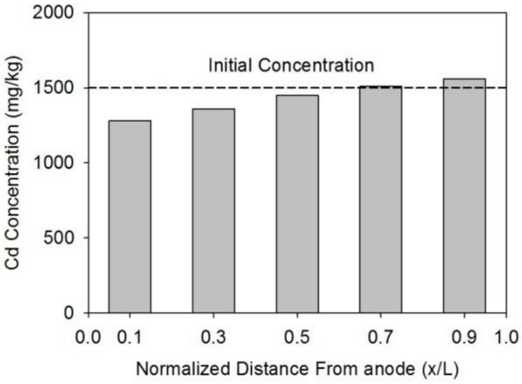

Figure 14: Cadmium concentration in soil after (EK-2).

Fig. 15 shows the pH changes versus normal distance from the anode of the treated soil. Compare with the initial value of soil pH, pH was decreased near the anode compartment and increased near the cathode compartment; because the electrolysis reactions occurred in both compartment.

Due to the low pH at the anode, the Cd concentration decreased and transported toward the cathode compartment. While at the high soil pH, $Cd(OH)_2$ was formed and precipitating near the cathode compartment and Decrease the soil conductivity. That may explain the low removal efficiencies in the solar panel tests (EK-2).

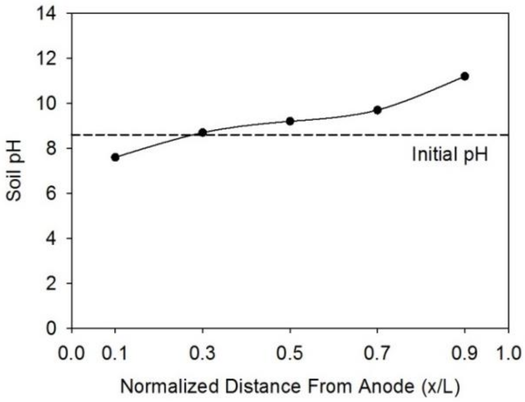

Figure 15. pH in soil after (EK-2).

4. Conclusions

1. The removal efficiency of cadmium from contaminated soil was only 20.8% when 24 V of DC power supply (EK-1) was applied. That can be explained by the high pH at cathode compartment which caused OH^- ions to enter the soil and possibly interfering with the mobility of cadmium ions. The high pH environment was favorable to make $Cd(OH)_2$ which precipitate near the cathode sections (as shown in Fig. 6) and decreases the removal efficiencies of cadmium in EK-1. While only 4.53% removal efficiency of cadmium was reached by the DC volte from two solar panel, each one have maximum voltage of 17 V (as shown in Fig. 14). That can be explained by the same reason in EK-1 an addition to two other reasons. The first one is the time of voltage applied from solar panel at day time and night time. The peak voltage during daytime was 27 to 36 V while 0 V was reported during the night. The second reason is the value of the voltage applied by the solar panel at day time and night time. Hence, an increase in the voltage gradient results in an increase in the removal efficiency of the contaminant. However, and at high voltage gradients, high current densities are induced which increase the rate at which H^+ and OH^- ions are generated due to electrolysis reactions. The rate at which the soil can allow these ions to migrate through it is limited and hence the removal efficiency does not increase proportionally to the rate at which electrolysis reactions occur [23].
2. The use of solar energy at the condition used in EK-2 to remove cadmium from artificially contaminated soil is not efficient because of the low removal efficiencies.
3. Because this low removal efficiencies, condition used in EK-2 can be used to prevent the cadmium transported in soil and accumulated near cathode compartment if used this method as a barrier to prevent cadmium mobility in contaminated soil [24].

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