Remediation of Lead-Contaminated Soil, Using Clean Energy in Combination with Electro-Kinetic Methods

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ABSTRACT: The present study aims at investigating the feasibility of using solar energy as a power source to run electro-kinetic remediation in order to clean-up lead from three types of Iraqi soil. In order to do so, it carries out six tests with enhancement conditions, involving pH control and injection wells. Conducted in the city of Baghdad, Iraq, the experiment, is divided into two groups so that the effect of applying continuous and constant voltage from solar panels study, by means of charge control and battery along with non-continuous and non-constant DC voltage from solar panel could be studied. The DC voltage has been generated by two Solar panels, each with a maximum voltage of 17 volts. All experiments have commenced in March 2017, wherein the soil has been contaminated with a concentration of Pb, equal to 1500 mg/kg as well as initial moisture content equal to 30%. The remediation lasts for seven days, with a potential gradient of about 1.2 V/cm. At the end, the experimental results show that the overall removal efficiencies of 90.7%, 63.3%, and 42.8% have been achieved for sandy, sandy loam, and silty loam soils, respectively, when using solar panels with charge control and battery.

Keywords: Soil pollution, lead removal, Electro-kinetic, solar panel, Soil remediation.

INTRODUCTION

Human activities like agriculture, mining, and –especially-- industrial activities normally cause soil pollution in many ways. This can be in the form of toxic heavy metals (e.g., Cd, Hg, Ni, Pb, and Cr), inorganic compounds (e.g., F^-, CN^-, and arsenic compounds), and a wide variety of organic compounds (e.g., hydrocarbons, BTEX, PAH, PCBs, pesticides, and energetic compounds). Generally, the most common soil contamination occurs in the form of heavy metals, petroleum-based hydrocarbon compounds and solvents, and agricultural pesticides (Cameselle et al., 2013; Alatabe,2018a). Soil contamination by heavy metals becomes an environmental problem over the globe (Puga et al., 2015; Saby et al., 2009; Alam et al., 2015;; Liu et al., 2016; Hu et al., 2017; Gomes et al., 2012), with heavy metals in the soil, likely to exist as soluble compounds like ions and metal complexes or in exchangeable forms. These metals are generally immobilized through being associated with different soil fractions and compartments, such as carbonate, oxide, hydroxide, organic matter, and residual materials (Srivastava et al., 2007). Due to heavy metals' toxicity along with their ability of getting accumulated in the biota, pollution by these metals is a serious problem (Ghaderi et al., 2012). Nasrabadi et al. (2010) found that Sr, Pb, Co, and Cd are the most

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mobile metals, based on the chemical partitioning of metals in the sediments.

The primary industrial sources of lead (Pb) contamination include metal smelting and processing, secondary metal production, lead battery manufacturing, pigment and chemical manufacturing, and lead-contaminated wastes. Widespread contamination due to former use of lead in gasoline is also of high concern. Lead, released to groundwater, surface water, and land, is usually in form of elemental lead, lead oxides, and hydroxides, not to mention lead metal oxyanion complexes. Lead occurs most commonly with an oxidation state of 0 or +II (Evanko and Dzombak, 1997; Smith et al., 1995). It mainly exists in the environment in its stable ion Pb^{+2}, with its high contents in vegetables, grown in contaminated areas, potential to pose health-related risks to consumers [Alatabe & Hussein 2017]. Uptake rate of lead varies among and within species, being highly related to soil pH, with higher lead bioavailability in soils with lower pH. The metal is absorbed by root hairs and stored mainly in cell walls (Heidary-Monfared, 2011; Alatabe, 2018b). Remedial technologies such as incineration, thermal desorption, soil washing, and bioremediation have proven to be either expensive or less effective at field scale application (Srivastava et al., 2007; Virkutyte et al., 2002), with electro-kinetic remediation technique as one of the promising technologies to clean up heavy-metal-contaminated soils, proven to be a very effective tool to remediate contaminated low hydraulic permeability fine grained soils (Acar and Alshawabkeh, 1993; Baraud et al., 1997; Baraud et al., 1999; Saeedi et al., 2009; Caliman et al., 2011; Faisal and Hussein, 2015). 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 (Acar et al., 1995; Yeung et al., 1997; Kornilovich et al., 2005; Yuan and Chiang, 2007; De Battisti,

2007; Elsayed-Ali et al., 2009; Rosestolato et al., 2015). The main mechanisms, leading to the removal of pollutants from soil, include electro-osmosis, electrophoresis, electromigration, and diffusion (Acar and Alshawabkeh, 1993; Acar et al., 1995). There have been many theoretical and experimental studies on electro-kinetic remediation of heavy-metal-contaminated soil. For example, some studies dealt with the effect of initial sand pH (Elsayed-Ali et al., 2009; Alatabe & Hussein, 2018) in an electro-kinetic cell on the amount of copper, which can be extracted from different regions of the cell. Their results offered an optimum pH value of ~2.6, above which the OH^- ions caused copper precipitation and below which the hydrogen ions reduced electro-osmosis, decreasing the effective movement of copper ions to the cathode region. Other studies, however, addressed the effect of pH and washing solution on lead removal from sewage sludge via electro-kinetics (Hanay et al., 2009; Alatabe, 2012), making six experimental attempts at two different pH values of 3 and 4 by means of acetic acid, nitric acid, and phosphoric acid. According to these researches, the use of nitric acid as the washing solution resulted in the highest removal efficiency of Pb (39%), compared to all other experiments. Acetic acid also provided similar removal percentages (37% and 38%) at different pre-acidification conditions. On the other hand, the removal efficiencies stayed at 36% and 27% with pre-acidification, using phosphoric acid at pH value of 3 and 4, respectively, hence resulting in the lowest efficiencies. Results showed that the type of acid, used as washing solution, was less effective than the values of pH for removal of Pb from sewage sludge during the electro-kinetic process. Furthermore, investigation of electro-kinetic remediation of cadmium contaminated soil, driven by a solar cell, composed a third group of studies (Yuan et al., 2009; Alatabe & Hussein, 2017), which found that the output potential of solar cell depended on

daytime, being influenced by weather conditions, while the applied potential in soil was affected by the output potential and weather conditions, and the current achieved by solar cell could be compared with the one, achieved by DC power supply. Electromigration of cadmium in soils by solar cell was comparable with the one via traditional DC power supply; however, the running cost of EK remediation by solar cell was much lower than that of DC power supply. Moreover, a fourth category of experimental studies used solar cells to generate electric field for elector-kinetic remediation of a soft clay soil, contaminated with copper (Hassan and Mohamedelhassan, 2012) where three solar cell panels with peak outputs of 13.5, 27, and 41 V were connected to identical electro-kinetic cells to allow investigation of electro-kinetics' effectiveness for removal of copper after 168 hours of remediation. There, a control test was carried out without electric field to provide a baseline data. Results showed that solar cells could provide sufficient electric field for electro-kinetic remediation. It was also found that electro-osmosis was effective for removing water from the contaminated soil while the removal of copper was primarily due to electromigration. The removal efficiency was proportional to the applied voltage and distance from electrode, with the maximum removed copper being 86% of the initial concentration, itself occurring near the anode.

One of the existing soil pollution problems in Iraq is contamination with lead compounds at various sites with a maximum concentration of more than 33 g Pb/kg soil (MoEn and UNEP, 2005). Hence, the significance of the present study, in comparison with the other studies, lies in its introduction of a novel combination of clean energy and acid-injection well technique with pH control for the sake of a purging solution for the enhancement of lead decontamination from three types of Iraqi soils in the presence of electric field.

MATERIALS AND METHODS

Lead was chosen to represent heavy metal pollutants. It used the formula of $Pb(NO_3)_2$ from AVONCHEM Company (UK). Soil samples, contaminated with Pb concentration equal to 1500 mg/kg and initial moisture content equal to 30% by weight, were prepared by dissolving 7.992 g of $Pb(NO_3)_2$ in one liter of distilled water with 300 ml of this solution added to 1 kg of dry soil. The soil was cleaned and sieved to achieve satisfactory uniformity. Three types of Iraqi soils were used as a porous medium, with Table 1 showing their composition and properties.

Fig. 1 shows the schematic diagram of the electro-kinetic test setup, used for this study. The system was consisted of an electro-kinetic cell, contaminated soil section, collection cylinder, two electrode compartments, four injection wells, two solar panels, two batteries, solar charge controller, and a multi-meter. The glass electro-kinetic cell had an inner dimension of 35 cm x 6 cm x 10 cm and the actual length of the soil specimen in this cell was equal to 20 cm. Also the purging solution, used in the cell, was around 500 ml, with each electrode compartment having 250 ml. the two electrode compartment contained a valve to control the flow in the cell as well as a collection cylinder for collecting the effluent flow from the cell. In order to separate the soil at both ends, of the soil sample, the study utilized perforated plastic plates, which were 1.5 mm thick with 30 punctures, each with an inner diameter of 6 mm, as well as filter paper. (Whatman No. 40). Anode and cathode electrode was a graphite rode, 2 cm across, while the injection well were perforated plastic pipes, the inner diameters of which were equal to 1.27 cm (containing 24 punctures, distributed equally in all four sides, the inner diameter each being 6 mm). DC power came from solar panels (two solar panels, each with a maximum power of 45 watt and voltage of

17 volt) as well as batteries (two 12-volt batteries), and solar charge controller (MPPT type), thus allowing a constant voltage to the electrodes for removal of lead from artificially-contaminated soil. Multi-meter was used to monitor the voltage and measure the current flow through the soil sample during the test.

Table 1. Soil sample composition and properties

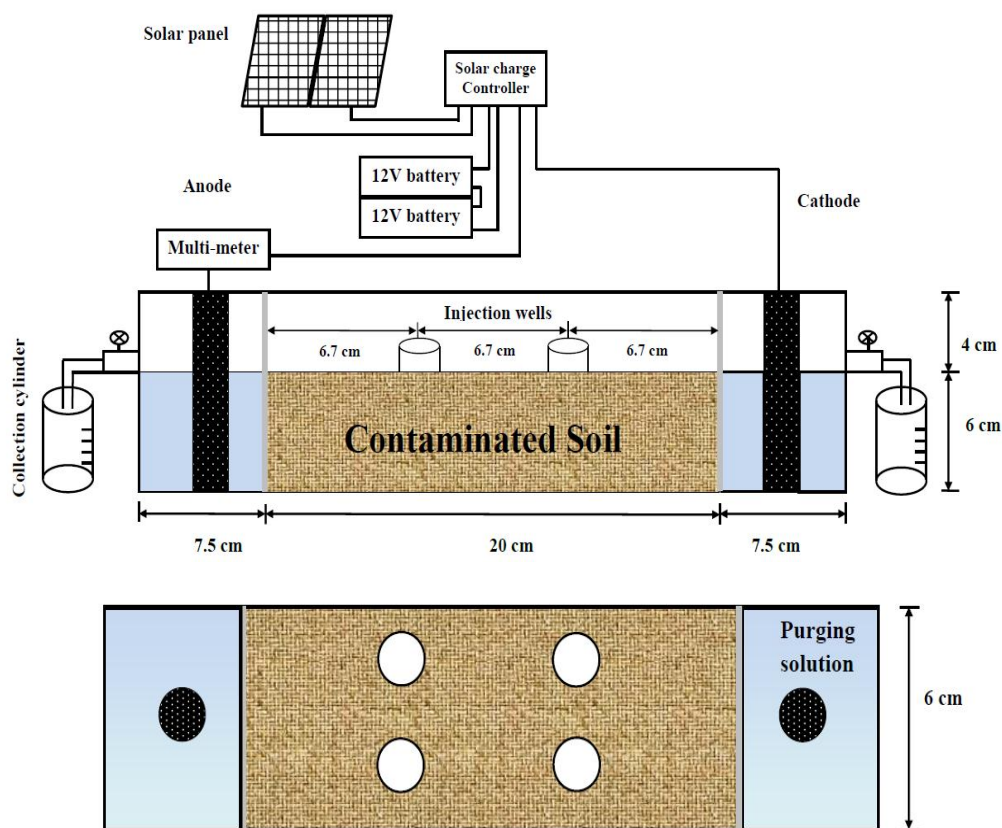| Property	Sandy loam	Silty loam	Sand
Particle size distribution (ASTM D 422)			
Sand (%)	56	4.7	96
Silt (%)	31	68	—
Clay (%)	13	27.3	4 (mud)
Atterberg limits (ASTM D 4318)			
Liquid limit (%)	NL	38.9	NL
Plastic limit (%)	NL	25.05	NL
Plasticity index (%)	NL	13.85	NL
Hydraulic conductivity (or coefficient of permeability) (cm s^{-1})	2.7×10^{-4}	1.2×10^{-8}	1.4×10^{-2}
Cation exchange capacity (meq /100 g)	6.25	17.16	3.1
pH	7.5	7.1	5.6
Organic content (%)	0.03	0.24	0
Electrical conductivity EC (µS/cm)	1356	11590	2210
Porosity (n)	0.41	0.496	0.324

Fig. 1. The experimental instrument of Electro-kinetic cell setup

Table 2. The plan of Electro-kinetic experiments

Test NO.	Soil type	Initial water content%	Processing duration (days)	Applied voltage (V)
EK-1	Sandy loam	30%	7	From solar panel with
EK-2	Silty loam	30%	7	batteries and solar charge
EK-3	Sand	30%	7	controller (~24V)
EK-4	Sandy loam	30%	7	From solar panel only
EK-5	Silty loam	30%	7	according to sun light (27-
EK-6	Sand	30%	7	36V)

Six tests were performed with enhancement conditions, involving pH control and injection wells. The first three tests were conducted, using continuous and constant voltage (~24 V) from solar panels with batteries and solar charge controller to clean-up lead from three types of Iraqi soil, whereas the other three tests employed non-continuous and non-constant DC voltage from solar panels only. What is more, three types of Iraqi soil were used to investigate the effect of applying continuous and constant voltage from solar panels with batteries and solar charge controller, and non-continuous, non-constant DC voltage from solar panels. Table 2 summarizes the test conditions of the experiments. Two solar panels were put into use, each having a maximum voltage of 17 V. The study applied purging solutions on the two electrode compartments to increase the remedial efficiency, where it mixed nitric acid HNO_3 with distilled water to reach a pH value of 3 (0.001 M), to be used as a purging solution. The pH was adjusted during the test in the anode, injection wells, and cathode compartments to range between 2.7 and 3.3 by adding HNO_3 at the cathode, injection wells, and NaOH at the anode compartment to enhance the advancement of the acidic front towards cathode with time. The latitude and longitude of Baghdad City (Iraq), wherein the experiments took place were 33.33333 and 44.43333, respectively. The solar panel was put at an angle of 50º to the horizontal. The experiments commenced in March 2017 with an average daytime length of 12h.
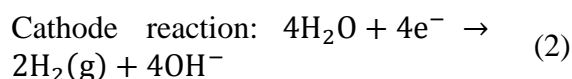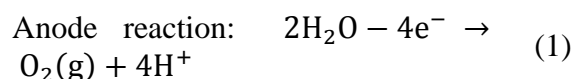
The soil samples were artificially contaminated with lead ions in all electro-kinetic tests. The required amount of the $Pb(NO_3)_2$ to yield the desired concentration was weighed and then dissolved individually in distilled water. The contaminant solution was then added to 1kg of the dry soil and mixed thoroughly with a plastic spatula in polyethylene container. In total, 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 a 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 and the electrode compartments were filled with purging solutions. The elevation of water level in the electrode compartment was kept constant to prevent any formation of hydraulic gradient across the specimen. Also, purging solutions were added to the anode compartment during the test in order to substitute the decrease of solution as the direction of fluid flow was from the anode to the cathode compartment. The electro-kinetic cell was then connected to the DC voltage source. The pH in anode, cathode, and injection wells was continuously adjusted between 2.7 and 3.3 throughout the experiment, wherein the electric current across the soil sample and the water flow got measured at different time periods. At the end of each test, the soil specimen was extruded from the cell. Sectioned into five parts, each part of these soil specimen was weighed and subsequently preserved in a

glass container. From each soil section, five grams of dry soil was taken from all layers 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 an hour. Afterwards, the soil pH was measured [Hansen et al., 2007].

Soil samples from the glass container were collected from all layers and subsequently lead ions was 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 1 g of dry matter and treating the sample in autoclave (JP SELECTA S.A. MICRO8(Spain)). The liquid was separated from the solid particles by vacuum through a 0.45 μm filter and diluted to 100 ml. Lead concentration was determined by atomic absorption spectrophotometer (novAA 300(Germany)) (Hansen et al., 2007).

RESULTS AND DISCUSSION

Electrolysis of water produces hydrogen ions in the anode compartment (Eq. 1), and the acid front migrates across the soil cell, desorbing the contaminants from soil surface in the process, thus initiating electromigration, i.e., transportation of ions and polar molecules in the electric field. Electric potential also leads to electro-osmosis, the flow of an ionic liquid under the action of an applied electric field relative to a charged surface. Electromigration and electro-osmosis are the important mechanisms in electro-kinetic soil processing, which removes contaminants from soil. One of the main defects of conventional electro-kinetics is precipitation of hydroxides near the cathode thanks to generation of hydroxyl ions by water electrolysis in the cathode compartment (Eq. 2), resulting in decreased effectiveness of this technology [Kim et al., 2001; Kim et al., 2003]:

Anode reaction: $2H_2O - 4e^- \rightarrow O_2(g) + 4H^+$ (1)

Cathode reaction: $4H_2O + 4e^- \rightarrow 2H_2(g) + 4OH^-$ (2)

The generated hydrogen and hydroxyl ions are transported through specimen in opposite directions via electrical migration, diffusion, and advection. The advance of base front is slower than that of the acid one, firstly due to the counteracting electro-osmotic flow, and secondly because the ionic mobility of H+ (=13.05 × 10^{-4} m^2/V h) is about 1.76 times higher than that of OH^- (=7.41 × 10^{-4} m^2/V h) (Kim et al., 2001). Consequently, the acid front dominates the chemistry across the specimen, except for some small sections close to the cathode. When electrical potential is applied across a wet soil mass, the cations are attracted to the cathode and the anions to the anode. To be mobile in the electric field, heavy metals must be present in the dissolved phase. Desorption and mobilization of most of the heavy metals during the remediation process are catalyzed by an acidic front that develops from the anode end of the soil. When the acidic front advances through the soil, the remediation action is complete. It has been reported that electro-kinetic technology can be used in combination with other cleanup techniques (Ouhadi et al., 2010; Chung and Lee, 2007; Greičiūtė and Vasarevičius, 2007).

Fig. 2 shows soil pH vs. normal distance from the anode after the end of each test. Electro-kinetic remediation of silty loam soil proceeded relatively slower than that of sandy loam and sand soils. Changes of pH occurred in the soil, as H^+ ions migrated from anode towards the cathode compartment during water electrolysis in the electro-kinetic cell (Acar and Alshawabkeh, 1993). The high-pH environment is favorable for making $Pb(OH)_2$ which precipitates near the cathode sections, decreasing soil conductivity. According to this fact, higher Pb concentrations are to be found near the cathode sections (Virkutyte et

al., 2002; Page and Page, 2002). The continuous adjustment of pH in anode, cathode, and injection wells prevent the precipitation of hydroxides near the cathode due to generation of hydroxide ions by water electrolysis in the cathode compartment.

Fig. 3 shows Pb concentration vs. normal distance from the anode after the end of each test. As can be seen, Pb migrated from the anode, accumulating towards the cathode. The initial concentration of lead throughout the soil was 1500 mg/kg, while the eventual concentration of this metal was lower, varying from 90-910 mg/kg near the anode to 185-1490 mg/kg near the cathode. The concentration profiles in Fig. 3 indicate that removal of lead from the soil in all experiments was below the initial concentration profile. Increased lead concentrations were observed only near the cathode.

Fig. 4 illustrates the electric current's profile during the tests, showing that the current initially varied between 41.2 and 63.4 mA, only to range from 99.1 to 609.2 mA, at the end. As shown, in EK-1 to EK-3 the current increased continuously, indicating that time is important to increase removal efficiency. In other words, removal efficiency rises when remediation time lasts longer. Also in EK-4 to EK-6 the current both increased and decreased to zero, depending on the sun light. The peak voltage, generated from solar panels during the 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 initiation (the test started at 6 am).
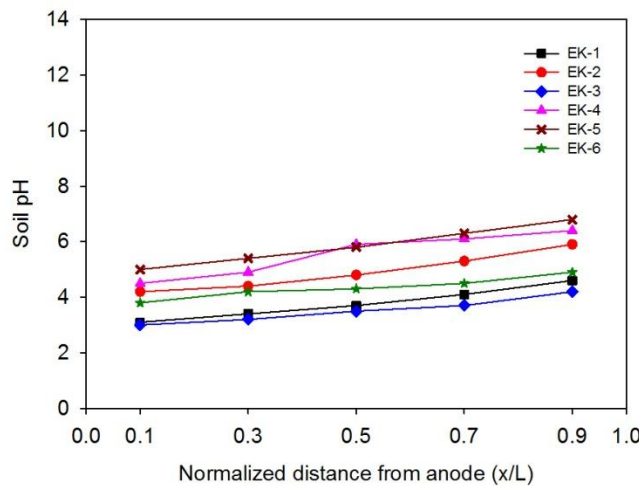

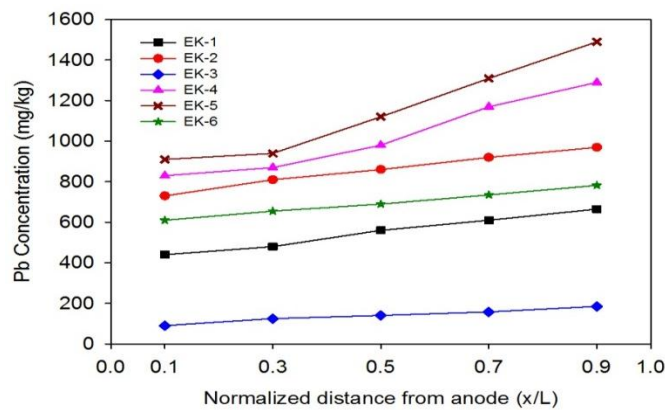**Fig. 2. Changes of pH in the soil with distance**
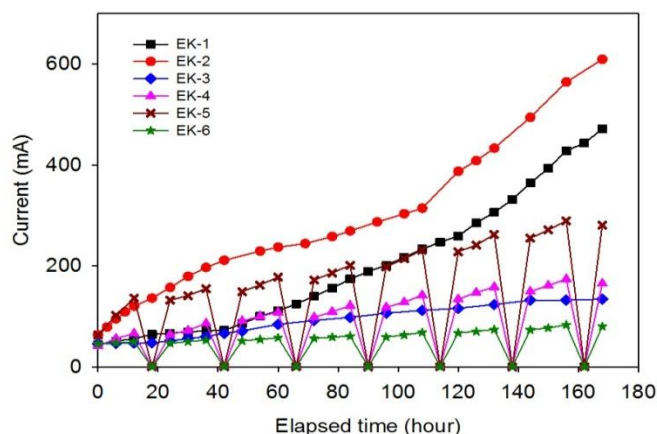

Fig. 3.Distribution of lead concentration in soil

Fig. 4. The change of Current versus time.

Direction of fluid flow was from the anode to the cathode reservoirs (zeta potential ζ <0) [Page and Page, 2002]. Fig. 5 demonstrates the plot of cumulative volume of effluent versus time as the effluent flow increased with time.

Fig. 6 presents the overall contaminant removal efficiencies (calculated based on the residual contaminant masses in the soil) for all conducted tests. The removal efficiencies were 63.3% in EK-1, 42.8% in EK-2, 90.7% in EK-3, 31.24% in EK-4, 23.07% in EK-5, and 53.7% in EK-6. Compared to sandy loam and silty loam soils, the highest removal efficiency was achieved in sand soil. The higher flow in this soil was probably responsible for removal of more lead when the 0.001 M HNO_3 purging solution was employed.

When cleaning the three types of polluted soil, in all cases a higher cleaning efficiency was observed in the anode zone, possibly because of lead ions' positive charge (Pb^{+2}) as well as the potential difference move towards the cathode and accumulation either in the electrolyte or in the nearby soil. It is likely that the extension of the cleanup process duration would enable equalization of concentrations in the cathode and anode sections, while guaranteeing the removal of all mobile lead ions. Electro-kinetic soil remediation was chiefly relevant for cleaning sandy loam and sand. This might be explained by the fact that cleaning efficiency strongly depends on electrical conductivity of soil pore water along how the way pollutant particles move. The grain structure in sandy loam and sand is mostly coarse, as compared with other types of soil, which ensure better movement of metal ions, thus ending up with better cleaning results (Greičiūtė and Vasarevičius, 2007).
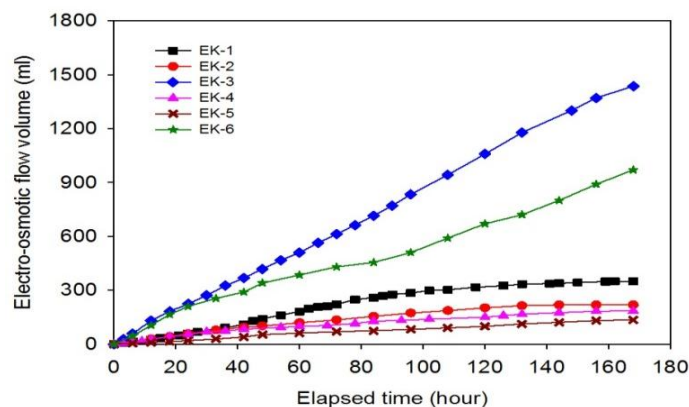

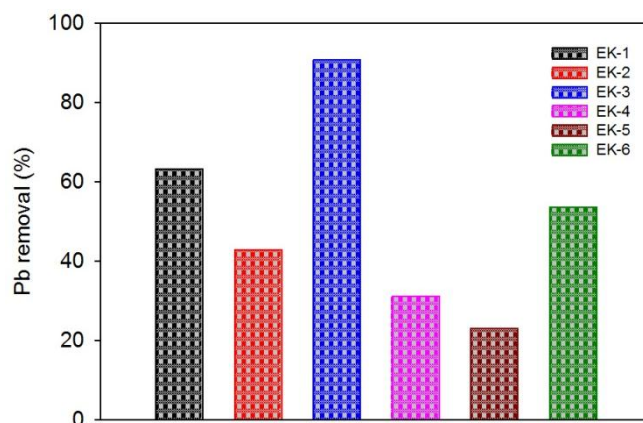**Fig. 5. Electro-osmotic changes against time**

Fig. 6. Lead removal from soil

CONCLUSION

According to the results, obtained from the experimental measurements, the following conclusions can be drawn: the use of solar energy as a power source for running electro-kinetic remediation to clean-up lead from contaminated soil was successful, when using 0.001 M HNO$_3$ as a purging solution, pH control, and injection well. When using solar panels without batteries and solar charge controller, removal efficiencies declined, as nightfall interruptions entailed more time to substitute these stops. The two major mechanisms, expected to govern ion transport process, were electro-osmosis and electromigration. On one hand, the measured flow was low for silty loam soil, indicating marginal electro-osmotic effects in comparison with other soil types, making electromigration the predominant ion transport mechanism in this kind of soil. On the other hand, the measured value of accumulative electro-osmotic effluent was high for sandy soil, indicating that the predominant ion transport mechanism in this type was electro-osmosis. Ionic contaminants must be desorbed before they can be removed and decontaminated. Soils which adsorb strongly and have high cation exchange capacities, e.g., silty loam, were more difficult to decontaminate than the sandy soil which have low cation exchange capacities. For the three soil types, investigated, sand was found to be most conducive to electro-kinetic Pb clean-up, with an overall efficiency of 90.7%, followed by sandy loam (63.3%), and lastly by silty loam (42.8%). As such, electro-kinetics remediation, being a friendly environmental procedure, is less risky and is applicable in low permeable soils.

ACKNOWLEDGEMENTS

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