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## Effect of electrokinetic treatment time on energy consumption and salt ions removal from clayey soils

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## PAPER

# Effect of electrokinetic treatment time on energy consumption and salt ions removal from clayey soils

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## Abstract

Electrokinetics effectively removes contaminants, but its field-scale applications are limited mainly due to its high energy cost. In previous studies, the energy consumption was determined either by changing the soil's specimens initial salt concentration while keeping the treatment time fixed or by changing the treatment time and keeping the same initial salt concentrations for all the specimens. Since both the initial salt concentration and treatment time are important parameters in determining reclamation cost, therefore, in this study, the soil specimens intentionally contaminated with different concentrations of sodium chloride (NaCl), i.e., varying from 3.7 to 15.5 g kg<sup>-1</sup>, were exposed to a constant DC electric field of 1 V cm<sup>-1</sup> for different time durations, i.e., varying from 6 to 72 h. The results show that electroosmotic flow (EOF) was directed from the anode to the cathode and higher for specimens contaminated with relatively low salt concentration, i.e., up to 7.6 g kg<sup>-1</sup>. Therefore, for these specimens, due to the combined effect of electroosmosis and electromigration, the removal of Na<sup>+</sup> was higher than the Cl<sup>-</sup>. However, for the specimen contaminated with a higher salt concentration, i.e., 15.5 g kg<sup>-1</sup>, the Cl<sup>-</sup> removal exceeded Na<sup>+</sup> due to the marginalization of EOF. Regardless of initial salt concentration, the electroosmotic flow and salt ions removal rates decreased with increasing treatment time, which might be attributed to the development of acidic and alkaline environments in soil. The collision of acidic and alkaline fronts resulted in a large potential gradient in a narrow soil region of pH jump, diminishing it everywhere else. This nonlinearity in the electric potential distribution in soil reduced the EOF and electromigration of salt ions.

## 1. Introduction

High salt concentration in the soil can bring drastic changes in its physical and chemical properties resulting in the development of an environment unsuitable for the growth of crops and seed germination. About 831 million hectares of irrigated agricultural land are salt-affected worldwide [1]. Due to salinity, annual global loss in the agriculture sector is estimated to be 12 billion US dollars and is increasing unceasingly [2]. In certain circumstances, routine irrigation and crop management practices cannot reduce excessive salinity in soils. Such situations demand the amelioration of saline soils to improve crop productivity. The common practice for the reclamation of saline soils is to apply excess water to leach salts. However, salt-affected soils are usually low permeable, so leaching water penetration is slow and inefficient. As a result, harmful salts, especially sodium salts, deposit near the soil surface, making it even less permeable [3]. The other soil-reclamation methods, e.g.

chemical amendment [4], soil washing [5] and bioremediation [6], have their limitations, such as environmental concerns, extensive cost, and prolonged treatment times, respectively [7].

To overcome the limitations posed by the methods mentioned above, in 1930, Puri and Anand, for the first time, applied electrokinetics on the field scale to reclaim low-permeable, saline soils [8]. Since then, electrokinetics have been extensively used to decontaminate salt and heavy metal-polluted soils [9–11]. In the electrokinetic method, the moist contaminated soil mass is sandwiched between the electrodes of opposite polarity. Electrokinetic transport processes in contaminated porous media cause the movement of water (electroosmosis) and ionic species (electromigration) under the effect of an applied DC electric field [12].

The soil surface charge dictates the direction of electroosmotic flow (EOF). Since most soils possess a negative surface charge [13]; therefore, in the presence of water (fluid) a mobile layer of positive ions is formed in the vicinity of soil pore surfaces to neutralize its negative surface charge. The formation of these two oppositely charged layers at the liquid-solid interface is known as the electrical double layer [14, 15]. Applying a DC electric field parallel to the interface moves the layer of mobile cations along with hydrated water molecules toward the cathode. Due to dipole-dipole interaction, the movement of the mobile layer induces a drag to the neighbouring molecules in the pore solution, causing a net flow of water from the anode to the cathode [16]. According to Helmholtz-Smoluchowski theory [17], the following equation gives the volumetric flow rate,  $q_{eo}$  ( $m^3 s^{-1}$ ), of water due to electroosmosis.

$$q_{eo} = \frac{\varepsilon \zeta n \Delta V}{\eta \Delta L} A \quad (1)$$

where  $\varepsilon$  ( $F m^{-1}$ ) is the dielectric permittivity of the fluid between the double layer,  $\zeta$  (V) is the zeta potential,  $n$  (–) is the porosity of the soil,  $\eta$  (Pa s) is the dynamic viscosity of pore fluid,  $\Delta V$  (V) is the applied electric potential difference,  $\Delta L$  (m) is the length of the specimen, and  $A$  ( $m^2$ ) is the cross-sectional area of the specimen perpendicular to the flow direction.

A decrease in water dielectric permittivity [18] and the magnitude of zeta potential of clay particles [19], and an increase in dynamic viscosity [20] of water with increasing salt (NaCl) concentration lead to a decrease in electroosmotic flow rate with increasing salt concentration. Furthermore, electroosmotic flow decreases with increasing salt concentration due to the compression of the electrical double layer [9, 21, 22]. Most of the clay's zeta potential values lie between 0 to  $-50$  mV [17] and change with pH [23]. A decrease in EOF rate with the pH changes and even a reversal in EOF direction was observed for  $pH < 3$ , where the sign of zeta potential changed from negative to positive [24, 25]. The soils containing a positive surface charge, i.e., a positive value of zeta potential, the EOF occurs from cathode to anode and is known as electro-endosmosis [26].

Electromigration causes the transport of dissolved ionic species under an applied DC electric field in which positive charges move towards the cathode and negative charges move towards the anode [27]. In porous media, the transport of ionic species by electromigration depends on their effective mobility. In addition to enhance salt ions transport in soil, the applied electric field produces  $H^+$  and  $OH^-$  ions at the anode and cathode, respectively. A higher transfer number of  $H^+$  and  $OH^-$  ions in soils limits the electromigration of targeted ions [28]. However, in the case of inorganic contaminants, the acidic environment promotes their solubility and facilitates their removal by electroosmosis and electromigration [29]. In a highly alkaline environment, the solubility of ions is decreased, which restricts their transport by electromigration [30]. The alkaline environment can be suppressed by using cation exchange membranes [31], chelating agents (such as EDTA, NTA and DTPA) [32] and acids (such as acetic, citric and nitric acid) at the cathode side [33]. In order to hinder the acidic environment, the anion exchange membranes or de-polarizing chemicals such as  $Ca(OH)_2$  are used at the anode side [34].

An electric field intensity of up to  $1 V cm^{-1}$  is preferred to accomplish the electro-reclamation of soils. The electric field intensity higher than  $1 V cm^{-1}$  can result in a fast corrosion of electrodes, especially the anode, due to the high production rate of  $H^+$  ions [35]. The corrosion of electrodes can affect the transport of ionic species and electroosmotic flow due to the production of metallic ions [36]. Therefore, to avoid corrosion, electrodes made of noble metals can be used [37]. However, to make electrokinetic reclamation cost-effective, usually low DC voltage or current is applied across iron/steel, carbon or titanium-coated carbon electrodes [38–40]. Applying a higher DC voltage or current across soil can also result in a rise in its temperature due to Joule's heating effect, which can harm the organic matter and increase energy consumption [41].

In previous studies, electrokinetic effects have been studied either by varying the treatment times only [10] or by changing the salt ions concentrations while keeping the treatment time fixed [19, 42]. The overall aim of this study was to optimize the EK treatment time in clayey soils contaminated with different concentrations of NaCl using stainless steel mesh electrodes. For this purpose, the soil samples were intentionally contaminated with different concentrations of NaCl ranging from  $3.7 g kg^{-1}$  to  $15.5 g kg^{-1}$ , with corresponding EC values varying from  $1.79 dS m^{-1}$  to  $4.85 dS m^{-1}$  as measured by keeping the soil/water ratio (1:5). Since the variations in soil pH, electric current, and electric potential distribution play a decisive role in determining the effective EK

**Table 1.** Physico-chemical properties of the reference soil.

Soil type	Clay soil
Clay	55.3%
Sand	30.2%
Silt	14.5%
pH	8.2
Electrical conductivity (1:5)	0.85 (dS m <sup>-1</sup> )
Hydraulic conductivity	1.6 × 10 <sup>-5</sup> (cm s <sup>-1</sup> )
Organic matter	1.42 (%)
Potassium	0.061 (g kg <sup>-1</sup> )
Calcium	0.166 (g kg <sup>-1</sup> )
Sodium	0.772 (g kg <sup>-1</sup> )
Chloride	0.768 (g kg <sup>-1</sup> )

treatment time and desalination efficiency, therefore, these parameters were measured during these experiments by applying a constant DC electric field of 1 V cm<sup>-1</sup> across the soil specimens.

## 2. Materials and methods

### 2.1. Experimental soil

In this research work, the surface soil (0–20 cm) was collected from the agricultural field of the rice track of Sheikhpura, Punjab, Pakistan (31°42'40"N 73°59'16"E). The soil used for this study is relatively young and dominated by illite-type clay minerals [43, 44]. This soil has been developed in alluvium derived from the Himalayas and deposited by River Indus and its tributaries during the late Pleistocene era [45]. This soil was first air-dried at 25 °C and then powdered with mortar and pestle. The powder was sieved through a 2 mm mesh and evenly mixed before determining the physico-chemical properties of the soil (table 1). The hydrometer method was used to determine the soil texture [46]. The soil pH was measured by mixing 10 grams of soil with 25 ml of distilled water and shaking it in a mechanical shaker (OS 1400) for one hour. After one hour, the suspension was filtered with Whatman quantitative filter paper (Grade 42). The pH of the filtered water was measured by a digital pH meter (HANNA, HI98128). While preparing samples for determining the electrical conductivity (EC) and concentration of salt ions (i.e., Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>+2</sup> and Cl<sup>-</sup>), the ratio of dried soil and distilled water was kept (1:5). The EC of soil samples was measured by EC meter (HANNA, HI99300). The Na<sup>+</sup>, K<sup>+</sup> and Ca<sup>+2</sup> concentrations were measured by a flame photometer (Sherwood-360). The concentration of Cl<sup>-</sup> ions was determined by the argentometric titration method [39]. The soil's organic matter was measured by the loss on ignition (LOI) method [47, 48] by using 20 g of soil and drying it in a furnace at 550 ± 10 °C for one hour. The constant head method was used to determine the hydraulic conductivity of soil [49].

### 2.2. Electrokinetic setup and experimental design

The schematic diagram of the electrokinetic (EK) setup used for the desalination of soil specimens is shown in figure 1. The EK setup comprises three compartments where the soil is sandwiched between the anode and cathode compartments. The soil compartment was built from a PVC pipe length of 9 cm and a diameter of 8.5 cm. The upper half of the PVC pipe was cut off to place soil specimens. This opening in PVC pipe was also used to place pH strips and insert platinum wire for periodically measuring the progression of acidic and alkaline fronts and the variation in electric potential across the soil. To avoid soil drying, the opening at the top side of the PVC pipe was immediately covered with a polythene sheet after periodically taking the pH and electrical potential measurements. The anode and cathode compartments were made from an Acrylic Plexiglass sheet of 6 mm thickness. The dimensions of each electrode's compartment were 7.5 (L) × 11 (W) × 6 (H) cm<sup>3</sup>. A slit connected with a hose was provided at the cathode compartment for the passage of excess water accumulated due to electroosmosis. A graded beaker was attached to the hose to collect and measure the water's electro-osmotically driven outflow rate.

The EK experiments were performed on soil specimens intentionally contaminated with different concentrations of NaCl (i.e., ranging from 3.7 g kg<sup>-1</sup> to 15.5 g kg<sup>-1</sup>). In the EK-1 specimen, no salt (NaCl) was mixed. However, EK-2, EK-3, and EK-4 specimens were mixed with 3.7 g kg<sup>-1</sup>, 7.6 g kg<sup>-1</sup>, and 15.5 g kg<sup>-1</sup> of NaCl, respectively. The dry weight of the soil used to prepare each specimen was 0.550 kg. The saturated paste of soil mixed with a known amount of salt and 170 ml of distilled water was prepared and placed in the soil compartment of the EK setup. Before starting EK experiments, each electrode compartment was filled with 360 ml of distilled water immediately after placing the specimen into the soil compartment. In the EK cell, filter

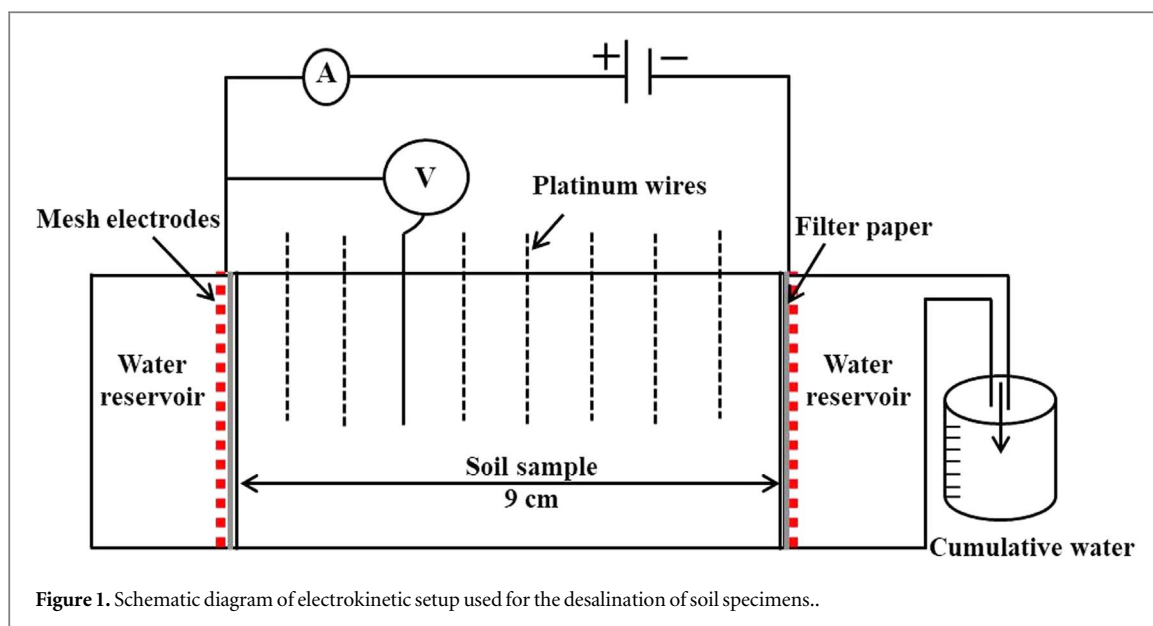


Figure 1. Schematic diagram of electrokinetic setup used for the desalination of soil specimens..

papers were introduced between the stainless steel mesh electrodes and the soil's specimen surface to restrict the movement of soil particles into the electrode compartments. Since the corrosion of stainless steel electrodes, especially the anode, can affect the transport of ionic species and electroosmotic flow due to the production of metallic ions [36], the anode was replaced during EK experiments to avoid the side effects of corrosion. The frequency of replacing an anode depends on the strength of the acidic environment at the anode compartment. For samples containing higher salt concentration, i.e., for EK-4, the anode was replaced every  $\sim 5$  h for the first 36 h. After 36 h of experiments, the anodes were replaced after  $\sim 14$  h up to 72 h due to the low production rate of  $H^+$  ions. However, for low salt contaminated specimens, i.e., EK-1, the anodes were replaced every  $\sim 22$  h.

The effect of treatment time on the removal of salt ions was studied by exposing the identical soil specimens to a DC electric field of  $1 \text{ V cm}^{-1}$  for five different time durations (i.e., 6, 12, 24, 48 and 72 h). The variation in electric current during the experiment was measured periodically (after every 15 min) by inserting a digital multi-meter (UNI-T, UT61A) in series with the DC power supply and the soil specimen. The variation in electric potential across specimens was measured periodically after 2 h by inserting a platinum wire, connected with a voltmeter, in the soil at a distance of every 1 cm from anode to cathode.

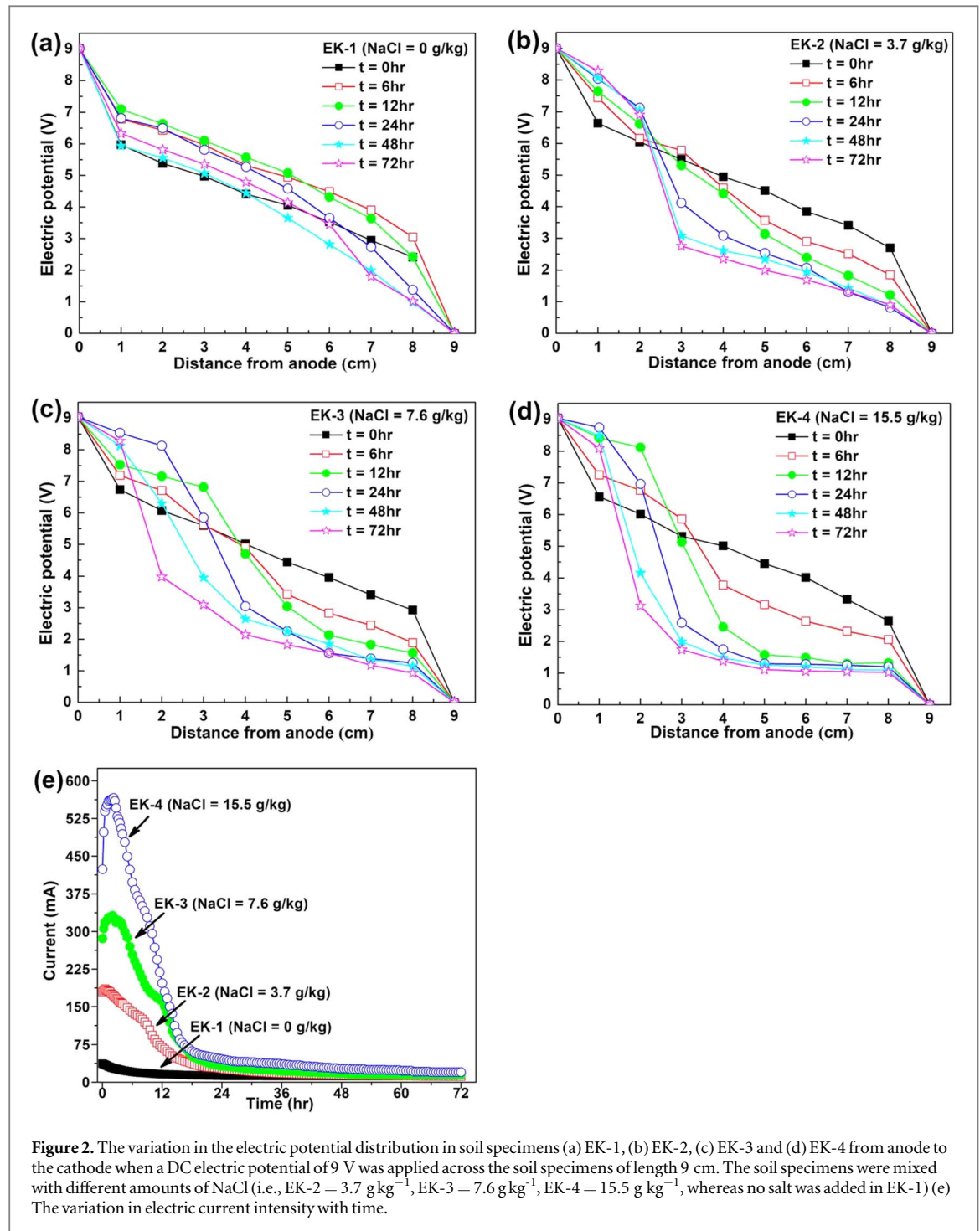
### 2.3. Analytical methods

At the end of each EK experiment, the treated soil samples were immediately segmented into nine equal parts. These soil segments were dried at  $105 \pm 10^\circ \text{C}$  for 5 h in a laboratory oven (DHG 9030 A) and then powdered. For determining the EC and concentration of salt ions, 10 g of powdered mass from each segment was mixed with 50 ml of distilled water for one hr on a mechanical shaker (OS 1400). After that, the suspensions were filtered with the Whatman quantitative filter paper (Grade 42). The electrical conductivity of soil samples was measured with an EC meter (HANNA, HI99300). The concentrations of sodium and chloride ions were measured by a flame photometer (Sherwood-360) and argentometric titration method, respectively. During all EK experiments, the pH changes across the soil were tracked with pH strips (Merck), whereas the pH in electrode compartments was measured by a pH meter (HANNA, HI98128).

## 3. Results and discussion

### 3.1. Electric potential, current, and pH

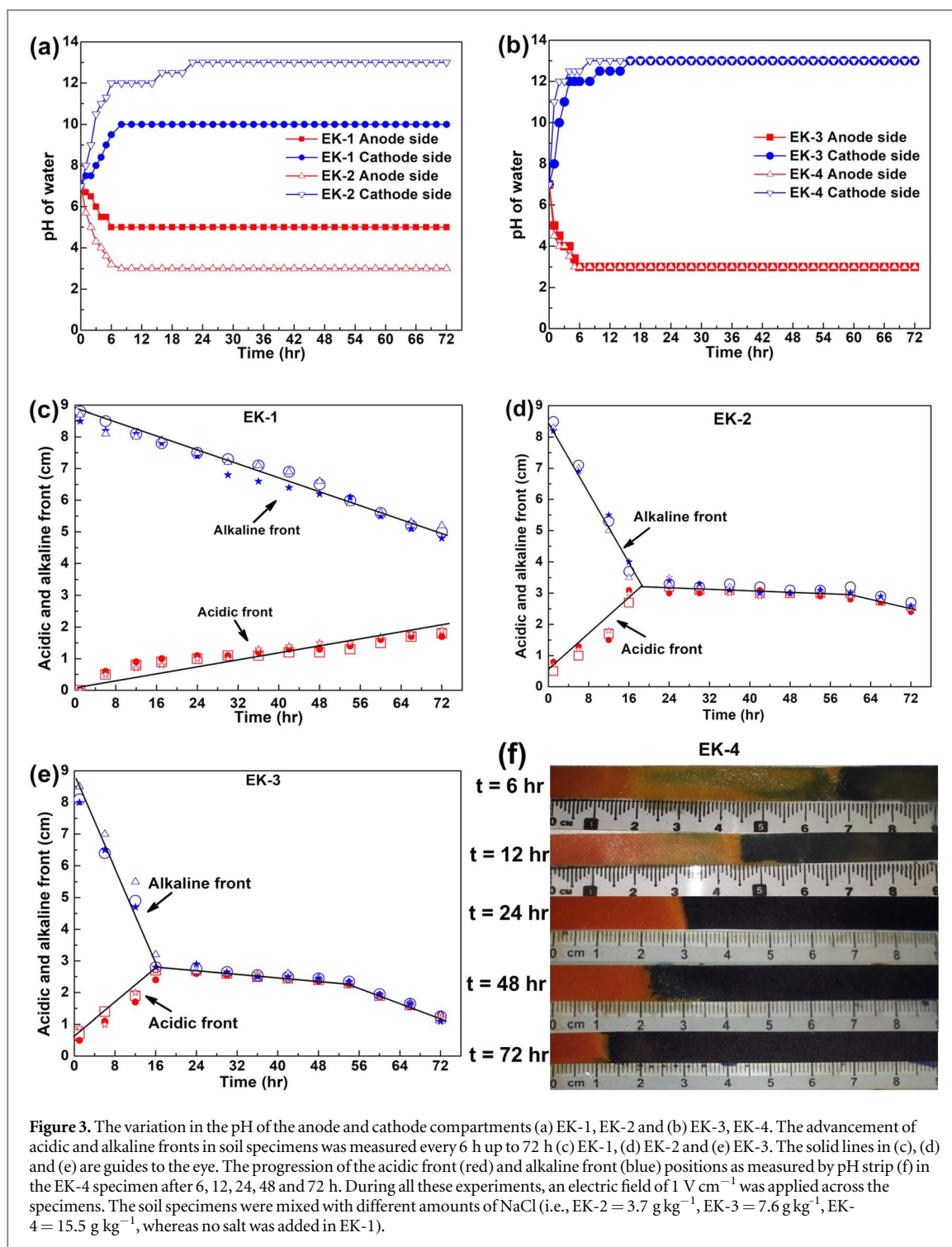
The variation in electric potential across the soil samples containing different salt (NaCl) concentrations is shown in figures 2(a)–(d). In these experiments, a DC electric potential difference of 9 V was applied across the soil specimens of 9 cm length for 72 h. At the beginning of all the EK experiments, a linear variation in electrical potential across the soil specimens was observed. This linearity in the electric potential profiles indicates an initially uniform distribution of moisture and salt ions in the specimens [39, 50]. However, with time, a deviation from linearity in the electric potential profiles was observed in all the specimens except for EK-1 (figure 2(a)), where no salt was mixed with the soil. The deviation from linearity in electric potential profiles indicates a non-uniform distribution of moisture and salt ions due to their transport by electroosmosis and electromigration and the ingress of  $H^+$  and  $OH^-$  ions in the soil's pores solution produced at the electrodes



**Figure 2.** The variation in the electric potential distribution in soil specimens (a) EK-1, (b) EK-2, (c) EK-3 and (d) EK-4 from anode to the cathode when a DC electric potential of 9 V was applied across the soil specimens of length 9 cm. The soil specimens were mixed with different amounts of NaCl (i.e., EK-2 =  $3.7 \text{ g kg}^{-1}$ , EK-3 =  $7.6 \text{ g kg}^{-1}$ , EK-4 =  $15.5 \text{ g kg}^{-1}$ , whereas no salt was added in EK-1) (e) The variation in electric current intensity with time.

compartments due to electrolysis of water. Furthermore, it was observed that the deviation from linearity in the electric potential profiles for specimens containing higher salt concentrations started earlier and became more prominent due to a relatively high potential drop within these specimens compared to specimens containing low salt concentrations.

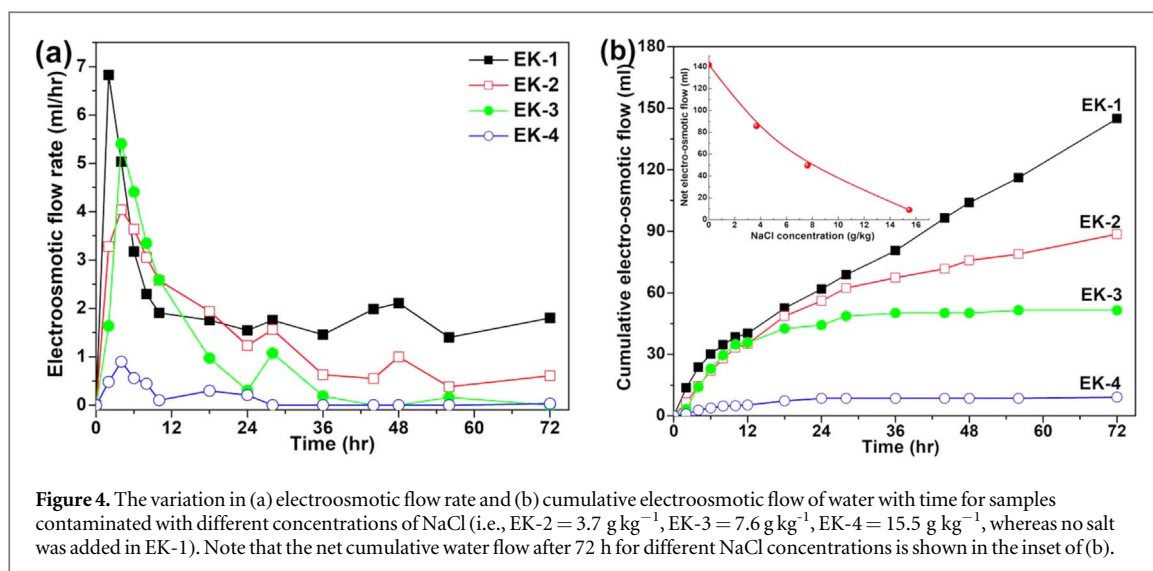
On the other hand, initially, a relatively higher potential drop at the edges of the specimens adjacent to the electrodes in a region of  $\sim 1 \text{ cm}$  was observed. This potential drop indicates a low electrical conductivity at the electrode-soil interface due to the insufficient free ionic species in the electrode compartments initially filled with distilled water. Therefore, the low electrical current intensity at the start of all EK experiments (that is more prominent for the specimens EK-3 and EK-4), as shown in figure 2(e), is due to an initially low electrical conductivity at the electrode-soil interface. However, with time the electric potential difference between electrodes and the edges of the specimens started to diminish due to an increase in the number of free ions in electrode compartments by the production of  $\text{H}^+$  and  $\text{OH}^-$  ions due to electrolysis and the transport of salt ions by the combined effect of diffusion, electroosmosis and electromigration. This increase in free ions



**Figure 3.** The variation in the pH of the anode and cathode compartments (a) EK-1, EK-2 and (b) EK-3, EK-4. The advancement of acidic and alkaline fronts in soil specimens was measured every 6 h up to 72 h (c) EK-1, (d) EK-2 and (e) EK-3. The solid lines in (c), (d) and (e) are guides to the eye. The progression of the acidic front (red) and alkaline front (blue) positions as measured by pH strip (f) in the EK-4 specimen after 6, 12, 24, 48 and 72 h. During all these experiments, an electric field of  $1 \text{ V cm}^{-1}$  was applied across the specimens. The soil specimens were mixed with different amounts of NaCl (i.e., EK-2 =  $3.7 \text{ g kg}^{-1}$ , EK-3 =  $7.6 \text{ g kg}^{-1}$ , EK-4 =  $15.5 \text{ g kg}^{-1}$ , whereas no salt was added in EK-1).

concentration in electrode compartments enhanced the electrical conductivity at the electrode-soil interface leading to an increase in electric current intensity (figure 2(e)), indicating an increase in the removal rate of salt ions. However, the electrical current intensity decreased after reaching its maximum value due to a decrease in salt concentration and the ingress of acidic and alkaline environments in the soil specimens.

The variation in pH of electrode compartments due to the production of  $\text{H}^+$  ions at the anode and  $\text{OH}^-$  ions at the cathode is shown in figures 3(a), (b). In the case of EK-1, where no salt was mixed in the soil, the pH of the anode reservoir varied from 7 to 5, and the cathode compartment's pH varied from 7 to 10 within  $\sim 6$  h. While during the same experimental time, i.e.,  $\sim 6$  h, the soil specimen EK-4, contaminated with  $15.5 \text{ g kg}^{-1}$  sodium chloride, the pH of the anode compartment varied from 7 to 3, whereas the pH of the cathode compartment varied from 7 to 13. This difference in pH indicates that the initial concentration of salt in the soil specimen influences the electrical current intensity, which in turn dictates the pH variation in the electrode compartments and, thus, in the soil [51]. The pH strips were used to track the progression of acid and alkaline fronts across the



**Figure 4.** The variation in (a) electroosmotic flow rate and (b) cumulative electroosmotic flow of water with time for samples contaminated with different concentrations of NaCl (i.e., EK-2 =  $3.7 \text{ g kg}^{-1}$ , EK-3 =  $7.6 \text{ g kg}^{-1}$ , EK-4 =  $15.5 \text{ g kg}^{-1}$ , whereas no salt was added in EK-1). Note that the net cumulative water flow after 72 h for different NaCl concentrations is shown in the inset of (b).

soil specimens, as shown in figure 3(f), where the red indicates acidic, and the blue indicates an alkaline environment. The variations in acidic and alkaline front positions with time in the soil specimens, i.e., EK-1, EK-2, and EK-3, are shown in figures 3(c)–(e). It can be seen (figures 3(c)–(e)) that acidic fronts travelled less distance than alkaline fronts in soil, irrespective of EOF directed from the anode towards the cathode and the higher mobility of  $\text{H}^+$  ions ( $\mu_{\text{H}^+} = 36.2 \times 10^{-8} \text{ m}^2\text{s}^{-1}\text{V}^{-1}$ ) than  $\text{OH}^-$  ions ( $\mu_{\text{OH}^-} = 20.6 \times 10^{-8} \text{ m}^2\text{s}^{-1}\text{V}^{-1}$ ) in dilute solutions [52]. The short distance travelled by the acidic fronts can be attributed to the alkaline nature of the soil (initial soil pH was 8.2, as given in table 1) and the buffering capacity of alkaline soils against the acid [10]. The effect of the acid buffering capacity of the soil on pH changes during electrokinetics can be observed by the receding of the acidic front towards the anode after its collision with the alkaline front, i.e., from 3 cm (after 24 h) to 1.5 cm (after 72 h) as shown in figure 3(f). A similar receding trend in the acidic fronts towards the anode side was observed for the specimens EK-2 and EK-3 after their collision with the alkaline fronts, as shown in figures 3(d), (e). However, for the specimen EK-1, where no salt was mixed with soil, though the alkaline front travelled more distance than the acidic front, the collision of these fronts did not occur due to the low production rate of  $\text{H}^+$  and  $\text{OH}^-$  ions as well as their slow movement due to low electric current intensity.

After the collision of acidic and alkaline fronts, the intensity of electric current became very low and remained steady (after  $\sim 16$  h) for all the specimens, irrespective of their initial and remaining salt ions concentration (figure 2(e)). A decrease in electric current intensity reflects a reduction in salt ions removal rate during the electrokinetic treatment. A reduction in the electrokinetic transport of  $\text{ZnCl}_2$  was also observed due to the precipitation of  $\text{Zn}^{+2}$  ions in the alkaline region of clay soil [53]. It was further shown by Kamran *et al* [54] that the ions that do not precipitate (i.e.,  $\text{Na}^+$ ) can also be halted due to the development of pH gradients during electrokinetic treatment. An enormous reduction in electric current intensity due to a sharp potential drop after the collision of acidic and alkaline fronts was also observed during the electrokinetic desalination of fired-clay bricks [39, 54].

### 3.2. Electroosmosis

The effect of salt concentration on electroosmotic flow (EOF) is shown in figure 4. The electroosmotic flow was measured every 2 h for the first 12 h after the start of experiments. However, after 12 h, the EOF was measured randomly, and the interval between any two consecutive measurements varied from 4 to 16 h. The EOF occurred from the anode to the cathode side of the specimen, indicating a net negative surface charge on the soil surface [13]. The maximum EOF was observed in the case of EK-1 (figure 4(b)), where no salt was mixed with the soil. In this case, the total amount of water collected at the cathode side of the reservoir was  $\sim 142$  ml after the completion of the experiment, i.e., after 72 h. However, with an increase in initial salt concentration, cumulative flow decreased, as shown in the inset of figure 4(b). The minimum EOF was observed for EK-4, where the total water collected at the cathode side of the reservoir was  $\sim 10$  ml. The decrease in EOF with increasing the salt concentration, according to Helmholtz-Smoluchowski theory, can be attributed to a decrease in dielectric permittivity and zeta potential (presented in the numerator of equation (1)) and an increase in the viscosity of pore fluid (in the denominator of equation (1)) [18–20]. In addition, a decrease in the thickness of the electrical double layer with increasing salt concentration can cause a decrease in EOF [9, 21, 22].

Moreover, the electroosmotic flow rate was initially higher and decreased with time, as shown in figure 4(a). Instead of a decrease, an increase in EOF rate was expected due to a decrease in soil's salt ions concentration with



an increase in treatment time. This decrease in EOF can be attributed to the changes in the zeta potential of soil due to the ingress of acidic and alkaline fronts [23] produced by the electrolysis of water at the electrodes. A decrease in EOF rate with the pH changes in soil and even a reversal in its EOF direction was observed for  $\text{pH} < 3$  at the anode side of the soil specimen [24, 50].

### 3.3. Salt ions removal and electrical conductivity

During electrokinetic (EK) desalination experiments, salt ions are transported from the soil to the electrode compartments due to diffusion, electroosmosis and electromigration. The variation in sodium ( $\text{Na}^+$ ) and chloride ( $\text{Cl}^-$ ) ions concentration in soil under the effect of an electric field of  $1 \text{ V cm}^{-1}$  after 6, 12, 24, 48 and 72 h for EK-1 to EK-4 are shown in figure 5. The dashed lines in figure 5 represent the initial concentration of  $\text{Na}^+$  and  $\text{Cl}^-$  ions in the soil specimens before the start of EK experiments. Under an applied DC electric field, the  $\text{Na}^+$  ions are transported towards the cathode side, while the  $\text{Cl}^-$  ions move towards the anode side. In this process, some of the  $\text{Cl}^-$  ions can be converted into chlorine gas due to an oxidation reaction at the anode side, as given in equation (2) [55].



The decrease in  $\text{Na}^+$  and  $\text{Cl}^-$  ions concentration in soil specimens after 6, 12, 24, 48 and 72 h due to the combined effect of diffusion, electroosmosis and electromigration shown in figure 5. It can be seen (figure 5) that an overall removal of both the  $\text{Na}^+$  and  $\text{Cl}^-$  ions increased with an increase in treatment time as was observed by Bessaim *et al* [10]. Though the overall removal of salt ions increased with time, their removal rate decreased with treatment time. As shown in table 2, during the first 6 h of treatment, 47%, 42%, 40% and 34% of the  $\text{Na}^+$  ions were removed from specimens EK-1, EK-2, EK-3, EK-4, respectively. For the next 66 h of treatment, i.e., from 6 to 72 h, the per cent increase in  $\text{Na}^+$  removal was 46%, 43%, 45%, and 29%, respectively, for the specimens from EK-1 to EK-4. A similar trend was observed for the  $\text{Cl}^-$  ions except for the specimen EK-4, where their removal exceeded  $\text{Na}^+$  ions due to the lack of electroosmotic flow. This indicates that the removal rate of salt ions is decreased by increasing treatment time. This decrease in the removal rate of  $\text{Na}^+$  and  $\text{Cl}^-$  ions is caused by the progression of  $\text{H}^+$  and  $\text{OH}^-$  ions in soil from the electrode compartments. Due to the exceptionally high mobilities of  $\text{H}^+$  and  $\text{OH}^-$  ions compared to  $\text{Na}^+$  and  $\text{Cl}^-$  ions, most of the ionic current in the soil is carried by these ions, thus affecting the removal of salt ions. However, after the collision of acidic and alkaline fronts (figure 3), a large potential gradient is developed in a narrow soil region of high pH jump, diminishing it everywhere else. Consequently, the electric current becomes very small (figure 2(e)), indicating a limiting effect of the electric field on the transport of salt ions [39, 54].

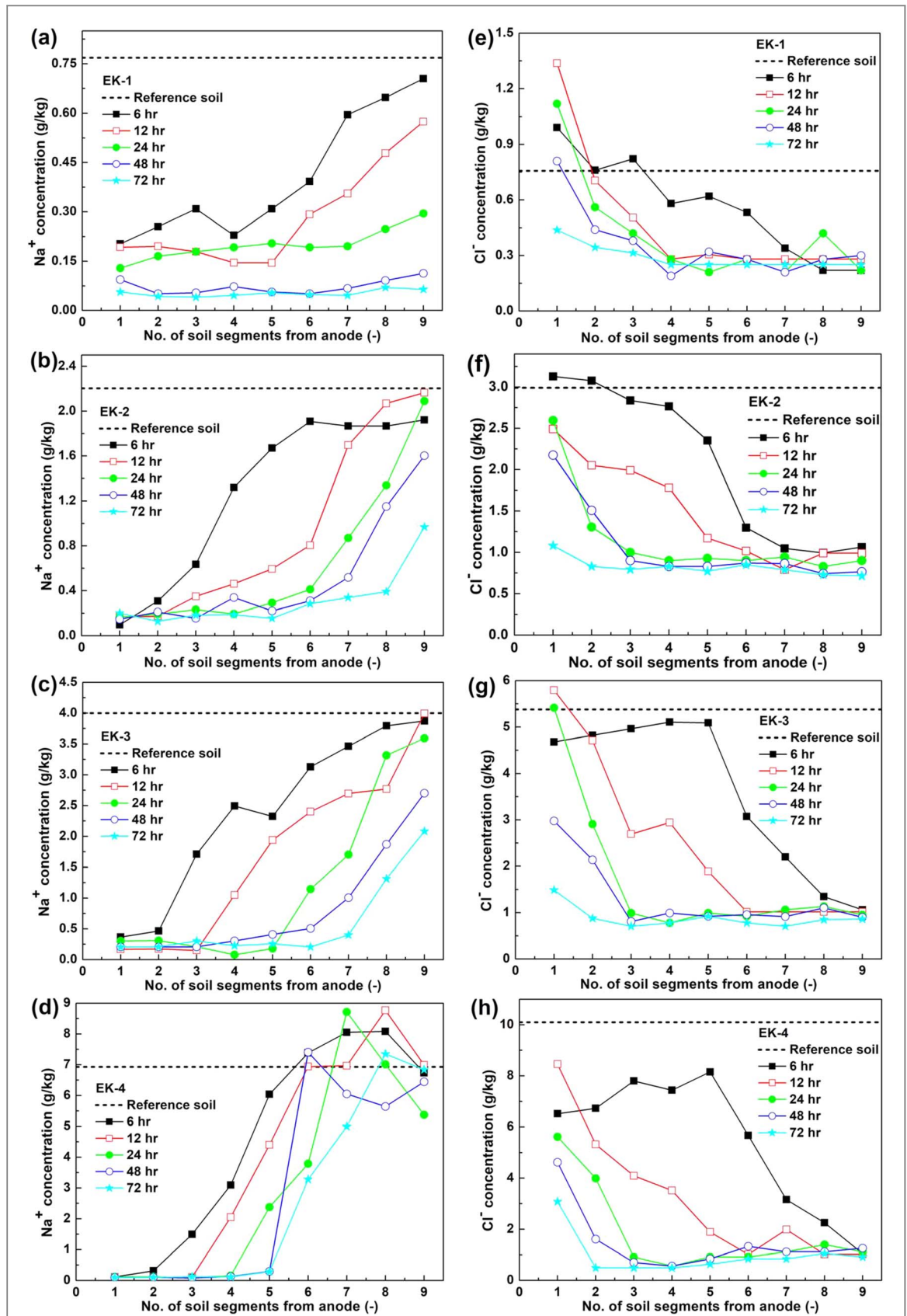
The electrical conductivity (EC) changes in soil (figure 6) followed the same trend observed by the salt ion concentration variation with treatment time. Since, under the effect of an applied DC electric field, the  $\text{Na}^+$  ions move towards the cathode, and  $\text{Cl}^-$  ions move towards the anode side of the specimen. Moreover, the ingress of the acidic and alkaline front is higher at the edges of the specimen. Therefore, the EC becomes higher at the edges, i.e., near the anode and cathode, whereas it becomes lower in the middle of specimens.

### 3.4. Salt ions removal efficiency

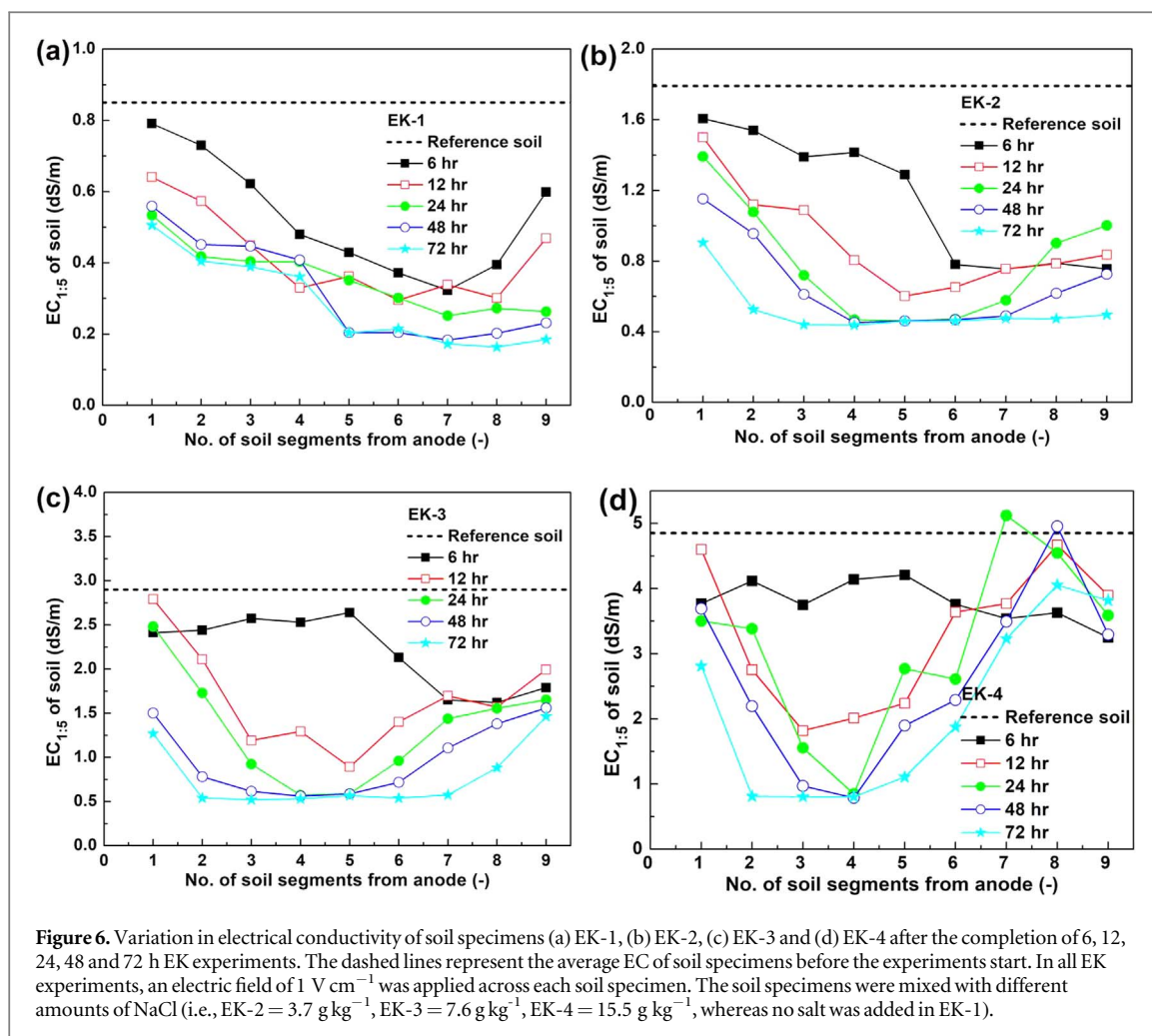
The removal efficiency ( $\eta$ ) of  $\text{Na}^+$  and  $\text{Cl}^-$  ions for different treatment times, i.e., 6, 12, 24, 48 and 72 h for all EK experiments, was calculated by using equation (3) [39].

$$\eta = \left( 1 - \frac{\sum_{f=1}^9 C_f}{\sum_{i=1}^9 C_i} \right) \times 100\% \quad (3)$$

Where,  $\sum_{f=1}^9 C_f$  is the sum of the residual concentration of  $\text{Na}^+$  and  $\text{Cl}^-$  ions in each segment of the soil specimen after EK treatments, and  $\sum_{i=1}^9 C_i$  is the sum of  $\text{Na}^+$  and  $\text{Cl}^-$  ions in each soil specimen before the EK experiment. Since  $\text{Na}^+$  ions are moved toward the cathode by the combined effect of electroosmotic flow and ionic migration, the effect of ionic migration of  $\text{Cl}^-$  ions is expected to be diminished due to electroosmotic advection. Therefore, irrespective of higher mobility of  $\text{Cl}^-$  than  $\text{Na}^+$  ions [56], due to the relatively enormous EOF rate in soil specimens contaminated with a relatively low concentration of NaCl, the removal of  $\text{Na}^+$  is higher than  $\text{Cl}^-$  ions for EK-1, EK-2 and EK-3. While, for the EK-4 specimen containing a higher salt concentration (i.e.,  $15.5 \text{ g kg}^{-1}$ ), the removal of  $\text{Cl}^-$  ions is higher than  $\text{Na}^+$  ions due to the suppression of the EOF. In the absence of electroosmosis, salt ions transport is dominated by electromigration. Therefore, an increase in the removal rate of  $\text{Cl}^-$  than  $\text{Na}^+$  ions (i.e., in the case of EK-4) is expected due to higher ionic mobility of  $\text{Cl}^-$  than  $\text{Na}^+$  ions [56]. The removal efficiency of both the  $\text{Na}^+$  and  $\text{Cl}^-$  ions for different treatment times, i.e., 6, 12, 24, 48 and 72 h, is given in table 2.



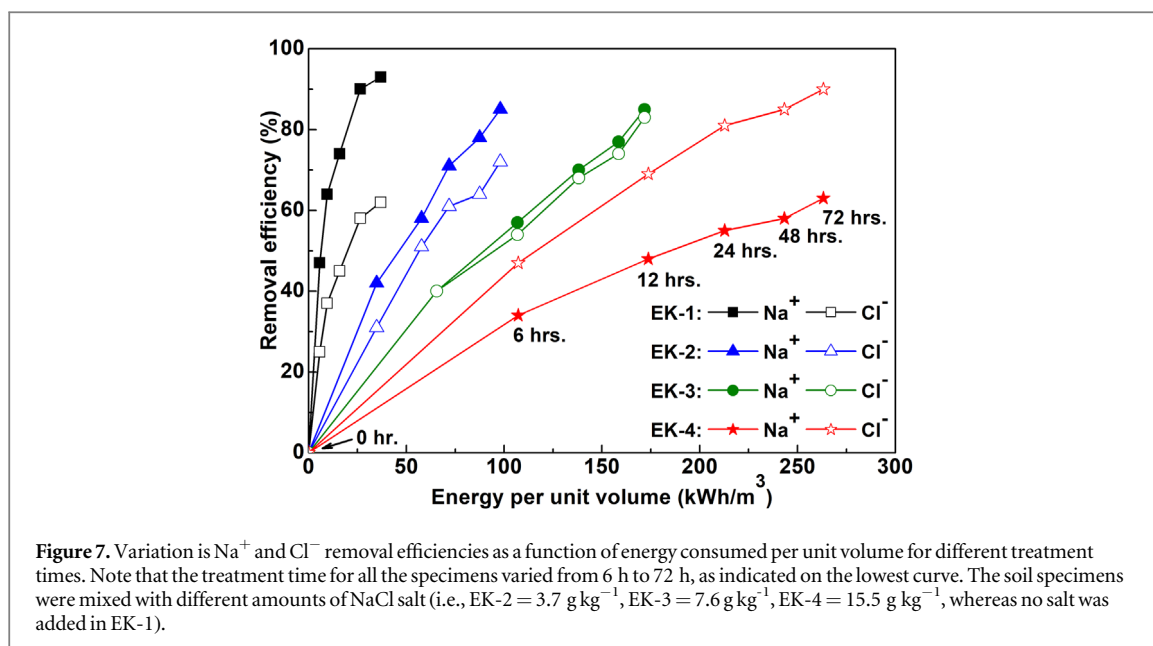
**Figure 5.** Here (a)–(d) represent the initial and final concentrations profiles of Na<sup>+</sup> in EK-1 to EK-4, respectively and (e)–(h) represent the initial and final concentration profiles of Cl<sup>-</sup> in EK-1 to EK-4, respectively after the completion of 6, 12, 24, 48 and 72 h. EK experiments. The dashed lines represent the averaged initial concentrations of sodium and chloride ions. During all these experiments, a DC electric field of 1 V cm<sup>-1</sup> was applied across the soil specimens. The soil specimens were mixed with different amounts of NaCl (i.e., EK-2 = 3.7 g kg<sup>-1</sup>, EK-3 = 7.6 g kg<sup>-1</sup>, EK-4 = 15.5 g kg<sup>-1</sup>, whereas no salt was added in EK-1).



**Figure 6.** Variation in electrical conductivity of soil specimens (a) EK-1, (b) EK-2, (c) EK-3 and (d) EK-4 after the completion of 6, 12, 24, 48 and 72 h EK experiments. The dashed lines represent the average EC of soil specimens before the experiments start. In all EK experiments, an electric field of  $1 \text{ V cm}^{-1}$  was applied across each soil specimen. The soil specimens were mixed with different amounts of NaCl (i.e., EK-2 =  $3.7 \text{ g kg}^{-1}$ , EK-3 =  $7.6 \text{ g kg}^{-1}$ , EK-4 =  $15.5 \text{ g kg}^{-1}$ , whereas no salt was added in EK-1).

**Table 2.** Variation in  $\text{Na}^+$  and  $\text{Cl}^-$  concentration, electrical conductivity, cumulative flow, and energy consumption during electrokinetic experiments performed for different durations under an applied electric field of  $1 \text{ V cm}^{-1}$ .

Specimen/Amount of NaCl added ( $\text{g kg}^{-1}$ )	Treatment time (h)	$\text{Na}^+$ Removal efficiency (%)	$\text{Cl}^-$ Removal efficiency (%)	Initial $\text{EC}_{1.5}$ ( $\text{dS m}^{-1}$ )	Final $\text{EC}_{1.5}$ ( $\text{dS m}^{-1}$ )	Reduction in $\text{EC}_{1.5}$ (%)	Cumulative flow ( $\pm 1 \text{ ml}$ )	Energy consumption per unit volume ( $\text{kW-hr m}^{-3}$ )
EK-1/(0 $\text{g kg}^{-1}$ )	6	47	25	0.85	0.53	38	30	5.71
	12	64	37	0.85	0.42	51	41	9.52
	24	74	45	0.85	0.35	59	62	15.87
	48	90	58	0.85	0.32	62	104	26.45
	72	93	62	0.85	0.28	67	145	36.91
EK-2/(3.7 $\text{g kg}^{-1}$ )	6	42	31	1.79	1.14	36	22	34.81
	12	58	51	1.79	0.90	49	35	57.75
	24	71	61	1.79	0.78	56	56	71.86
	48	78	64	1.79	0.66	63	76	87.41
	72	85	72	1.79	0.52	71	89	98.05
EK-3/(7.6 $\text{g kg}^{-1}$ )	6	40	40	2.92	2.19	24	23	65.47
	12	57	54	2.92	1.66	43	36	106.8
	24	70	68	2.92	1.31	55	44	138.2
	48	77	74	2.92	0.97	67	50	158.5
	72	85	83	2.92	0.76	74	52	171.8
EK-4/(15.5 $\text{g kg}^{-1}$ )	6	34	47	4.85	3.79	22	3.9	107.2
	12	48	69	4.85	3.26	33	5.4	173.7
	24	55	81	4.85	3.00	38	8.6	212.8
	48	58	85	4.85	2.42	50	8.8	243.3
	72	63	90	4.85	2.04	58	9.1	263.2



### 3.5. Energy consumption

Energy consumption is crucial in determining the feasibility of EK method for the reclamation of salt-affected soils. The energy consumption was calculated by using the values of periodically measured (after every 15 min) electric current intensity and the applied DC electric potential (9 V) for different experimental times (i.e., 6, 12, 24, 48, and 72 h). The energy consumption per unit volume was calculated by using the equation (4) [57].

$$E_V = \frac{E}{V} = \frac{\int I U dt}{V} \quad (4)$$

Where  $E_V$  is the energy consumed per unit volume of soil [ $\text{kW-hr m}^{-3}$ ],  $E$  is the energy consumed [ $\text{W-hr}$ ],  $V$  is the volume of soil [ $\text{m}^3$ ],  $U$  is the applied electric potential [ $\text{V}$ ],  $I$  is the electric current [ $\text{A}$ ] and  $t$  is the electrokinetic treatment time [ $\text{hr.}$ ]. Note that the volume of each soil specimen was  $\sim 2.55 \times 10^{-4} \text{ m}^3$ .

Since all the EK experiments were performed by applying a constant DC electric potential of 9 V across the soil specimen's fixed mass, the variation in electric current intensity is the only parameter determining the energy consumption. Furthermore, the electric current depends on the salt ions concentration in the soil. Therefore, both the current (figure 2(e)), as well as energy consumption per unit volume (table 2) are higher for samples initially contaminated with higher salt concentration. However, over time, the electric current starts to decrease due to the low removal rate of salt ions caused by the collision of acidic and alkaline fronts and the development of a large potential gradient in soil, resulting in a reduction in electrical energy consumption (figure 7).

## 4. Conclusions

The results show that the salt ions removal efficiency and electrical energy consumption depend on electrokinetic treatment time. Soils contaminated with higher salt concentration require more electrical energy due to increased electrical current intensity and electrokinetic treatment time. With increased salt concentration, the EOF decreased, and it became negligibly small for the soil specimen, i.e., EK-4, contaminated with  $15.5 \text{ g kg}^{-1}$  sodium chloride. The EOF was directed from the anode to the cathode, and a decrease in EOF resulted in a decrease in the removal efficiency of cations ( $\text{Na}^+$  ions). The salt ions removal efficiency was higher at the start. Irrespective of initial salt concentration, more than 45% of salt ions ( $\text{Na}^+$  and  $\text{Cl}^-$ ) were removed from all the intentionally contaminated specimens during the first 12 h of electrokinetic treatment. A decrease in salt ions removal efficiency at the later stage of treatment can be attributed to the ingress of acidic and alkaline environments and the development of a potential gradient in soil. The variation in soil EC followed the distribution of  $\text{Na}^+$ ,  $\text{H}^+$ ,  $\text{Cl}^-$  and  $\text{OH}^-$  ions concentrations, i.e., over time, its value increased at the edges, and it diminished in the middle of the specimens.

## Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

## Conflicts of interest

The authors declare that they have no conflict of interest.

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