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Remediation of Chromium-Contaminated Soils by Electro-Kinetic Technique

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ABSTRACT

Heavy metal contamination, especially chromium, poses severe environmental threats due to its persistence and widespread presence in soil. This research investigates the effectiveness of electrokinetic remediation (EKR) for removing chromium from contaminated soil. Experiments were conducted on sandy and sandy clay soils, with pH adjusted to 2 and oxalic acid used as an enhancement agent. A constant voltage of 1.2 V/cm was applied. The results showed a chromium removal efficiency of up to 81.9% in the sandy soil remediation with oxalic acid, while the sandy loamy soil showed a lower removal efficiency of 65.26%. These results suggest that EKR, especially when combined with oxalic acid, is a promising method for in situ chromium removal due to the ability of oxalic acid to increase the solubility of chromium, which facilitates its removal. However, differences in soil types significantly affect the effectiveness of the remediation process.

1. Introduction

soil contamination is causing irreversible land losses all over the world, and there are serious concerns regarding the sustainability of soil resource. Even following source removal, soil contaminants might remain accumulated for many years, impacting soil functions, reducing soils' capability for supporting life systems, and reducing soils' capability to offer ecosystem services [1]. Generally speaking, the majority of pollutants enter the environment through sewage, waste, unintentional discharge, or as residues or byproducts from the creation of something beneficial [2]. Metals are among the

most prevalent and harmful kinds of contaminants in soil today. Soil contamination through heavy metals represents a major environmental problem on a global scale, and the industrialized world [3], [4], [5]. In order to limit, prevent, or mitigate risk to property, public health, and the environment, soil remediation is the management of soil contaminant at a site [6][7]. Chemicals such as heavy metals, if introduced to the environment through a specific technique, may disperse into other environmental elements, which could be due to the nature of interactions occurring in such natural system. heavy metals can interact with natural compounds in either a physical or

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chemical way, changing how they exist in nature. precipitate [8], [9]. Soil pollutants include trace elements like cadmium (Cd), arsenic (As), copper (Cu), zinc (Zn), lead (Pb), nickel (Ni), mercury (Hg), manganese (Mn), and chromium (Cr). The majority of Cr compounds found in the air are small dust particles which fall to the surfaces of land and water. It binds strongly to soil particles and only a little amount is dissolved in water, which may seep to lower soil horizons or groundwater [10], [11], [12]. In general, the issue of soil contamination has been addressed through the use of numerous techniques and plans. The two primary methodologies for remediation technologies are ex situ remediation and in situ remediation [13], [14]. Electrokinetic remediation is an economical, environmentally-friendly method for separating, migrating, and removing pollutants from soil as well as sediment under an

electric field. Variably referred to as electromigration, electrokinetic soil processing, electro reclamation, or electrochemical decontamination. These processes use electric currents to recover organic wastes, mixed inorganic species, radionuclides, heavy metals, and some organic compounds from soils and slurries. The removal of various kinds of pollutants, like organic pollutants such as thorium, phenanthrene, aniline, phenol, triclosan, and heavy metals like Cu, Cr, Pb, Zn, and Cd, was made possible by the widespread usage of electrokinetic remediation [15], [16]. Figure 1, shows the main mechanisms that occur throughout Electrokinetic Remediation are electrolysis, electroosmosis, electrophoresis, diffusion, and electromigration as the five fundamental processes in achieving decontamination of polluted environments [17].

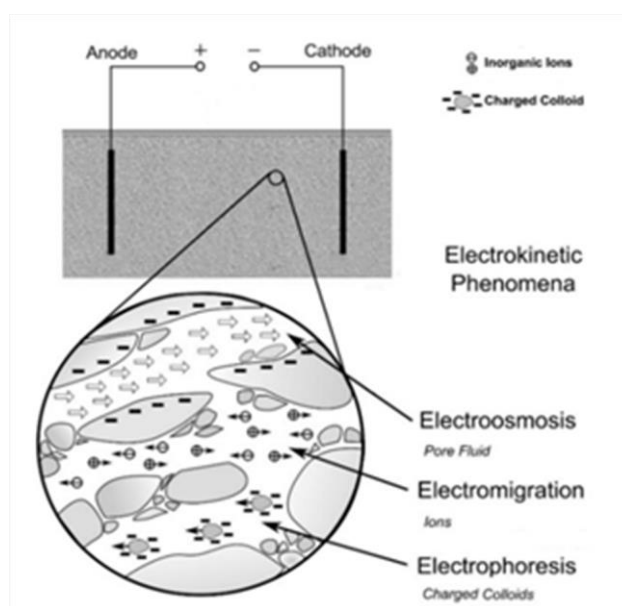


Figure 1. the main mechanisms that occur throughout Electrokinetic Remediation [17].

Through applying an electric-field to the contaminated soil or sediment, enriching the pollutants in the anode or cathode zone by electro-osmosis, electro-migration, and electro-

phoresis, KR accomplishes the remediation goal. In the process of the electrokinetic remediation, electric current is thought to go along 3 distinct paths inside the soil cell: (1) liquid-phase path-

way; (2) alternating solid–liquid phase path-way; and (3) a solid path-way through interconnected soil particles [18], [19], [20].

2. Methodology

2.1 Soil

Two types of soil were used in this study. Table 1 shows the physical and chemical

Table 1: Soils chemical and physical properties

Property	Soil I	Soil II
Particle size distribution (ASTM D 422)	97.12	80.35
Sand (%)	0.71	17.52
Silt (%)	2.12	2.11
Clay (%)	sand	Loamy sand
Texture class		
Atterberg limits (ASTMD2487)		
Liquid limit (%)	ND	ND
Plastic limit (%)	ND	ND
Plasticity index (%)	ND	ND
Specific gravity	2.66	2.72
Electric conductivity EC ($\mu\text{S}/\text{cm}$)	486	725
Organic content (%)	0.03	5.30
Primary pH	7.4	6.8
Porosity (n)	54.25	56.37

2.2 Wheat straw

Wheat straw collected from a farm located in the Al-Qazaniya region of the barley Diyala Governorate, following collection, the straws was washed with distilled water and air dried. The dried wheat straw was subsequently in a ball mill for a brief period to obtain a suitable particle size that is approximately 1-2 cm in length. Lastly, the wheat straw was placed in a perforated bag and put inside a chamber that measures 5 by 12 by 14 cm.

2.3 Chromium contaminant

To prepare the chromium contaminated solution at a concentration of 2000 mg/kg, 15.9 g of $\text{Cr}(\text{NO}_3)_2 \cdot 9\text{H}_2\text{O}$ was added and dissolved in 1000 ml of distilled water. Chromium nitrate

properties of these soils, the sandy soil was obtained from an agricultural nursery in Baghdad, while the sandy clay soil was obtained from an industrial site for leather industry in Al-Zafaraniya area in Baghdad. The two soils were air dried to preserve their properties, then they were sieved with a 2 mm sieve and stored in sealed containers.

hexahydrate ($\text{Cr}(\text{NO}_3)_2 \cdot 9\text{H}_2\text{O}$) is a blue-green solid. This material was obtained from the Scientific Equipment Offices in Bab Al-Muadham area, Baghdad.

2.4 Enhancing solution

Oxalic acid was used in this study as an enhanced reducing agent for the removal process, as it was obtained from the Scientific Equipment Office in Bab Al-Muadham area, Baghdad. 25.21 g of oxalic acid dihydrate ($\text{HO}_2\text{CCO}_2\text{H}_2 \cdot \text{H}_2\text{O}$) was dissolved in 1 liter of distilled water. Then 100 ml of oxalic acid was taken and added to 3.5 kg of soil contaminated with 200 mg/kg of chromium.

2.5 Purging solution

This research involves the use of tap water as a disinfection solution in the treatment process, and the pH level of tap water was adopted at 2. The pH and electrical conductivity values of tap water before modification for the four experiments were 7.15, 7.12, 7.06, and 7.17 for pH and 576, 587, 610, and 586 $\mu\text{S}/\text{cm}$ for electrical conductivity, respectively. Then, hydrochloric acid and sodium hydroxide obtained from the Water Treatment Laboratory at Al-Mustansiriya University-College of Engineering were used by adding it through biuret to adjust the tap water to pH 2.

2.6 Soil preparation

In the current study, the amount of pollutant in sandy soil was controlled to simulate polluted soil, a concentration of 200 mg/kg was used as the initial pollutant concentration, this polluted soil was prepared by adding 300 ml of $\text{Cr}(\text{NO}_3)_2 \cdot 9\text{H}_2\text{O}$ solution to the sandy soil by spraying until all parts of the soil were saturated. While loamy sandy soil was industrially polluted as it contained 376 mg/kg of the polluted chromium concentration.

3. Electrokinetic reactors

Figure 2. Three-dimensional diagram of the electrokinetic cell connected to the power

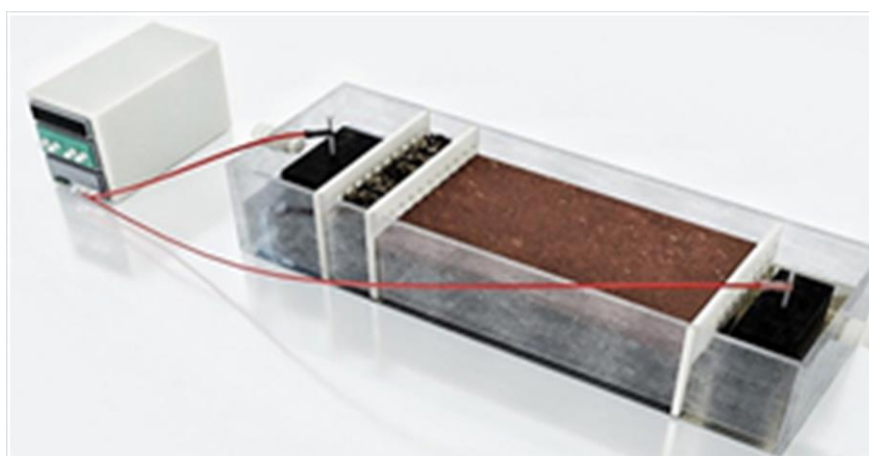


Figure 2. 3D diagram of electro-kinetic

supply through electrodes and connecting wires. The soil portion is separated from the electrodes by plastic sheets and filter papers on the anode side and an absorbent material on the cathode side. The cell was made of glass with internal dimensions ranging from 50 cm in length to 12 cm in width and 14 cm in height. The dimensions of the soil sample in the cell were determined to be 25 cm in length and 12 cm in width. Perforated plastic sheets were used as a barrier between the chambers. The sheets measure 12×14 cm and include holes with a diameter of 6 mm. At a distance of 5 cm from the soil chamber, another plastic sheet was separated to form a chamber in which the perforated bag containing wheat straw was placed as a barrier between the soil and the cathode chamber. The height of the wheat straw barrier was 14 cm. Each cell contains a valve on both sides of the cell and at a height of 10 cm from the bottom of the cell to regulate the osmotic flow. This cell used cylindrical electrodes of 10 cm height and 4 cm width made of graphite material, a nail was fixed on top of these electrodes for the purpose of facilitating electrical conduction, Whatman 40 filter papers were used in front of each plastic plate, this cell was connected by electrodes to a power source to generate a constant voltage DC current.

4. Experimental work

At the beginning of the treatment process, the cell was washed with distilled water to ensure the accuracy of the work, and the barrier was separated from the soil using filter paper. After that, the sandy soil was filled with a concentration of 200 mg/kg of the pollutant in the designated chamber with an amount of 3.5 kg and pressed homogeneously until it was unified. Then, the disinfection solution was added to the cell in an amount equal to the surface of the soil and the end of the valve, as shown in Figure 3. After ensuring that the electrodes were connected to a direct current source, the electrical source was turned on with a voltage of 1.2 volts/cm³ and the treatment process began. In this study, four experiments were conducted to determine the effect of changing the improvement solution and pH on sandy soil and sandy clay soil in removing metal pollutants (chromium). The first experiment was based on treating sandy soil with a concentration of 200 mg/kg of chromium pollutant at a voltage of 1.2 V/cm³ using tap water with a pH of 2. In the second experiment, the same conditions were adopted in the first experiment, but by adding 100 ml of oxalic acid to the cell while maintaining the pH value of 2 and the voltage of 1.2 V/cm³. In the third experiment, the sandy soil was replaced with sandy clay soil with an initial concentration of 376 mg/kg of chromium pollutant using tap water with a pH of 2. In the fourth experiment, the same standards were maintained in the third experiment, but using oxalic acid by adding 100 ml of oxalic acid to the aqueous medium. Table 2 shows the specific criteria for the laboratory standards used in the removal process. In all previous experiments, the treatment process continued for 5 consecutive days without power outage while the cell was continuously monitored. It was noted that the pH and EC values were constantly changing throughout the treatment

process as a result of hydrolysis. To maintain the pH value of 2, it was necessary to carry out continuous adjustments by adding HCl and NaOH in varying amounts during the experiment, the pH of the solution was carefully maintained at 2 using hydrochloric acid. Sodium hydroxide (NaOH) was periodically introduced to neutralize excess acid and maintain the pH balance within the reactor. This ensured the stability of the experimental conditions, as pH control is critical for optimizing the electrokinetic process and preventing adverse chemical reactions within the soil. After each experiment, the soil chamber was divided into 5 equal parts and a sample was taken from each part along with a sample of wheat straw and stored in order to examine the concentration of chromium remaining or absorbed on the wheat straw after the treatment process. To find the pH and EC values for each sample of the four experiments, 5 grams of the sample were taken and mixed with 12.5 ml of distilled water and left for an hour without stirring, which allows the soil particles to settle at the bottom. Then the pH and EC values of these samples were determined using measuring devices (HANNA type) obtained from the offices for scientific equipment in the Bab Al-Muadham area, Baghdad/Iraq. As for the process of determining the concentration of chromium in the samples after treatment, the analysis followed the EPA's Method 3050B for acid digestion of soils (EPA, 1996)[21], a mixture of nitric acid 5%, 1% and 3%, perchloric acid and hydrochloric acid, respectively, is used. This mixture is used to digest the materials in order to prepare them for chemical examination. An amount of 1 gram of dried soil sample is taken and 5 ml of the acid mixture is added to it and left for a period of not less than 12 hours. After that, this compound is exposed to a high-temperature surface until it reaches a state of almost dryness, then left to cool. This step is followed

by adding pure water to the sample, then the resulting mixture is filtered so that the final volume of the solution becomes 25 ml. Then the filtered solution is taken and entered into

the atomic device (atomic absorption spectrometer Shimadzu ASC-7000), where the device begins the process of determining the concentration of chromium.

Table 2: Operational conditions

EX. NO.	pH of solution	Chromium concentration (mg/kg)	Voltage (V/cm)	Remediation duration (in days)	Objective of the experiment
EX-1	2	200	1.2	5	pH impact
EX-2	2	376	1.2	5	pH impact
EX-3	2	200	1.2	5	OX impact
EX-4	2	376	1.2	5	OX impact

5. Results and discussion

5.1 effect of pH value on soil type

As shown in the Figure 4. we note that the concentration of chromium removed at the end of the removal process was in the sandy soil with an initial contaminant concentration of 200 mg/kg, ranging from 29 mg/kg to 59 mg/kg from the anode to the cathode. While in the loamy sandy soil with an initial concentration of 376 mg/kg and under the same experimental conditions of pH 2 and 1.2 v/cm and for a period of 5 days, we note that the concentration of chromium removed at the end of the experiment was 133 mg/kg to 191 mg/kg from the anode to the cathode, i.e., The removal efficiency for the sandy soil was 78.3% and 57.71% for the loamy sandy soil. This is due to the larger pore size of the sandy soil than it is in the sandy clay soil, which allows the free movement of chromium ions through the soil matrix. This means that chromium ions are more affected by electromotive forces, which speeds up their removal process. Sandy soil usually has lower electrical resistance than loamy sandy soil, which means that the electric current flows easily, which increases the removal efficiency. Figure 5. shows the pH values for sandy soil and

loamy sandy soil, where we notice that they range in sandy soil from 5.2 - 5.93 towards the cathode, while for loamy sandy soil, the pH values ranged between 6.3 - 6.9 from the anode to the cathode, which is greater than in sandy soil. To explain this difference in pH values between sandy soil and sandy clay soil, it can be attributed to several factors, including the nature of the chemical reactions that occur during the process. The oxidation reactions that occur at the anode and the reduction reactions that occur at the cathode lead to the formation of acids. These acids affect the pH value of the soil surrounding the anode. Figure 6. shows the effect of soils on the EC value. We notice that in sandy soil, the EC values were lower than in sandy clay soil, as they were 2.78 to 1.29 $\mu\text{s/cm}$ from anode to cathode in sandy soil and 6.9 to 6.1 $\mu\text{s/cm}$ from anode to cathode in sandy loam soil. This is due to the large pore size of sandy soil, which reduces the contact area between dissolved ions. Consequently, the movement of ions is easier and less restricted, leading to a decrease in electrical conductivity.

5.2 Effect of oxalic acid (OX) on soil type

The third and fourth experiments were conducted to study the effect of oxalic acid on sandy soil and loamy sandy soil. As shown in

Figure 4. at the end of the remediation process in the presence of oxalic acid, the concentration of chromium removed for sandy soil with an initial contaminant concentration of 200 mg/kg ranged from 27 mg/kg to 47 mg/kg from anode to cathode with a removal efficiency of 81.9%. While in loamy sandy soil with an initial concentration of 376 mg/kg and under the same conditions of the third experiment, the concentration of chromium removed was 109 mg/kg to 156 mg/kg from anode to cathode with a removal efficiency of 65.26%. This difference

in removal between the two soils is due to the difference in the mineral composition of loamy sandy soil from sandy soil, as this difference affects the soil's ability to absorb and interact with chromium ions. In addition, oxalic acid increases the solubility of chromium, which facilitates its removal, in addition to forming stable complexes with chromium ions, thus reducing their activity. In addition to other factors that characterize sandy soil, such as larger pore size and lower electrical resistance.

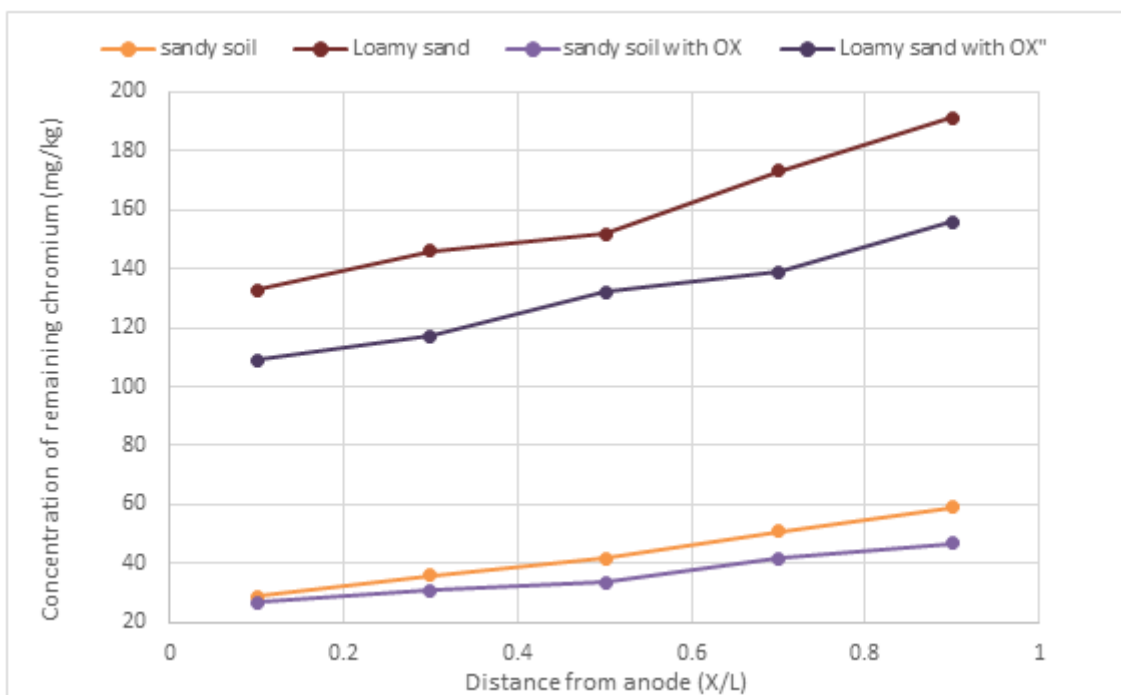


Figure 4. Chromium concentration (mg/kg) at pH 2 with and without oxalic acid.

In Figure 5. we note that the pH values of sandy soil and loamy sandy soil range from 4.9-5.6 to 5.6-6.233 from the anode towards the cathode for sandy soil and loamy sandy soil. We note from these results that the pH values in loamy sandy soil are greater than in sandy soil, due to the chemical reactions that occur more in sandy

soil and in addition to the presence of oxalic acid, all of which are factors that affect the pH value of the soil. The electrical conductivity ranged from 7.2 to 6.6 $\mu\text{s}/\text{cm}$ near the anode to about 6.47 to 5.7 $\mu\text{s}/\text{cm}$ near the cathode for sandy and clayey soils, respectively.

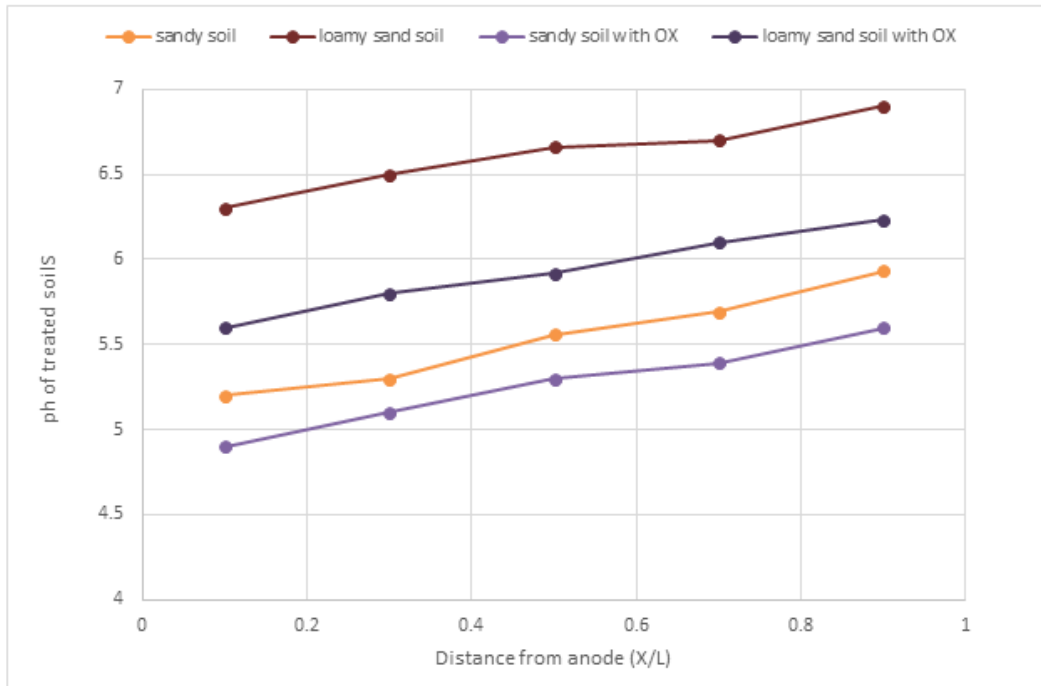


Figure 5. pH of treated soils at pH 2 with and without oxalic acid.

An increase in electrical conductivity was observed as shown in Figure 6. for clayey soil in the presence of oxalic acid, compared to clayey soil with tap water only. This is due to the dissolution of oxalic ions in water, which increases the free ions of the soil. This increase in the number of ions leads to an increase in electrical conductivity.

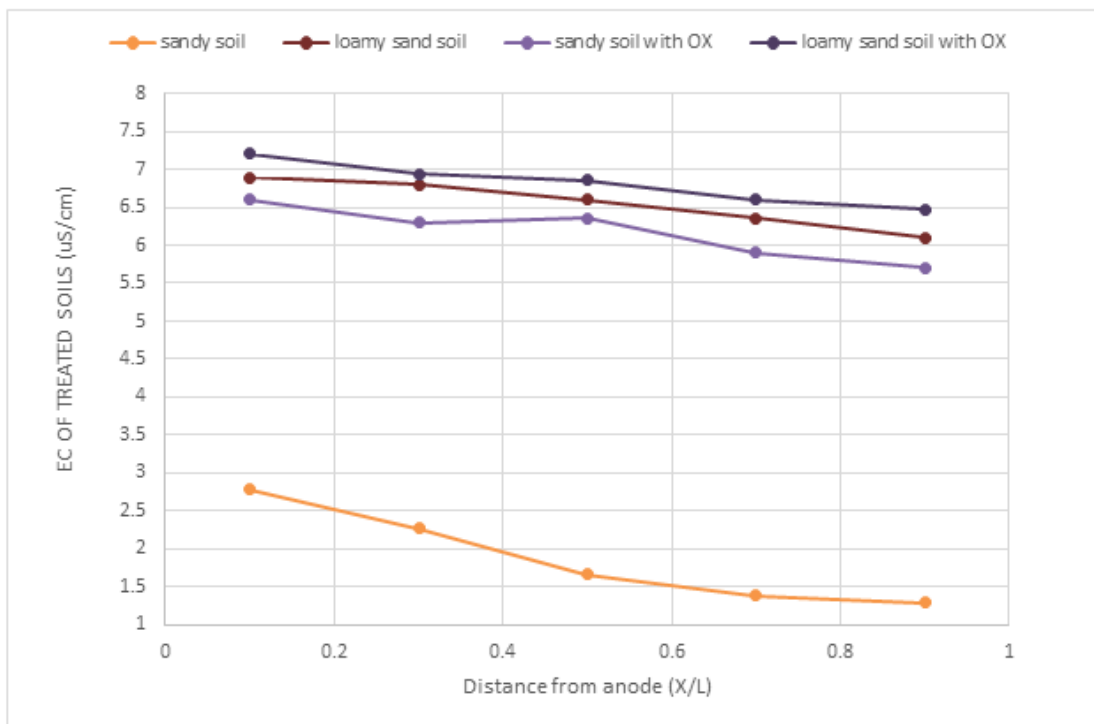


Figure 6. EC of treated soils at pH 2 with and without oxalic acid.

7. Removal Efficiency of chromium

Figure 7. shows the results of chromium removal under different experimental conditions. The removal efficiency was calculated based on the mass of the initial and residual pollutants in the contaminated soil, as shown in Equation (1)[22].

$$\eta \% = \frac{\text{Initial conc.} - \text{Residual conc.}}{\text{Initial conc.}} * 100 \dots (1)$$

Where:

η %: Removal Efficiency

We note that the highest removal efficiency was in experiment 3 at 81.9% when OX acid was used as an enhancement solution at a rate of 100 ml in the experiment 3 sandy soil. In contrast, the loamy sandy soil showed a greater removal

efficiency of the pollutant (chromium) 65.26% in the experiment 4 when OX acid was used as an enhancement compared to the experiment 2 without OX acid 57.71%. This improvement is due to the strong adhesion properties of OX acid, environmental friendliness, and minimal interaction with the soil. We note in Figure 4 that the removal efficiency 57.71% in the loamy sandy soil in the experiment 2 experiment was lower compared to the 78.3% experiment 1 in the sandy soil. This is due to the fact that the loamy sandy soil has adsorption forces, which hinder the movement of chromium. This leads to an increase in the salt content in this soil, and an increase in the soil's buffering capacity. This increase in the ability to delay the formation of the acid front, which will lead to lower rates of pollutant removal in these soils.

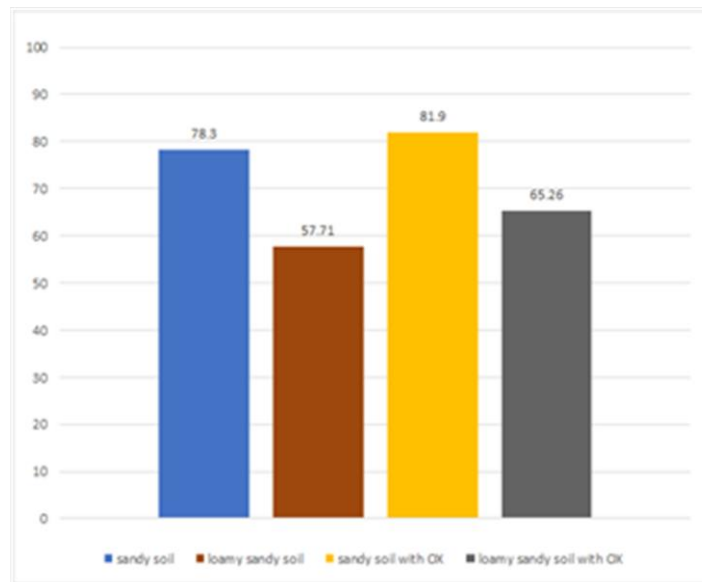


Figure 7. Removal Efficiency of Chromium at different parameters

7. Conclusions

The electrokinetic remediation technique proved to be highly effective in treating soil contaminated with chromium, as demonstrated by the varying levels of removal efficiency

across different soil types in this study. The results indicate that soil composition plays a critical role in the migration and removal of contaminants. In sandy soil, the electrokinetic process achieved removal efficiencies of 78.3%

and 81.9% in experiments 1 and 3, respectively, while in loamy sandy soil, removal efficiencies were slightly lower at 57.71% and 65.26% in experiments 2 and 4. The presence of oxalic acid as an enhancing agent significantly improved the removal process, particularly in sandy soils, where the efficiency increased from 78.3% without oxalic acid to 81.9% with its inclusion. A similar pattern was observed in loamy sandy soils, where the addition of oxalic acid raised the removal efficiency from 57.71% to 65.26%. The success of these experiments aligns with findings from previous studies, despite the lack of explicit comparisons in the results section. For instance, Han et al. (2021) reported chromium removal efficiencies of approximately 75% under similar electrokinetic conditions but without using an enhancing agent. In contrast, the use of oxalic acid in this study not only improved removal rates but also enhanced the solubility of chromium, facilitating its migration and extraction from the soil. This highlights the importance of optimizing the remediation process by considering both soil type and the use of enhancing solutions like oxalic acid. These findings confirm the effectiveness of electrokinetic remediation, particularly for large-scale soil decontamination projects, though future studies should provide direct comparisons to further validate the technique's success.

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