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ELECTROKINETIC REMEDIATION TECHNOLOGY OF HEAVY METAL CONTAMINATED SOIL BASED ON IMPROVING SOIL ELECTRICAL CONDUCTIVITY

An electrokinetic remediation technique taking Cr(VI) as an example is proposed to improve the conductivity of contaminated soil, which significantly increases the current density in the soil. The improvement of soil conductivity was achieved by continuous spraying of NaCl solution with a concentration of $4 \text{ g}\cdot\text{dm}^{-3}$ on the soil surface. The distances of electrode pairs were 2.0 m and 1.5 m, respectively. The heavy metal-contaminated soil thickness was 25 cm, and the DC power supply voltage was 90 V. The experiment demonstrated that under the condition of continuous spraying of NaCl solution on the soil surface, the current density variation was related to the salt content in the soil, and the current density in the soil generally increased linearly with time. The effectiveness of soil remediation is related to the electric field strength and current density, and there exists an optimal electric field that can reduce the heavy metal content in the soil at any point by minimizing the electric field strength and current density. Most of the heavy metals can be concentrated within a diameter of about 15 cm around the anode under the optimal electric field, which can be remediated after removing the soil.

1. INTRODUCTION

At present, many methods to repair heavy metal-contaminated soil are known although the remediation of heavy metal pollutants is difficult as they occur in various forms, have weak migration ability, high toxicity, and long retention time [1, 2]. Microbial remediation techniques include the reduction of the toxicity of heavy metals in soil by changing the redox status of heavy metals by microorganisms [3]. In addition, the strong affinity of microorganisms for heavy metals can be utilized to enrich various

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heavy metals and reduce the heavy metal content in soil and groundwater [4, 5]. Phytoremediation technology uses the roots of hyperaccumulated heavy metal plants to absorb large amounts of soluble and insoluble heavy metals from the soil. Chemical remediation technology involves the addition of modifiers to the soil to reduce the biological effectiveness of heavy metals through adsorption, redox, antagonism or precipitation of heavy metals [6].

Electrothermal remediation technology uses high-frequency voltage to heat the soil and accelerate the separation of volatile heavy metal contaminants (mainly Hg and Se) from the soil [7]. Soil leaching technology is the transfer of heavy metals from the solid phase to the liquid phase in soil using leaching solutions (water or reagents that increase the solubility of heavy metals), or the use of strong acids leaching solution to change the surface charge of soil particles so that heavy metals on the surface of soil particles are adsorbed into the soil in the liquid phase by desorption and then recovered for wastewater treatment.

Electrokinetic remediation technology is that the metal ions or metal complex ions in soil separately migrate electrodes when the cathode and the anode are directly inserted into the soil to form an electric field, and eventually accumulate around the electrodes and finally be removed [8, 9]. This electrokinetic remediation technology is especially suitable for clay and silt and also has applications in wastewater [10]. The research of electrokinetic remediation has made great progress in the world but most of the results are not suitable for practical application [11].

Nanomaterials have high adsorption capacity and are also used in heavy metal-contaminated soil remediation [12, 13]. Citric acid and polyaspartic acid are used as new electrolytes to improve removal efficiency [14]. The combination of electrokinetics and solar energy is feasible and effective to some extent for the remediation of Cr-contaminated soil [15]. Each of the above soil remediation technologies has advantages and disadvantages, so there are great limitations in practical application [16].

Electrokinetic remediation is an in situ technique that can simultaneously remove metals and organic pollutants. The advantages of electrokinetic remediation in the removal of chromium are apparent, as follows: compared with the ectopic remediation technology, its impact on the landscape and architecture is negligible. It will not destroy the structure of the soil itself and will not be affected by low soil permeability. No secondary pollutants are generated during the remediation process, and the method is effective for both saturated and unsaturated layers.

The key problem for the wide application of electrokinetic remediation technology is how to improve soil conductivity. In this paper, it is proposed that the soil conductivity can be continuously increased by spraying suitable conductive liquid on the soil surface continuously or intermittently during the soil remediation process. With the gradual penetration of good conductive liquid to deep soil, the soil conductivity gradually increases from the surface to the deep, the current density in the soil also increases continuously, and then the heavy metal ions in soil acquire greater migration power, and

ultimately we achieve the ideal effect of soil remediation. This new remediation technique was successful in the experiment, in which NaCl solution was chosen as a liquid with good electrical conductivity.

2. CHOICE OF GOOD CONDUCTIVE LIQUID

Two factors should be considered in the selection of conductive fluid. Firstly, it should not produce secondary pollution to the soil. In addition, it can maximize the conductivity of the soil. As long as the water content of the soil increases, the soil conductivity will also increase, so tap water is also a choice for conductive fluids [17]. However, the conductivity of tap water itself is not high enough (typically $200\text{--}400\ \mu\text{S}\cdot\text{cm}^{-1}$) to substantially increase the conductivity of soil, while NaCl can significantly increase the conductivity of conductive liquids. As shown in Table 1, the conductivity of tap water with different NaCl content was measured using a temperature conductivity meter (temperature correction function). In the experiment, the solution of $4\ \text{g}\cdot\text{dm}^{-3}$ NaCl was used as the conductivity liquid.

Table 1

The conductivity of tap water with different NaCl content

Concentration of NaCl in tap water [g/dm^3]	Conductivity of NaCl solution [$\mu\text{S}/\text{cm}$]
0	194
2	2160
3	4320
4	5848
5	7850
6	9888

Excessive Na^+ ions adsorbed on the surface of soil particles for a long time will also have a certain impact on the soil texture. Therefore, it is not appropriate to use excessive NaCl solution as conductive fluid. At the same time, a certain amount of tap water can be sprayed a certain time after the remediation and then Na^+ ions can be washed off the surface of the soil particles, which can minimize their impact on the soil texture.

3. EXPERIMENTAL DESIGN

In the experiment, K_2CrO_4 was selected as the heavy metal contamination component, silty clay was selected as the soil, and K_2CrO_4 was evenly mixed in clay as the heavy metal contaminated soil. The experimental apparatus for storing contaminated

soil was made of plexiglass ($205 \times 150 \times 40$ cm). The thickness of contaminated soil set in this experiment was 25 cm.

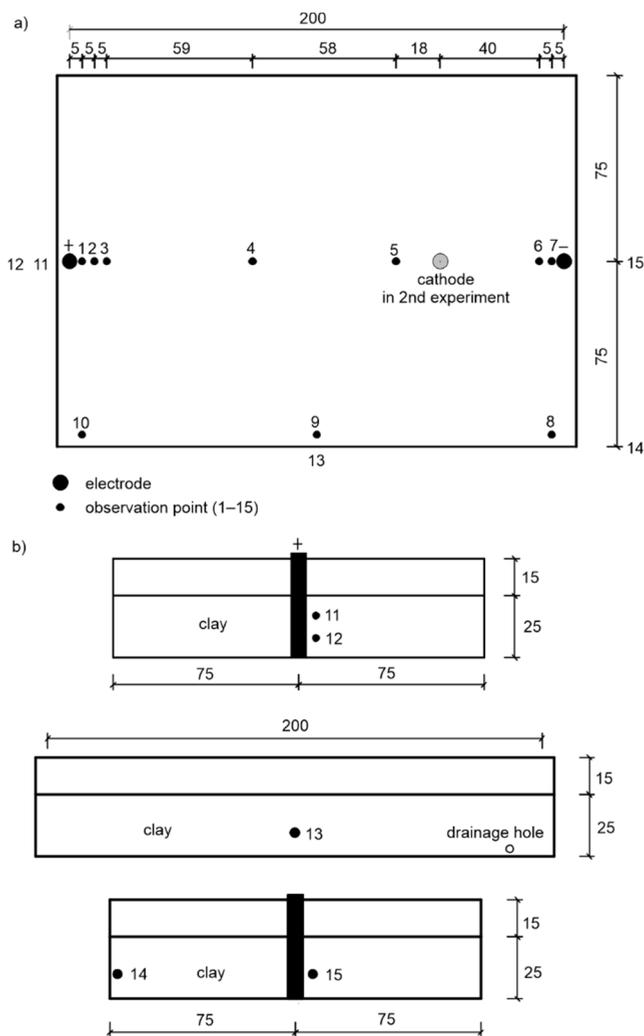


Fig. 1. Layout of observation points: a) horizontal (1–10), b) vertical (11–15)

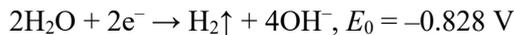
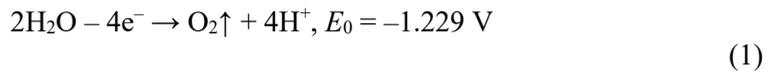
Preparation of contaminated soil. K_2CrO_4 solution was prepared in advance at a concentration of $4 \text{ g} \cdot \text{dm}^{-3}$ for a total of 14 dm^{-3} , and then the K_2CrO_4 solution was mixed in the field with silty clay. The mixing process was as follows: the original soil sample was taken in the field about 1.5 m^3 mashed and spread out, and the $1/3$ part of K_2CrO_4 solution was sprayed on the crushed soil. Then the soil was mixed up and down so that the soil and the K_2CrO_4 solution were mixed as thoroughly as possible and the

process was repeated for the other 2/3 K_2CrO_4 solution 2 times. The soil thoroughly mixed with all K_2CrO_4 solution was transported to the laboratory and slowly stratified and pressed into the designed plexiglass frame. Electrodes, conductive liquid spraying equipment and pH controllers were then installed separately. The contaminated soil in the plexiglass frame was left for at least half a month, and then the electrodes were energized to create an electric field to start the remediation.

Electrode, electric field and current device. A hollow graphite electrode with an outer diameter of 35 mm and an inner diameter of 17 mm was used, and holes of $\varnothing 4$ mm were distributed on all the sidewalls. The holes on the sidewall were arranged to facilitate the escape of gas generated by water electrolysis around the electrode. The 220V AC power is transformed into a constant power supply DC voltage 90 V by a step-down transformer. The electrode distance in the earlier experiment was 2.0 m, and that in the latter experiment was 1.5 m (Fig. 1a). The ammeter specification 5A is fixed to the step-down transformer.

Observation points. There were 1–15 observation points to measure the heavy metal content in the soil during electrokinetic remediation. The observation points 1–10 were located on the top of the soil (Fig. 1a) to record the remediation effect at different locations on the plane and the migration rule of heavy metals. The observation points from 11 to 15 were arranged on the side (Fig. 1b) to trace the remediation effect and heavy metal migration rule at various depths. The observation points of 1–3 and 6–7 were close to the cathode and the anode, respectively to check the enrichment degree of heavy metal ions towards the electrode under the action of the electric field, and ultimately consider the scope of soil excavation after enrichment.

pH control. Soil pH is an essential factor affecting the effect of electrokinetic remediation of heavy metal contaminated soils [18, 19]. Under the action of the electric field, OH^- ions around the cathode, and H^+ ions around the anode will be produced by the interaction of electrodes and soil moisture (see the following chemical reaction). Thus, acid-base titration devices were designed in the experiment to let the OH^- ions around the cathode neutralized by the acid titration solution (citric acid) and the H^+ ions around the anode neutralized by the alkaline titration solution (NaOH).



Spraying facility of conductive liquid. The spraying facility consisted of sprinkler heads, a hose, a micro-pump, a conductive liquid container, and a timer, which were installed above the soil. The conductive fluid was ejected from all sprinkler heads through a hose after pressurizing the micro-pump and sprayed on the soil surface. The timer was used to control the time interval of spraying. The time interval set in the experiment was

15 minutes, that is, spraying was twice an hour, to ensure that the soil surface is always in a wet state. The wet cover can expand deep into the soil so that the soil moisture content and conductivity gradually increase.

Determination of Cr(VI) concentration and removal rate in the soil. In the presence of an electric field, Cr(VI) ions in the soil undergo only physical migration and no chemical changes occur. The valence state of Cr(VI) ions remains unchanged. Therefore, the content of Cr(VI) in soil was determined by diphenylcarbazide spectrophotometry using the ultraviolet-visible spectrophotometer (UV-2600i, Shimadzu, Japan). The determination of Cr(VI) in soil was done in two steps. The first step was the pretreatment of contaminated soil. First, the soil sample was air-dried to remove large particles, then the soil was crushed and passed through a 10-mesh sieve and a 100-mesh sieve (aperture 0.149 mm) in that order. In addition, nitric, hydrochloric, hydrofluoric, and perchloric acids were used to remove various organic substances from the soil in turn, and the treated soil samples were dissolved in distilled water to form a solution containing Cr(VI). In the second step, the concentration of Cr(VI), which was linearly related to the absorbance measured at a wavelength of 540 nm, was determined spectrophotometrically. The linear dependence (standard curve) of absorbance on the Cr(VI) concentration was shown in Fig. 2, where the Cr(VI) concentration has been converted to Cr(VI) concentration per unit of soil mass.

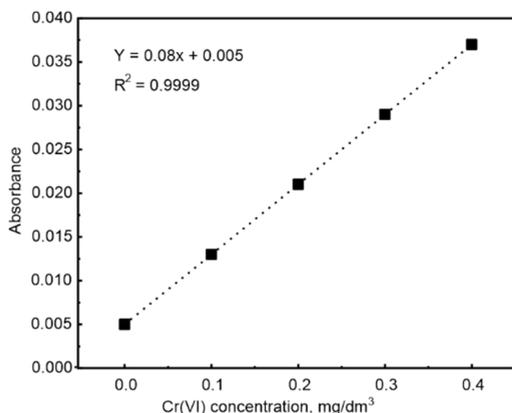


Fig. 2. Dependence of the absorbance on Cr(VI) concentration

All results in this study were mean values \pm standard deviation from three independent samples with an experimental error below 10%. All the electrokinetic experiments were carried out at the same room temperature (20 ± 3 °C). The removal rate ρ , %, of Cr(VI) at each observation point can be calculated according to:

$$\rho = \frac{\sigma_1 - \sigma_2}{\sigma_1 - \sigma_0} \times 100\% \quad (2)$$

where: σ_1 , σ_2 , σ_0 – initial, final, and background contents of Cr(VI), $\mu \cdot \text{g}^{-1}$, in the soil.

4. RESULTS AND DISCUSSION

The graphite electrodes were energized and the soil samples were collected from the observation points for testing the next day and the ammeter readings were recorded. The whole experiment was conducted in two phases: the first phase lasted 15 days with an electrode distance of 2.0 m, which was terminated due to unsatisfactory remediation results. At the same time, we modified the electrode spacing to 1.5m and began the second phase of the experiment for 9 days.

4.1. CURRENT DENSITY

The variation of the current density during the two experimental phases, which were calculated from the ammeter readings and the cross-sectional area of the soil, were shown in Fig. 3.

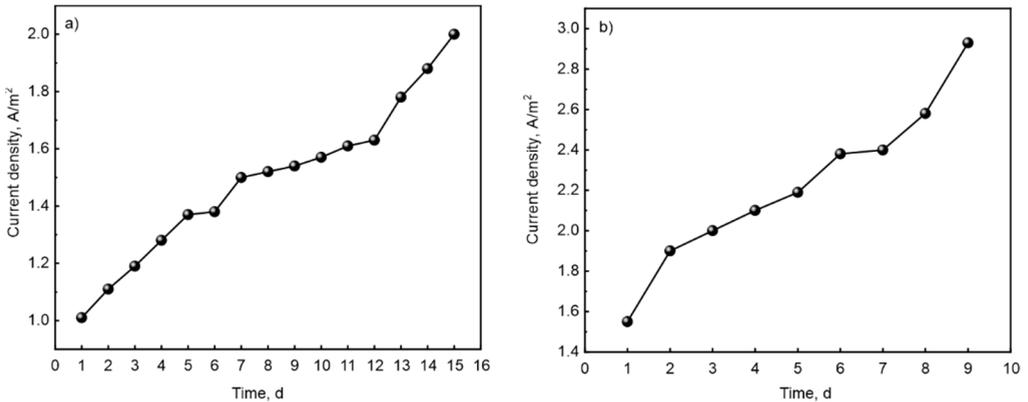


Fig. 3. Current densities in the first (a) and second (b) period

As shown in Fig. 3a, the first phase of the experiment shows a gradual increase in the current density of the soil between the two graphite electrodes, which is consistent with the transformation trend of the second stage (Fig. 3b). These results indicate that continuous spraying of the conductive solution on the soil surface can significantly increase the soil conductivity. The current density value in the second period (1.55–2.93 A·m⁻²) was significantly higher than that in the first period (1.01–2.00 A·m⁻²), which were attributed to two reasons. First of all, the electrode distance in the second period of the experiment was changed from 2.0 m to 1.5 m, which reduced the soil resistance between the two electrodes, and the soil resistance decreased while the voltage remained constant. Secondly, after the spraying of the first period, the soil salt content increased while the soil conductivity increased at the same time. These results suggested that electrode distance and the fluid with good conductivity can synergistically increase current density.

The curves in Figs. 3A and 3B were characterized by 3 different sections: a significant change in the pregradient, a slowing down of the middle gradient, and an increase in the post-gradient again, which was related to the process of increasing saline soil content. Before the experiment, the soil water content was the lowest, especially in the top layer of the soil. After salt spraying, the surface soil first acquired salt and the soil resistance decreased significantly, leading to a significant increase in the current density gradient. As the salt spraying continued, the salt in the soil increased and spread to deeper soil, but the swelling process was relatively slow and the current density gradient decreased accordingly. The process usually ends when the brine reaches the bottom of the soil. After that, the salt content at the bottom of the soil increases significantly as more and more salt reaches the bottom of the soil. At this point, the soil resistance decreased again while the current density gradient increased.

4.2. Cr(VI) CONCENTRATION OF OBSERVATION POINT

Four in-situ soil samples were collected before the addition of K_2CrO_4 solution to the pulverized clay. The content of Cr(VI) in the four in situ soil samples was measured and the results are presented in Table 2.

Table 2

Cr(VI) content in silty clay [$\mu\text{g/g}$]

Observation point	Cr(VI) content	Observation point	Cr(VI) content
1	4.1±0.2	9	21.0±1.2
2	6.5±0.3	10	25.0±1.5
3	6.5±0.3	11	25.0±1.5
4	8.0±0.4	12	27.0±1.6
5	30.0±2	13	6.5±0.3
6	12.0±0.6	14	13.8±0.6
7	17.0±0.8	15	5.3±0.2
8	12.0±0.6		
No. of silty clay in situ	Cr(VI) content [$\mu\text{g/g}$]	No. of silty clay in situ	Cr(VI) content [$\mu\text{g/g}$]
1	6.1±0.2	3	3.7±0.2
2	2.5±0.1	4	3.0±0.1

After the addition of K_2CrO_4 solution to the pulverized clay, the initial Cr(VI) content in soil samples was measured from 15 observation points and the results are shown in Table 2. The content of Cr(VI) ions in the in-situ soil samples was extremely low and uniformly distributed. The content of Cr(VI) in the soil samples of the 15 observation points ranged from 4.1 to 30.0 $\mu\text{g/g}$. The content of Cr(VI) in observation points 1, 2, 3, 13 and 15 was less than 6.5 $\mu\text{g/g}$, which was comparable to the content of Cr(VI) in

the in-situ soil samples. These results indicated that K_2CrO_4 solution mixed in clay has a certain degree of non-homogeneity, which is due to the fact that clay is mostly blocky, and different block sizes lead to different Cr(VI) contents, Thus it is normal that the Cr(VI) content of the surface and the interior of the same block are different. However, this does not affect the remediation process; as long as there exists charge in the electric field, Cr(VI) can migrate towards the electrode and thus produce a remediation effect.

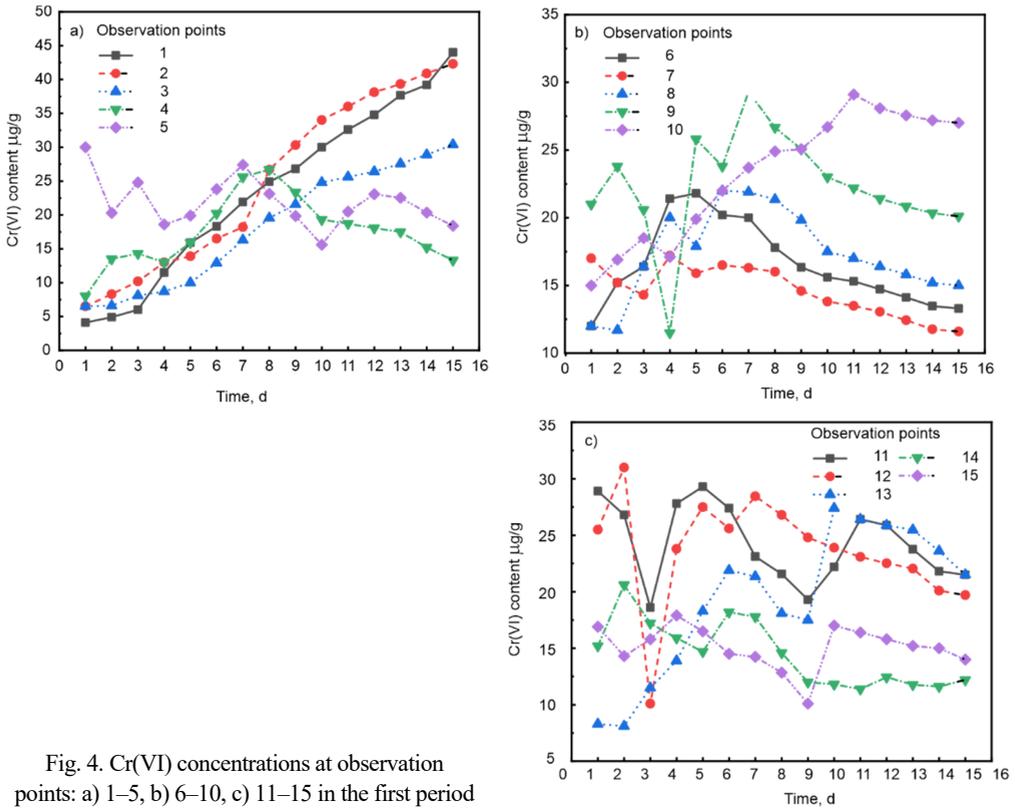


Fig. 4. Cr(VI) concentrations at observation points: a) 1–5, b) 6–10, c) 11–15 in the first period

The results of remediation experiments in the first period are shown in Fig. 4. Overall, the migration of Cr(VI) in the soil to the anode was very slow, which can be ascribed to the lack of the electric field strength and current density. In addition, the migration kinetics of Cr(VI) in soils of different depths and regions varied greatly, especially the migration kinetics of Cr(VI) in deeper soils was insufficient, so the phase I experiments were suspended.

In the second period of the experiment, the electrode distance was changed from 2.0 to 1.5 m, while other conditions remained the same. In addition, the number of observation points was reduced and the experimental results are shown in Fig. 5. The experiments in the second stage exhibited that the electric field and current density provided

enough motivation for the migration of Cr(VI) to any location of the soil so that the Cr(VI) migrated and concentrated toward the anode from all directions. It is obvious that the content of Cr(VI) in the soil decreases continuously and even reaches the initial value of Cr(VI) content.

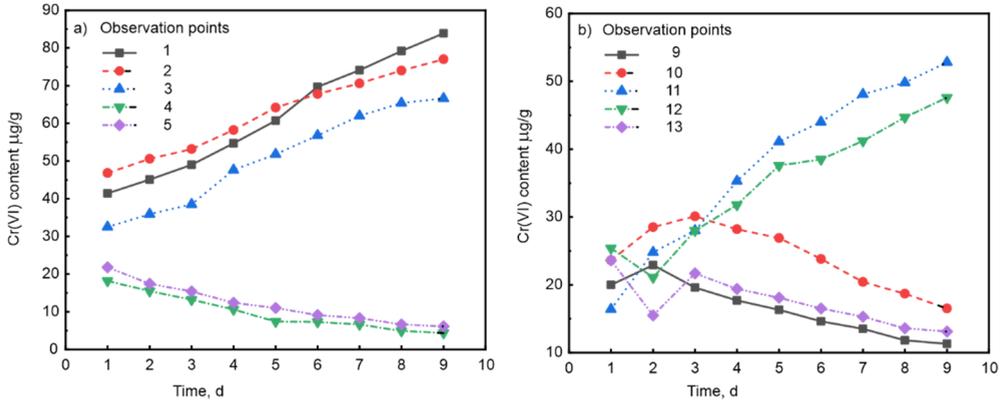


Fig. 5. Cr(VI) concentrations at observation points: a) 1–5, b) 9–13 in the second period

Table 3

Removal rate of Cr(VI) at the observation point

Observation point	Cr(VI) initial content [µg/g]	Cr(VI) final content [µg/g]	Removal rate [%]
4	12.2±0.2	4.4±0.1	93.1±0.5
5	17.8±0.3	6.1±0.1	83.7±0.8
9	19.7±0.3	11.3±0.2	52.9±2.3
10	25.2±0.4	16.5±0.2	40.7±2.8
13	21.5±0.4	40.7±0.8	47.5±2.6

The removal rates of Cr(VI) in five observation points are given in Table 3. It was evident that observation points 4 and 5 exhibited the highest removal rates and the final values of Cr(VI) content were close to the initial values. In addition, the removal rates of observation points 9, 10, and 13 were around 50%, which indicated a successful remediation.

4.3. HEAVY METAL MIGRATION IN SOIL UNDER THE ACTION OF ELECTRIC FIELD AND OPTIMAL ELECTRIC FIELD PROPOSED

In the first experimental stage, when the electrode distance was 2.0 m, and the electric field intensity and current density were low, a peak was observed at each observation point except for observation points 1–3, 15 from the Cr(VI) content variation curve. In the second experimental stage, the electrode distance was adjusted to 1.5 m, and the

electric field intensity and current density were relatively increased. At this time, only observation points 9 and 10 displayed peaks in the Cr(VI) content variation curves, which can be attributed to their location at the edge of the electric field, leading to their relatively low electric field intensity and current density. Based on the results of the two experiments, it is evident that the transport of heavy metals in soil under the action of electric fields is a dynamic process. Cr(VI) ions at any point leave while receiving the arrival of nearby Cr(VI) ions. The amount of transferred Cr(VI) ions is smaller than the amount of accepted Cr(VI) ions with the low electric field strength and current density, leading to the increase of Cr(VI) content. On the contrary, the amount of transferred Cr(VI) ions is higher than the amount of accepted Cr(VI) ions when the electric field and current density are enhanced, resulting in the decrease of Cr(VI) content. Therefore, we propose the concept of optimum electric field for remediation of heavy metal-contaminated soil by electrokinetic method, which refers to the optimal electric field strength and optimal current density. The optimum electric field for remediation of heavy metal-contaminated soils is regarded as the minimum electric field intensity and the minimum current density, which can drive the content of heavy metals at any point in the soil descend continuously from the beginning of the experiment. In the remediation experiments of the first and second periods, the electric field intensity and current density do not reach the optimum electric field, but the electric field in the second period is very close to the optimum electric field.

5. CONCLUSIONS

Continuous spraying of NaCl solution on the contaminated soil can greatly increase its electrical conductivity and the soil current density. Meanwhile, the electrode distance can be set to more than 1.0 m, which makes the electrokinetic remediation method have the potential for practical application.

Under the condition of continuous spraying NaCl solution, the current density in soil increased linearly, and the gradient of the current density versus time was related to salt content.

The soil remediation effect is closely related to the electric field intensity and current density. The optimum electric field is the minimum electric field strength and minimum current density that can cause a continuous reduction of heavy metal content at any point in the soil.

Under the action of the optimal electric field, Cr(VI) ions in the soil can be migrated and concentrated in a range of about 15 cm in diameter around the anode, and the soil can be remediated by removing these soil around the anode.

The removal rate of heavy metals in soil is related to the time of electric field action. The longer the time of electric field action leads to higher removal rate of heavy metals.

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