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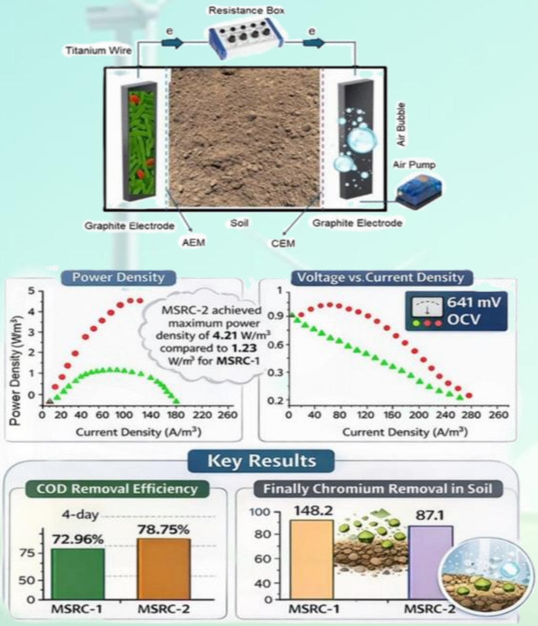
Green Energy Generation and Sustainable Chromium Remediation in MSRC by Focusing on the Role of Microbial Bio-Supports

Marzie Razavi

Highlights

- **A hybrid microbial fuel cell–electrokinetic system was developed to simultaneously remediate hexavalent chromium soil and generate bioelectricity.**
- **Electrodes modified with activated carbon granules significantly outperformed plain ones, increasing voltage to 641 mV and power density to 4.21 W/m³.**
- **The modified system achieved 83.5% COD removal and reduced soil chromium concentrations to 68–99 µg/g.**
- **This approach proves to be an efficient, eco-friendly solution for simultaneous energy recovery and in-situ soil remediation.**

Graphical Abstract



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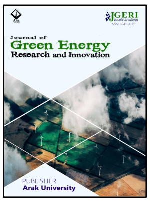

Green Energy Generation and Sustainable Chromium Remediation in MSRC by Focusing on the Role of Microbial Bio-Supports

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ABSTRACT

In this study, a hybrid microbial fuel cell–electrokinetic remediation system (MSRC) was developed to remediate soil contaminated with hexavalent chromium while simultaneously generating bioelectricity. Two configurations were compared: MSRC-1 with plain graphite electrodes and MSRC-2 with graphite electrodes modified using activated carbon granules. The results demonstrated that electrode modification significantly enhanced biofilm development and electron transfer, leading to higher system efficiency. MSRC-2 achieved an open-circuit voltage of 641 mV, a maximum power density of 4.21 W/m^3, and 83.5% COD removal, compared to 406 mV, 1.23 W/m^3, and 62.3% in MSRC-1. Chromium migration toward the cathode was also more effective in MSRC-2, reducing soil concentrations to 68–99 μg/g. These findings highlight the novelty of integrating activated-carbon-modified electrodes into a microbial fuel cell–electrokinetic system, offering an efficient and environmentally friendly approach for simultaneous energy recovery and in-situ remediation of Cr (VI)-polluted soils.

1. Introduction

The discharge of wastewater containing heavy metals into receiving waters generally results in physical, chemical, and biological alterations in the environment [1,2]. The nature and extent of these changes depend largely on the type and concentration of heavy metals present in both the water and soil [3]. Due to their toxicity, these metals pose significant threats to the health of living organisms [4]. Furthermore, because these compounds cannot be degraded by microorganisms in soil, they persist for extended periods, making their removal from aquatic environments crucial [5]. The heavy metal chromium is widely distributed in soil, sediment, water, and biological materials and is considered a serious hazard to them [6-10]. The main source of chromium entering the environment is the output of metal plating processes, textile and paint industries, leather manufacturing, and its direct disposal [11,12]. Chromium exists in two forms in soil: trivalent chromium and hexavalent chromium [13]. Trivalent chromium is essential for the body in small amounts, but it has been shown that hexavalent chromium has carcinogenic effects in living organisms [14]. According to studies by Costa and Klein in 2006 and also by Owlad et al. in 2009, it has been shown that trivalent chromium can be converted to hexavalent chromium by oxidation and may accumulate in aquatic habitats through bioaccumulation processes [15,16]. Therefore, chromium removal is very critical for human health and the environment. Various soil remediation technologies have been developed to remove this contaminant based on physicochemical, thermal, and biological methods [17]. One of the physicochemical reduction techniques is the electrokinetic remediation method. Electrokinetic remediation is a well-known environmental method for removing contaminants from porous matrices with low permeability, soils, sediments, and wastes [18]. In this technique, organic materials and heavy metals are removed from the contaminated environment or transferred to another place using a weak electric field through various charge transfer mechanisms, charged particle operations, and the effects of electric potential on fluid transport within the pores of the media [19].

However, the high consumption of electrical energy is one of the limitations of the electrokinetic method in soil and sediment remediation [20]. Also, electrolytic reactions near the electrodes cause changes in the pH of the soil, such that the pH near the anode decreases sharply and increases sharply near the cathode [21]. This can cause a cemented zone near the cathode electrode and greatly slow down the migration of pollutants there [17]. For this reason, researchers have sought to provide useful methods combined with the EKR method to reduce the costs of electricity consumption in the process and, by controlling the pH, prevent cementation conditions and interruption of electroosmosis flow in the cathode zone, and simultaneously remediate contaminated soil or sediment.

Among the methods combined with EKR, solar energy and microbial fuel cells can be mentioned [20, 22-25]. In recent years, microbial fuel cells have been proposed as an emerging technology in the production of clean water [26-28] and green energy [28,29] using waste materials and have provided a new way to use renewable and low-cost energy sources [30-32]. The main obstacle in the microbial fuel cell process is the high cost of the materials used in its construction [33]. Since the shape and design of the electrodes are key factors in reducing the price of the electrode material, many efforts have been made to provide cheaper electrode materials and higher energy output [34-36]. Carbon materials are considered one of the most suitable options for electrodes in microbial fuel cells due to their high chemical resistance, low cost, and sufficient electrical conductivity [36]. It is noteworthy that the efficiency of the microbial fuel cell is increased by optimizing the formation of biofilm on the electrode, increasing the rate of organic matter decomposition, and electron transfer between the bacteria and the anode electrode. Achieving all of the above is possible by modifying the electrode surface and has become a new research field among researchers [37-39]. The first concept of a three-compartment microbial fuel cell design with an electrokinetic process for the purification of vegetable garden soil was proposed by Chen et al. in 2015, which resulted in the removal of 25% and 18% of zinc and cadmium metals, respectively [40].

Considering the challenges in the electrokinetic remediation process, including high energy consumption, severe pH changes in the vicinity of the electrodes, and reduced process efficiency due to phenomena such as the formation of a cement zone near the cathode, the need to utilize new and combined technologies to improve the efficiency and sustainability of the remediation process is increasingly felt. In the meantime, the use of microbial fuel cells as a renewable and low-cost energy source has opened a new horizon in the development of environmental cleaning technologies. These systems, with the ability to simultaneously utilize organic materials in the environment to generate electricity and remove pollutants, have paved the way for the design of more efficient combined processes. Considering the potential of microbial fuel cells in providing the energy required for the electrokinetic process, and on the other hand, the lack of comprehensive studies on the combination of these two technologies in removing heavy metals from contaminated soil, investigating and evaluating the efficiency of combined remediation systems under real conditions is considered an important research gap. According to the findings of the researchers of this study, no study has been conducted so far in the field of contaminated soil cleaning using a combined method of microbial fuel cell and electrokinetic. This study is conducted to remediate chromium-contaminated soil using an integrated Microbial–Soil–Remediation Coupled system (MSRC), as a novel hybrid approach. Also, in the present study, the performance of the combined MSRC process using three different electrodes, plate graphite, scratched plate graphite, and a combination of plate graphite and activated carbon granules as electrodes, was compared. The results of this study can be presented as a new technique for in-situ remediation of soil contaminated with heavy metals in coastal areas.

2. Materials and Methods

2.1. MSRC reactor structure

Figure 1 illustrates the schematic of the MSRC reactor, which is constructed from Plexiglas and consists of three equal-sized cubic chambers. The first chamber, known as the anode chamber, contains the wastewater; the middle chamber holds the contaminated soil sample; and the third chamber, referred to as the cathode chamber, is filled with distilled water and a phosphate buffer solution. Two ion exchange membranes (Anion exchange membrane and a cation exchange membrane) are used to separate the middle chamber from the other two chambers. In this study, two MSRC reactors with numbers 1 and 2 were used. A smooth graphite electrode was used in the anode chamber of reactor number 1. The dimensions of the anode and cathode electrodes were selected to be 3 × 3 × 0.5 cubic centimeters. The useful dimensions of the side and middle chambers were 3 × 5 × 5 cm3 and 4 × 5 × 5 cm3, respectively, in length, width, and height. A piece of titanium wire was used to connect each of the electrodes and place them in the circuit.

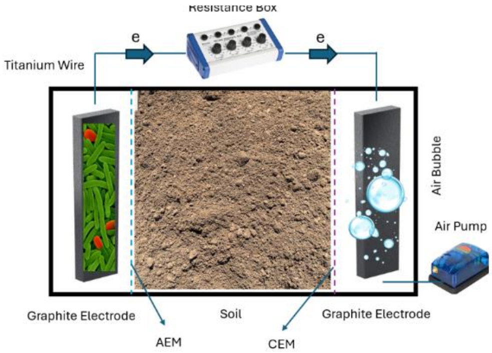

Figure 1. Schematic of MSRC in this research.

In order to start the MSRC reactor, the anode chamber was fed with a volumetric mixture of 50% anaerobic sludge obtained from the Borujerd city wastewater treatment plant as the inoculum and 50% synthetic wastewater. The specifications of the synthetic wastewater used in the system are shown in Table 1.

2.2. Soil Sampling and Characterization

The soil used in the study was obtained from the Silakhor Plain agricultural region in Lorestan Province. This plain, with an area of approximately 377 square kilometers, is located between longitude 48°28″ to 49°30″ east and latitude 33°15″ to 34°10″ north, southeast of Borujerd. After visual inspection and field observations,10 topsoil samples were collected from all parts of the plain from a depth of 0-10 cm. The location and number of sampling points in the study area are shown in Figure 2.

Then, after drying in the open air, the soil samples were passed through a No.200 sieve and then subjected to physical and chemical tests. The chemical characteristics of the studied soil are presented in Table 2. An amount of 265 g of oven-dried soil was introduced into the reactor's middle chamber and compacted layer by layer in three stages to ensure uniform density. After the reactor parts were fully assembled, deionized water was introduced into the reactor through a hole in the top of the middle chamber. This operation was repeated once a week during the test period to keep the chromium-contaminated soil moist. The reactor was operated under discontinuous feeding conditions throughout the entire commissioning period. The anolyte was drained from its chamber every 4 days to maintain the appropriate level of nutrients. This process was also performed to replace the catholyte. After achieving stable electrical potential changes, the MSRC reactor operated for two more cycles. All tests were performed at room temperature. It should be noted that the average time for cleaning the chromium-contaminated soil was 78 days for each of the reactors.

3. Analysis and Calculation Methods

The pH of the solutions was measured and recorded daily using a pH meter (HI6221, HANNA, USA). The chromium concentration in the soil was measured using an inductively coupled plasma emission spectrometer (ICP-OES, Optima 5300DV, Perkin Elmer, US). The chromium reading and measurement method was performed according to standard methods [41]. The power density of the MSRC reactor presented in this study is obtained using Equation (1).

$$P = V \times I/\nu \tag{1}$$

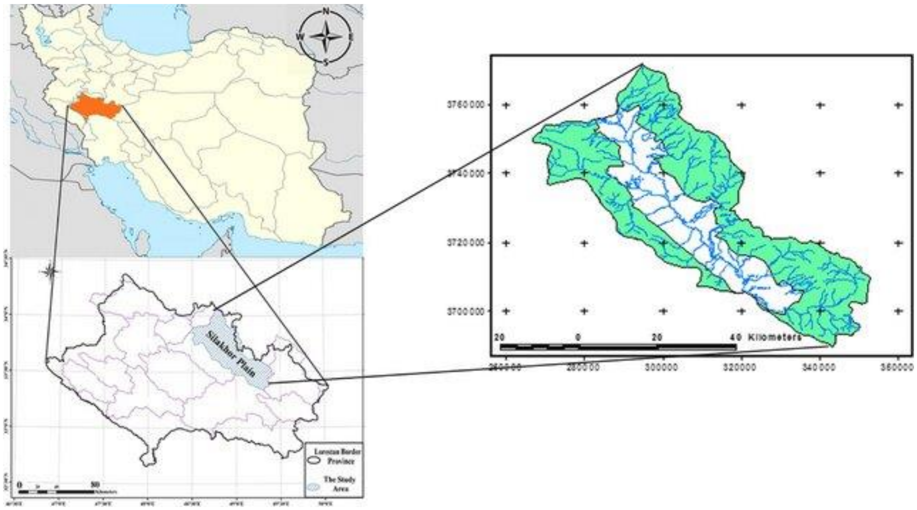

Figure 2. Silakhor plain (location and borders).

Table 1. Specifications of sewage [28].

Chemical	Amount (grams) per liter of distilled water
CH_3COONa	1.6
K_2HPO_4	1.07
KH_2PO_4	0.53
NH_4Cl	0.15
NaCl	0.5
$MgSO_4$	0.015
$CaCl_2$	0.02
Yeast extract	0.1

Table 2. Chemical characteristics of the studied samples.

	pH	CEC	EC
Unit	-	$cmol_c$/ kg	dS/m
Amount	7.6 ± 0.28	18.28 ± 4.3	0.16 ± 0.04

The resulting electric current is calculated according to Ohm's law from Equation (2):

$$I = V/R \qquad (2)$$

In these relations, V is the potential difference recorded by the data logger in volts, R is the external resistance in ohms, and ν is the useful volume of the anode chamber in liters. The polarization curve was obtained based on the studies of Watson et al. to determine and calculate the internal resistance and also the maximum power density [42]. The reactor voltage was recorded every 20 minutes using a comprehensive information system with the model (AEP-DG40-WiFi, Universal, Iran). The Coulombic efficiency (CE) was calculated in Equation (3).

$$CE = \left(\sum_{i=1}^{n} I_i t_i \Big/ FbVC_R\right) \times M \times 100\% \qquad (3)$$

In the equations presented, F is the Faraday constant, b is the number of moles of electrons produced per mole of substrate consumed (24 moles of electrons are produced per mole of glucose oxidized under anaerobic conditions), S is the concentration of the substrate used, and M is the molecular mass of the substrate consumed. COD was measured in triplicate and daily (every 24 hours) according to methods extracted from the standard method and using a spectrophotometer model (Hach DR 6000, USA) at a wavelength of 600 nm [43]. At the end of the experiments, the different parts of the reactor were separated from each other, and the soil sample, electrodes, and activated carbon granules (if present in the experiment) were stored in appropriate conditions for other examinations and measurements. The modified soil was stored in a refrigerator at 4 ℃.

4. Results and Discussion

4.1. Power and Current Generation

In the present study, a mixture of synthetic wastewater and aerobic-anaerobic sludge from a municipal treatment plant was used as feed (substrate) in the reactors. The highest OCV values were obtained in MSRC-1 and MSRC-2 reactors, with values of 641 ± 12 mV and 406 ± 34 mV, respectively, as shown in Figure 3. When the open circuit voltage showed a stable performance, the operation of two more cycles was continued. Then, the polarization and power density curves were measured and plotted at 20-minute intervals using external resistors of 50000 to 10 Ohms (resistor box) that were closed on the circuit [42]. From these curves and the polarization gradient, the internal resistance of the reactor is obtained [44]. According to the information obtained from the polarization curve of the MSRC-1 reactor, the highest power density obtained was 4.21 W/m^3, which showed a better performance compared to the MSRC-2 reactor, with the highest power density of 1.23 W/m^3 (Figure 4). The comparison between the highest power production of the two reactors shows that the type of anode electrodes can be effective in improving the performance of the reactors. The results clearly indicate that reactor number 2, due to the presence of activated carbon granules and having a large surface area for the establishment of microorganisms and creating a suitable shelter for them, has caused the formation of a suitable biofilm on it [45]. Biological systems of adhesive growth with activated carbon granules are known as the biological activated carbon process [46]. In fact, the combination of surface adsorption and biological removal of organic matter led to the production of higher power in reactor number (2). Figures 3 and 4 show the power density and polarization curves.

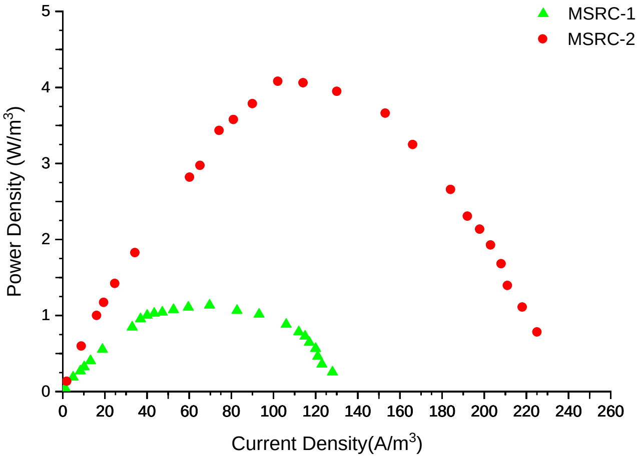

Figure 3. Power density curve of reactors.

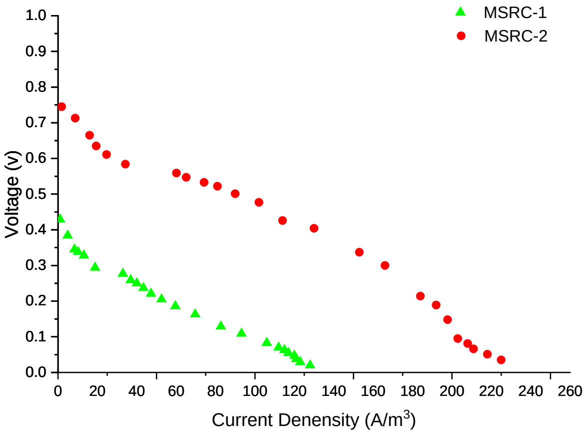

Figure 4. Polarization curve of reactors.

4.2. *pH changes in soil samples*

After transporting the soil sample to the laboratory and preparing it, the initial pH of the soil was measured, which was 6.8 ± 0.2. After the test period in two reactors, the pH changes in the chromium-contaminated soil were measured from the anode area to the cathode area. According to the results obtained, during the cleaning operation of the chromium-contaminated soil, the pH in the anode area was acidic and reached 6.11 ± 0.04. This is while the cathode area was basic and had a pH of 8.6 ± 0.2. According to the results of other researchers, acid is produced in the anode area and in the cathode area, and it is regenerated. Therefore, the pH at the anode decreases and increases at the cathode [47]. In general, the electrolysis process leads to alkalization and acidification of the soil, respectively, near the cathode and anode, and the production of H+ at the anode and OH- at the cathode [21]. H+ ions are transported by electroosmosis and electromigration processes, while OH- moves in the opposite direction and towards the anode. It should be noted that the mobility of H+ ions is 1.76 times higher than that of OH- ions. The migration of H+ and OH- from the anode and cathode along the contaminated soil sample as soon as an electric current is applied leads to dynamic changes in the pH of the soil [47]. In general, pH is one of the important and influential parameters in the mobility of heavy metals [48]. During the process of H+ movement in the sample, heavy metal desorption occurs, although with the presence of OH- and the formation of metal hydroxides such as Cr(OH)3 and their deposition in the sample, their mobility decreases [20,49]. During the implementation of the electrokinetic process for heavy metal removal, it is vital and essential to maintain an acidic pH throughout the soil sample and prevent the formation of metal hydroxides and the trapping of metals in the pores of the contaminated soil. Oxygen in the cathode chamber is reduced and produces water by consuming protons [50]. As a result of the consumption of protons in the oxygen reduction process, the proton transfer rate along the sample decreases and may cause the precipitation and trapping of chromium in the sample [10]. The results of this study also show that in both reactors, MSRC-1 and MSRC-2, the pH at the anode decreased and increased at the cathode. The increase in pH in both reactors is shown in Figure 5. The largest pH changes were observed in the MSRC-2 reactor. In fact, the pH changes in reactor number 2 are influenced by the electrochemical operations inside the cell and anaerobic respiration of microorganisms, the formation of more biofilm attached to the activated carbon granules and the electrode, and also the increase in the electron exchange rate between the GAC and the electrode [36,51]. In the case of reactor number 1, due to the smoothness of the graphite surface and less biofilm formation on it, resulting in reduced anaerobic respiration of bacteria, the rate of electron exchange was lower and therefore the pH changes were lower than in the previous two reactors.

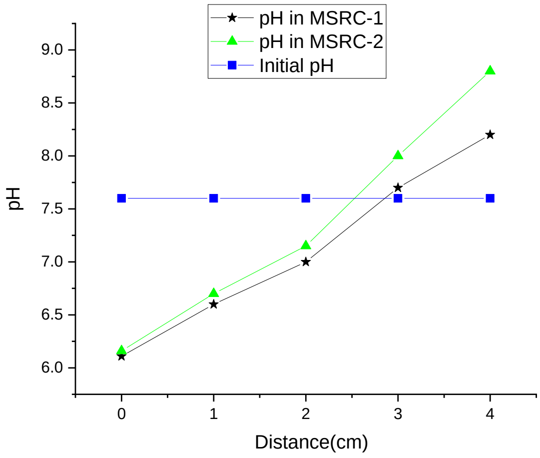

Figure 5. Relationship between pH and distance from the anode.

4.3. Chromium migration

The results of this study confirm the electrokinetic migration of chromium within the soil matrix under the electric field generated by the microbial fuel cell. As shown in Figure 6, the distribution of chromium across different sections of the soil sample after remediation by the MSRC system is compared to the initial concentration of 320 µg/g. Soil sampling was performed at 1 cm intervals along the length of the reactor. The observed trend clearly shows that chromium ions migrated from the anode region toward the cathode region, with some ultimately entering the catholyte solution. This is evident from the gradual increase in chromium concentration from the anode to the cathode side of the soil sample [10].

In this study, the soil concentration in the cathode area decreased to 132.45 µg/g and 99.1 µg/g in reactors 1 and 2, respectively. Also, the chromium concentration in the cathode region of both reactors reached 87.55 µg/g and 68 µg/g, respectively, and indicates its migration from the soil sample. Considering the presence of chromium in the catholyte (cathode chamber), it can be concluded that chromium migration from the soil sample has taken place.

4.4. COD Removal and Coulombic Efficiency in MSRC Reactor

The results of COD removal in the reactors of the present study are presented in Figure 7. Each MSRC reactor was initially operated in an open circuit mode and discontinuously, and after obtaining three suitable cycles in a closed-circuit mode. The highest COD drop in the 4-day operating time intervals was from 344.32 ± 22.35 to 763.81 ± 30.14 mg/L, respectively. The organic matter in the anolyte chamber of the reactors is consumed by the microorganisms present in it. The microorganisms form biofilms on the electrode as well as the activated carbon granules and cause the transfer of the released electrons to the electrode through the existing circuit. Activated carbon granules are mainly used for the removal of natural organic matter (NOM) and synthetic organic compounds (SOCs) [46]. A comparison of COD removal efficiencies in the two reactors shows that the low COD removal rate in the initial days of operation could be due to the growth of methanogenic bacteria that occurs in the presence of high substrate concentrations. However, the COD removal rate gradually increases as a result of the adaptation of bacteria in the system [39]. In this study, the comparison of COD removal rates clearly shows the relationship between the microbial population in the anode compartment and the COD removal rate. In fact, the increase in the anaerobic microbial population causes more electron release and consequently increases the cell voltage. The highest COD removal efficiencies were obtained in reactors 1 and 2 at 4-day HRTs of 62.28% and 83.45%, respectively. The results show that in reactor number 2, due to the presence of activated carbon granules and the provision of a more suitable substrate for creating a biofilm on it and creating ideal conditions for reducing the substrate, the COD removal efficiency was higher than in reactor 1. Also, the Coulombic efficiency obtained from the reactors is evidence of the number of electrons obtained from the substrate to produce an electric current.

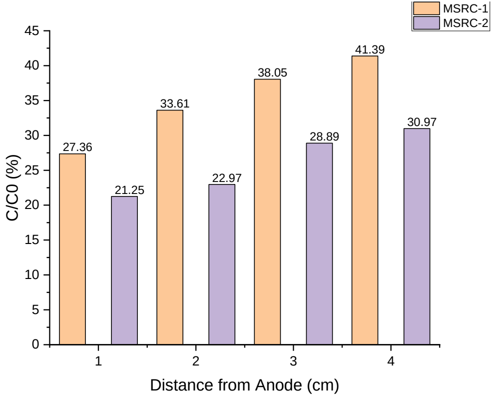

Figure 6. Chromium concentration ratio (C/C0) along the contaminated soil sample.

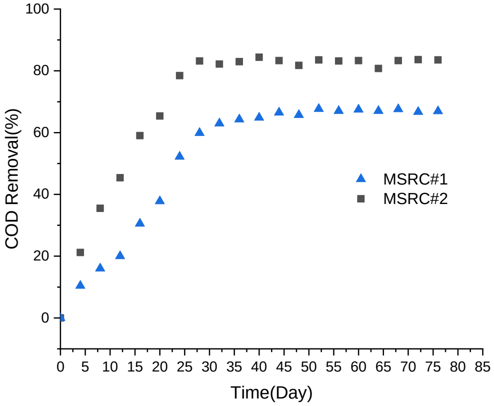

Figure 7. COD removal efficiency over time in reactors 1 and 2.

5. Conclusion

In the present study, the removal of the heavy metal chromium from contaminated soil and the impact of varying anode electrode operating conditions on the efficiency of the MSRC system were investigated. The results from reactors No. 1 and 2 indicated that pH levels decreased near the anode and increased near the cathode in both systems. The highest pH changes were observed in the MSRC-2 reactor, which was a result of electrochemical operations inside the cell and the formation of more biofilm adhering to the activated carbon granules and the electrode, as well as an increase in the electron exchange rate between GAC and the electrode. In the case of reactor No. 1, due to the smoothness of the graphite surface and the formation of less biofilm on it, the electron exchange rate was lower, and therefore the pH changes were lower than in the previous reactor. The highest OCV values were obtained in MSRC-2 and MSRC-1 reactors, with values of 641 ± 12 mV and 406 ± 34 mV, respectively. According to the information obtained from the polarization curve of the MSRC-2 reactor, the highest power density obtained was 4.21 W/m^3, which showed better performance compared to the MSRC-1 reactor, with the highest power density of 1.23 W/m^3. The comparison between the highest power production of the reactors shows that the type of anode electrodes can be effective in better performance of the reactors. Also, the research findings showed that the highest COD removal efficiency in 4-day HRTs was 78.75% and 72.96%, respectively, which indicates a more favorable performance of reactor number 2 compared to reactor number 1. Previous studies did not report COD removal efficiency under comparable MSRC operating conditions, highlighting the novelty of the present work. The use of activated carbon granules, due to the creation of a suitable substrate for bacteria to settle and anaerobic respiration, as well as electron transfer, can be a very good option for increasing the efficiency of microbial fuel cell systems. The results also showed that the use of a combined microbial fuel cell and electrokinetic process method can be considered as an environmentally friendly method due to the favorable results in cleaning chromium-contaminated soil.

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Declaration of competing interest

The author declare that she has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. The ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, have been completely observed by the author.

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