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RESEARCH NOTE

Electrokinetic and Sediment Remediation in Microbial Fuel Cell

M. Razavi[a], D. Yousefi Kebria*[a], A. Ebrahimi[b]

[a] *Environmental Engineering Department, Faculty of Civil Engineering, Babol Noshirvani University of Technology, Babol, Iran*
[b] *Department of Civil-Environmental Engineering, Mazandaran Institute of Technology, Babol, Iran*

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A B S T R A C T

Recently developed man-made structures have caused environmental pollutions, and unfortunately, in spite of the deteriorating affairs and repeated warnings by scientists and experts, the degree of contamination is increasing considerably. One of the natural sources undergoing changes is the coasts. It is mainly due to human activities which have led to a change in the quality and quantity of sediments. These regions can be contaminated by a variety of hazardous pollutants such as heavy metals and hydrocarbons. In this work, a combination of electrokinetic and MFC process was used for Cr removal from contaminated sediments. According to the obtained results, a maximum power density and current of 1.06 W/m^3 and 52.05 A/m^3 were achieved during the process. Given the presence of chromium in the catholyte, it can be concluded that the chromium migration from sediment sample to the cathode chamber has been taken. In addition, the maximum Cr measured in catholyte was 0.056 mg/l. Overall, the results confirmed the high efficiency of the proposed cell for contaminant removal from sediments.

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1. INTRODUCTION

During recent years, various technologies have been introduced in order to purify and reduce the pollution of contaminated soils and sediments, which indicates the importance and necessity of the issue. Heavy metals are one of the major problems in the aquatic and terrestrial environment, which have negative impacts on the environment and the health of the living organisms. The concentration of heavy metals in soil and sediment is increasing in various ways because of using different fertilizers and pesticides, agricultural runways, and the use of untreated urban sewage and industrial wastewater for irrigation of agricultural products. Natural processes, including atmospheric subsidence and erosion of rock and soil, may also cause accumulation of contaminants in soils and sediment [1, 2]. Sediment contaminated to heavy metals can threaten the health and life of aquatic animals and, consequently, human health, and will increase the risk of a variety of diseases in the living organism. Therefore, sediments remediation plays a key role in the purification of the aquatic environment. Soil or sediment can be considered as a complex compound that often stores and maintains metals [2]. The term "sedimentation" according to the definition of a European sediment network (SedNet) refers to a suspended solid matrix or organic and mineral sediment that is susceptible to being transported by water [3]. Electrochemical process is an efficient method in order to clean soil and sediment in which, an electric field is produced by an external source in a soil or sediment between two electrodes. Contaminants are activated and moved toward opposite electrodes by passing this low-intensity electric current [4]. Electrokinetic remediation (EKR) is a well-known environmental method for cleaning the contaminated soils and sediments. The removal efficiency of this method is generally high and the duration of the treatment process is short [5, 6]. However, the high energy required to perform this process can be considered as one of the main drawbacks of this method [7]. In such a situation, the use of other energy production methods along with the electrochemical process is essential. The equation of mass conservation for a pore water solute species (e.g.

*Corresponding Author Email: *Dy.Kebria@nit.ac.ir* (D. Yousefi kebria)

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an ion) in an EKR system can be expressed as Equation (1):

$$\frac{\partial C_k}{\partial t} = -\vec{\nabla}.\vec{J_k} + R_k \quad (1)$$

where J_k is the flux of the k_{th} ion, and C_k and R_k are the concentration and production rate of the k_{th} ion per unit of volume, respectively. This flux includes electromigration, electroosmosis, diffusion, and advection contributions [6]. Microbial fuel cell is an efficient method which can be successfully coupled with electrokinetic process. In this process, bacteria are used as a catalyst to oxidize organic material and generate electric current. The electrons produced by the bacteria, from the substrate, are transmitted to the cathode through an external circuit and protons are transmitted internally through the membrane to the anode. Then, oxygen is used to form water or other chemical oxidants in the cathode compartment to form the reduced product [8-12]. Various studies have shown that the presence of a weak electric field can lead to the movement of nutrients and microorganisms in the environment of contaminated soils [13]. Hua et al. [14] investigated chromium removal from sediments using the electrochemical method and achieved up to 60% removal. This process was carried out by placing electrodes in the sediment and creating an electric field. Current created by a microbial fuel cell can transport and remove heavy metals, although it should be stated that the obtained voltage is relatively low compared to the voltage used by other methods such as electrokinetic, which usually have higher voltage gradients [15]. Therefore, the aim of this study is to investigate the possibility of removing heavy metals from the contaminated sediment using the combination of microbial and electrochemical fuel cell processes. In this system, instead of using direct electric current, a bacterium is used to generate flow electrons. Also, the efficiency of removing heavy metal of chromium from sediments in this system is investigated. The results of this research can be used as a new method for the purification of contaminated sediments.

2. MATERIAL AND METHOD

2. 1. Cell Structure In the present study, a rectangular cubic reactor was used for chromium removal. The fabricated reactor consisted of three Plexiglass chambers that were separated from each other by an anion and cation exchange membranes (Mega Company, Czech Republic). The anion membrane was used to separate the anode and middle chambers. The cation membrane was used to separate the cathode from the middle chamber. The central chamber was filled by contaminated contaminated sediment. The working dimensions of both anion and cation chambers were 6 × 6 × 3 cm^3. The dimension of the middle chamber was 6 × 6 × 4 cm^3. The roughened surface graphite with dimension of 4 × 4 × 1 cm^3 was used as both anode and cathode electrodes. In addition, 50% of anode volume was filled by granular activated carbon (GAC). The connection between graphite electrodes and titanium wires is shown in Figure 1.

As it is indicated in the above figure, the main branch of titanium wire was connected to smaller pieces of that to make a multi-branch wire. Titanium wire was used to connect the graphite electrodes. A multi-branch titanium wire was inserted among GACs and was also connected to the wire inserted in the graphite electrode; to make a better connection and more transfer of the electrons among activated carbon granules and wire (see Figure 1). GACs were used because of their biocompatibility and low prices [16, 17]. The electrodes were washed with 1 M HCl.

2. 2. Preparation of Sediment Sample The anolyte solution was inoculated (50% v/v) with return sludge obtained from the municipal wastewater treatment plant, located in Babol, Iran. The specifications of synthetic sewage used in the system are described in Table 1. The trace element solution used in this study was reported in our previous work [18].

The reactor was operated under the batch condition at room temperature. The sediment sample investigated in this study was obtained from the Fereydounkenar beach in the southern part of the Caspian Sea with a geographical location of 36 ° 41'N and 52 ° 32'E. It was washed, and dried in an oven at 105 °C for 24 hours, in order to remove trace metals. Middle chamber was filled by sediment in three steps to eliminate any embedded air. The sample in middle chamber was compacted at each step by using a special hammer. After assembling various sections of the cell, the deionized water was injected into the middle chamber and this process was repeated in every week during the experiment. In order to increase the level of chromium contaminant in the sample, 300 µg/g $K_2Cr_2O_7$ was added to the sediment, and the sediment sample were completely mixed before being placed in the middle chamber of the cell.

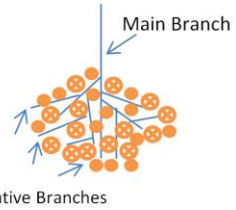

Figure 1. Schematic of connection between GAC and titanium wire

TABLE 1. Specifications of synthetic sewage [18]

Chemical	Amount in gram per a 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
Extrac yeast	0.1

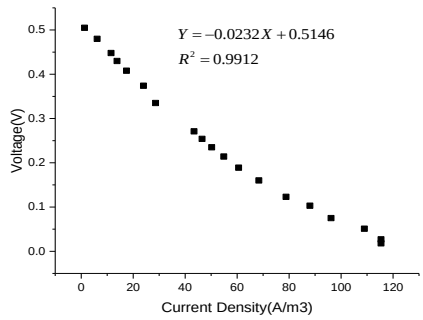

Figure 2. Polarization curve

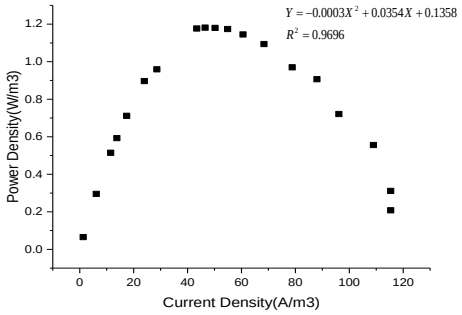

Figure 3. The power density curve

3. MEASURMENT AND CALCULATIONS

The system voltage was measured using a data logger (AEP-DG30.1, Universal, Iran) every 10 minutes. The samples were centrifuged and Chromium removal was measured according to the standard method using spectrophotometer (UNICO 2800, USA) at a wavelength of 540 nm [19]. The pH was measured using a pH meter (WA-2017SD, Lutron, Taiwan).

3. 1. Polarization and Power Density Curves The cell was operated for duration of more than two months under repeated batch cycles with a time interval of five days. During operation, electricity was continuously generated and recorded by data-logger. The maximum voltage of 505 mV was obtained when the cell was operated under open circuit condition. When a stable performance was observed in voltage generation during open circuit mode, the polarization and power density curves were plotted with changing applied external resistance from 50k to 10 Ω [10, 20]. The high voltage obtained in this study was probably attributed to the high concentration of organic matter in anode chamber.

The polarization and power density curves show the internal resistance of the cell, as well as the highest power density of reactor. The obtained results are illustrated in Figures 2 and 3. An internal resistance of 512Ω was achieved in the cell. Furthermore, the highest power density obtained in the cell was 1.2W/m^3. The obtained results are comparable with previous research [15]. The transmission of the electric current through soil sample or sediment depends heavily on the ionic conductivity of solutions and internal resistance. All experiments were measured at the environment temperature, and so there was no specific control over temperature. According to previous studies, an increase in temperature can affect the metabolism of bacteria and consequently improve the performance of the cell [21, 22]. As regards to voltage generation, it was observed that electrical energy production was high in the early stages of each cycle and it decreased significantly over the batch cycle. Besides, PH variation in the sediment led to sequestration and trapping of metallic ions near the cathode electrode, which increased the resistance of the cell and reduced the electricity production [15, 23, 24].

3. 2. pH Variation The pH of the anolyte and catholyte was measured on a daily basis and shown in Figures 4 and 5. The initial pH of sediment was equal to 7.39. The results have shown that during the purification of the sediment, the pH of the anolyte solution decreased gradually through each batch cycle, while an opposite trend was observed in catholyte compartment. Low pH in anode chamber had an inhibitory effect on exoelectrogenic bacteria activities and resulted in decreasing the electricity production during the last days of each batch cycle. Similar results were reported by other previous researchers [15]. In a microbial fuel cell, microorganisms oxidize the organic matter and produce electrons that would be transmitted by a set of respiratory enzymes in the cell and produce energy like ATP for the cell [9]. Released electrons are delivered to a final electron receiver and will be reduced in cathode chamber [25]. In addition, due to the limited proton transfer in the soil, the sediment pH has increased from

the anode compartment region to the cathode one [5]. Similar results are obtained in all soil remediation by electrokinetic method [26]. It can be concluded that pH variation is affected by two important mechanisms; electro-osmosis transfer along with electromigration. Ion transfer or electromigration is the process of ion transferring water among soil pore, under an electrical field. This process involves the migration of H^+ and OH^- to the electrodes with opposite loads [6]. During the process of sediment remediation by electrokinetic, soil pH was reduced because of the migration of proton from the anode to the cathode in the sediment environment, and the electrical conductivity of the soil was increased as a result of ionization of minerals and metals [27]. H^+ and OH^- moved by electromigration to the cathode and anode, respectively. Hydrogen ions are lighter than produced hydroxyl ions, and hence, they passed longer distance than hydroxyl ion by electro-osmosis process [28, 29]. In order to keep a charge balance in the soil, an eco-friendly method should be employed. Sedimentation and absorption of heavy metals can prevent their displacement or limit their removal from soil or sediment. The development of the acidic ions migration from anode to the cathode helps heavy metals dissolution on the soil. However, high pH conditions near let the cathode lead to settlement and sequestration of metal hydroxide [21]. Thus, controlling pH in anode and cathode compartments and preventing high changes are one of the effective factors on the electrokinetic process [21].

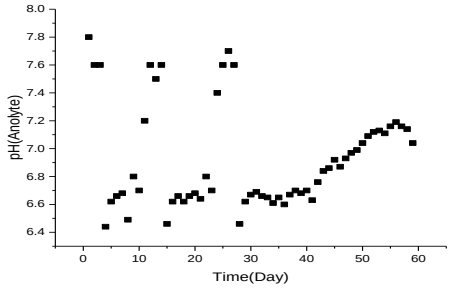

Figure 4. Measured Value of pH in Anolyte

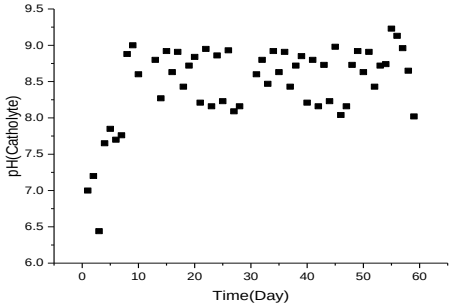

Figure 5. Measured Value of pH in Catholyte

Also, soluble forms of heavy metal cations and anions, as a result of soil pH or sediment reduction, can lead to the increase of the electrical conductivity and reduce its resistance [15]. Hence, pH control was carried out continuously in the present work and all changes were controlled. The pH range was adjusted by adding phosphate buffer during the process when it was necessary.

3. 3. Metal Migration This study investigated the purification of the sediment contaminated to heavy metal (Chromium) by using a combination of electro-kinetics and microbial fuel cells methods. The results of the study have shown that in the sediment sample, the migration of chromium was occurred under the electric field produced by microbial fuel cell. The initial concentration of chromium in the sediment sample was considered to be 300g/µg. During purification of sediment sample, the chromium concentration in catholyte was daily measured. After more than two months operations, Cr in the sediment was migrated from the anode to the cathode regions in microbial fuel cell. Given the presence of chromium in the catholyte, it can be concluded that the chromium migration from sediment sample to the cathode chamber has been taken. In addition maximum Cr measured in catholyte was 0.056 mg/l. Habibul et al. [15] investigated the cadmium and lead removal from the local vegetables garden soil with a combination of electrokinetic and MFC processes and achieved 31% and 44.1% removal after 143 days, respectively. In this study after remediation, the concentration of Cr in the anode region decreased to 114 µg/g, indicating that about 68% of Cr in the sediment would be mitigated. After more than 70 days remediation, the Cr concentration in the cathode region reached to 83.5µg/g.

4. CONCLUSION

Microbial fuel cells are another technology which can be used for contaminant removal from sediment. Microbial fuel cells are an environmentally-friendly technology that uses bacteria as a catalyst to oxidize organic materials and generate electric current. Results showed the possibility of the technique for the remediation of toxic metal contaminated sediments. The low voltage was generated from organics oxidation by microbial culture, resulting in lower metal migration rates compared with those in the conventional electrokinetic remediation technique with a high voltage gradient. Nevertheless, results indicated that the weak electrical field generated from the microbial fuel cell could power the electrokinetic remediation. According to obtained results, a maximum power density and current of 1.06W/m^3 and 52.05A/m^3 were achieved

during the process. After more than 60 days of operations, Cr in the sediment migrated from the anode to the cathode regions in microbial fuel cell. Given the presence of chromium in the Catholyte, it can be concluded that the chromium migration from sediment sample to the cathode chamber has been taken. In addition, the maximum Cr measured in catholyte was 0.056 mg/l and the Cr removal in anode region was 68% in 70 day after remediation. Overall, the results confirm the high efficiency of the proposed cell for contaminant removal from sediments.

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RESEARCH NOTE

M. Razavi[a], D. Yousefi Kebria[a], A. Ebrahimi[b]

[a] *Environmental Engineering Department, Faculty of Civil Engineering, Babol Noshirvani University of Technology, Babol, Iran*
[b] *Department of Civil-Environmental Engineering, Mazandaran Institute of Technology, Babol, Iran*

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چکیده

توسعه اخیر ساخته های دست بشر، موجبات آلودگی محیط زیست را فراهم آورده است و متاسفانه علیرغم هشدارهای کارشناسان و دانشمندان این حوزه میزان آلودگی به شکل چشمگیری افزایش یافته است. یکی از این منابع طبیعی دستخوش تغییرات سواحل است که عمدتا به دلیل فعالیتهای انسانی منجر به تغییر کیفیت و کمیت رسوبات گردیده است. این مناطق توسط انواع آلاینده های خطرناک همچون فلزات سنگین و هیدروکربنهای نفتی آلوده شود. در تحقیق حاضر از فرآیند ترکیبی الکتروکینتیک و پیل سوختی میکروبی برای حذف کروم از رسوبات استفاده شده است. با توجه به نتایج به دست آمده بیشترین چگالی توان ۱/۰۶ وات برمترمکعب و بیشترین چگالی جریان ۵۲/۰۵ آمپر بر مترمکعب در طی فرآیند حاصل گردید. با توجه به حضور کروم در محلول کاتولیت، می‌توان نتیجه گرفت که مهاجرت فلز کروم از نمونه رسوب به درون محفظه کاتد انجام شده است. علاوه بر آن حداثر میزان کروم بدست آمده در نمونه کاتولیت ۰/۰۵۶ میلی گرم بر لیتر بوده است که حاکی از کارایی بالای سلول پیشنهادی برای حذف آلودگی از رسوب می باشد.

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