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Evaluating the removal efficiency of cadmium and mercury from industrial wastewater using microbial desalination cell

Fariba Mirzaienia[a], Ahmad Jonidi Jafari[b], Mohammad Malakootian[a,c,*]

[a]*Department of Environmental Health, School of Public Health, Kerman University of Medical Sciences, Kerman, Iran, Tel. +98 343 132 5128; Fax: +98 343 132 5105; emails: m.malakootian@yahoo.com (M. Malakootian), fariba.mirzaie@gmail.com (F. Mirzaienia)*
[b]*Research Center for Environmental Health Technology & Department of Environmental Health Engineering, School of Public Health, Iran University of Medical Sciences, Tehran, Iran, email: ahmad_jonidi@yahoo.com*
[c]*Environmental Health Engineering Research Center, Kerman University of Medical Sciences, Kerman, Iran*

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ABSTRACT

Microbial desalination cell (MDC) is a process converting the energy stored in chemical bonds of organic compounds into electrical energy by exoelectrogenic bacteria. In this study, polyester fabric membranes were used as selective membranes, and microorganisms were obtained from activated sludge, so that this reactor would be more cost-effective. Synthetic samples were provided with the concentration of 25, 50, 75, and 100 mg L^{-1} of cadmium and mercury, and removal efficiency was analyzed after 60, 90, and 120 min of retention time in psychrophilic, mesophilic, and thermophilic phases and with 3–4, 4–5, and 5–6 mg L^{-1} dissolved oxygen (DO). Optimum conditions for removing cadmium and mercury were observed in 100 mg L^{-1} concentration, 4.43 mg L^{-1} DO, 26.25°C, and 120 min of retention time. Experiments were performed on the wastewater of Isfahan Steel Company, Iran, as an actual sample. The removal efficiency of cadmium and mercury was 69.57% and 67.26%, respectively. MDC is a good choice for industries because it can simultaneously treat municipal and industrial wastewater in separated chambers as an effective method with a relatively high efficiency for removing cations in industrial wastewater.

Keywords: Microbial desalination cell; Exoelectrogenic bacteria; Industrial wastewater; Isfahan Steel Company

1. Introduction

Humans are highly dependent on energy to develop science and technology [1–5]. One way to alleviate energy shortages and global warming problems is using renewable energy technologies [4,6]. Electricity generation from renewable energy sources is highly desirable as it has no carbon dioxide emissions. Application of the microbial fuel cell (MFC) technology is popular because microorganisms convert the energy stored in chemical bonds of organic compounds into electrical energy via catalytic reactions [7,8].

Microbial desalination cell (MDC) is modified from MFC reactors by inserting a chamber between the anode and cathode chambers and a pair of ion exchange (IX) membranes. IX membranes remove ions from water with low energy consumption, relatively [9–12], such that the first MDC made for desalination had three chambers of anode, cathode, and middle chambers between anode and cathode [10].

In the anode chamber, exoelectrogenic bacteria oxidize organic compounds. During the oxidation process, electrons are removed from the substrate and transmitted to the terminal electron acceptor (TEA) (i.e., cathode) through the electron transport chain. In the cathode chamber, oxygen is as TEA. Reaction of electrodes creates an electrical potential gradient [12,13] causing the desalination of aqueous solutions

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* Corresponding author.
Department of Environmental Health, Environmental Health Engineering Research Center, Kerman University of Medical Sciences, Kerman, Iran.

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with the transfer of ions from the middle chamber to anode and cathode chambers through IX membranes [10,12].

The concept of MDC was first introduced by Cao et al. [10] on a small scale (volume of salt water: 3 mL), and later Jacobson et al. [11] created an MDC on a large scale (volume of salt water: about 1 L). After that, Zhang and He [14] increased the scale of MDC volume to about 105 L. MDC has a high potential in desalination systems and can be used as a pretreatment for the reverse osmosis process, significantly decreasing the desalination energy cost and ion sedimentation on the membranes [11,15].

Cadmium (Cd) is used in electroplating, batteries, paint pigments, and alloys with various other metals, but it is not essential for plants and animals. The metal is extremely toxic and accumulates in the kidneys and liver, with prolonged intake at low levels sometimes leading to dysfunction of the kidneys. Mercury (Hg) is used in amalgams, mirror coatings, vapor lamps, paints, measuring devices, pharmaceuticals, pesticides, and fungicides. It is often used in paper mills as a mold retardant for paper. Inorganic mercury, dimethylmercury, is very toxic and can concentrate in the aquatic food chain [16].

Heavy metals should be removed from water, wastewater, and the environment to protect the health of individuals. Some studies have measured the amount of heavy metals such as arsenic, copper, lead, Cd, and iron concentration in the drinking water resources of Central and Southern Bardsir Plain, Iran, in 2014 [17].

The MDC was used in various studies for removing heavy metals, including nickel, lead, copper, and zinc from industrial wastewater. Moreover, the efficiency of MDC in the removal of arsenic from aqueous solutions has been examined [18–20]. An et al. [21] in China also reduced hexavalent chromium to 75.1% with MDC.

Different methods have been employed for removing heavy metals from paint industry wastewater using lightweight expanded clay aggregate (LECA) as an available adsorbent [22], In addition, the efficiency of perlite as a low-cost adsorbent applied for the removal of Pb and Cd from paint industry effluent [23], the effectiveness of wood ash in the removal of Cd from paint industry effluent [24], and the efficiency of lead biosorption removal from industrial wastewater by micro-alga *Spirulina platensis* [25] have also been investigated.

The removal efficiency of Cd from actual samples was mostly examined by the MDC method compared with other methods, which was 53% in the study of the nanofiltration process efficiency in Pb, Cd, Cr^{6+}, and Cu ion removal from sulfate-containing water and 28% in comparison with the efficiency of nano-zerovalent iron particles and manganese compounds in Cd ion removal from aqueous environments, studied by Malakootian and Khazaei [26], and Malakootian et al. [27].

In the present study, the removal of Cd and Hg was investigated by the MDC method. The novelty of this study lies in the use of membranes made by polyester fabrics instead of gel polystyrene used in most other studies. The ion exchange capacity of anion exchange membrane (AEM) and cation exchange membrane (CEM) was 1.6 and 1.9 meq g^{-1}, respectively. To swell these membranes, demineralized water was used instead of salt water (NaCl). Both polyester fabric membranes and demineralized water are cheaper than their counterpart. The applied microorganisms were obtained from activated sludge, so this reactor was more cost-effective. To the best of our knowledge, there is no study on the removal of Cd and Hg by the MDC in psychrophilic, mesophilic, and thermophilic phases and 3–4, 4–5, and 5–6 mg L^{-1} DO to understand what would happen if environmental conditions change the exact temperature and DO in the real process. Thus, we chose a range of temperature and DO values.

2. Materials and methods

This experimental study was performed in 2016 on 108 synthetic samples containing Cd and 108 synthetic samples containing Hg. The wastewater of Isfahan Steel Company was used as an actual sample containing Cd and Hg.

The reactor was made by Plexiglas with 10 mm of thickness. The dimension of the reactor was 48 cm (length), 18 cm (width), and 18 cm (height), and its volume was about 8 L. The reactor operated in a batch system with three chambers, a bioelectrochemical chamber or anode, a middle chamber, and an electrochemical chamber or cathode aerated with an aquarium pump. The inside dimension of each chamber was 14 × 14 × 14 cm^3, the inlet of each chamber was at the top, and their outlet was at the bottom.

Anode and middle chambers were separated by an AEM (Famasep FTAM-E of FuMA-Tech GmbH Company, Germany), and the middle and cathode chambers were also separated by a CEM (Fumasep FTCM-E of FuMA-Tech GmbH Company, Germany). AEM and CEM were made of polyester fabrics. The membranes were soaked in deionized water for 48 h in accordance with the company's guideline, each was placed between two perforated supporting Plexiglas plates to prevent damage due to the fluid pressure in the reactor.

There were two carbon graphite electrodes with the dimension of 4 × 1 × 14 cm^3, one of them placed in the anode chamber (called the anode electrode) and the other inserted in the cathode chamber (called the cathode electrode). The electrodes were linked to each other by copper wire and a digital ohmmeter. The electrodes were soaked in deionized water for 24 h.

Return activated sludge was used as the microorganism source, and the wastewater of the aerated tank was applied as the organic material source in the anode chamber called the anolyte. Return activated sludge and wastewater were collected from Kerman Wastewater Treatment Plant, Iran. The catholyte in the cathode chamber was 0.1 M phosphate buffer, and the middle chamber contained the synthetic (first phase) or actual (second phase) sample. Synthetic samples were obtained from cadmium sulfate hydrate and mercury (II) sulfate, with the pH set at 7 using 0.1 M sulfuric acid and 0.1 M sodium hydroxide. The MDC pilot is depicted in Fig. 1.

The schematic representation of the MDC function and supporting plates is illustrated in Figs. 2a and b, respectively.

To study the removal efficiency, about 25 mL of the sample was collected from the outlet of the middle chamber in each case. Experiments were performed for each metal concentration (25, 50, 75, and 100 mg L^{-1}) after 60, 90, and 120 min of retention time and at different temperatures of psychrophilic (0°C–20°C), mesophilic (20°C–35°C), and thermophilic (40°C–55°C) phases at 3–4, 4–5, and 5–6 mg L^{-1} ranges of dissolved oxygen (DO). Analysis of samples was performed by a spectrophotometer atomic absorption device (Younglin AAS 8020 model manufactured by YL Instruments, South Korea). Experiments were performed on the wastewater of Isfahan Steel Company as an actual sample under the same condition. The quality of the actual sample was determined and is given in Table 1. Finally, the removal efficiency of each metal was calculated by Eq. (1).

$$\eta = \frac{C_0 - C_1}{C_0} \times 100 \tag{1}$$

where η is the removal efficiency percentage, and C_0 and C_1 are the primary and secondary concentrations of each metal, respectively.

Experiments were conducted in accordance with the standard methods for the examination of water and wastewater (2540 D, 5210, 5220 C, 4500 D, 4500 E, 5560 C, and 3111B) [16]. All chemicals and materials were purchased from Merck Company, Germany. SPSS 16 was used for data analysis using descriptive statistics.

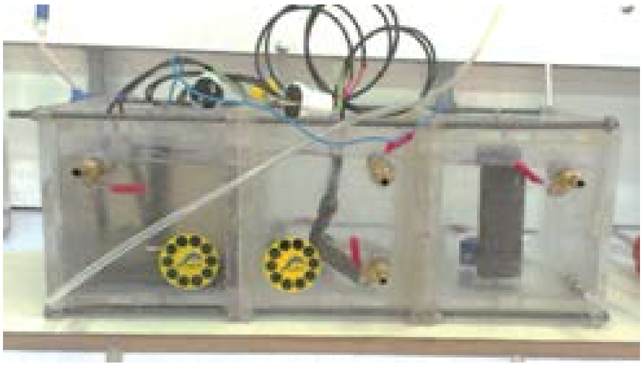

Fig. 1. MDC pilot.

3. Results and discussion

The quality of the wastewater of Isfahan Steel Company is shown in Table 1.

The results of the effect of DO on the removal efficiency of Cd and Hg in synthetic and actual samples by the MDC method with increasing retention time are presented in Fig. 3.

When the amount of DO in the cathode chamber was set to 3–4 mg L^{-1}, with increasing the contact time, the removal efficiency of Cd and Hg decreased. At this time, exoelectrogenic bacteria on the anode electrode adapted with the environment were in the Log growth phase. They were oxidizing organic matters in the substrate of the anode chamber, and the amount of produced electrons was large. However, as the TEAs in the cathode chamber were low, their transfer from the anode to the cathode electrode and the removal of the metals were not maximum. The amount of DO was increased to 4–5 mg L^{-1} and the removal efficiency of Cd and Hg in synthetic and real samples increased as well because the amount of oxygen as TEA had increased in the cathode chamber. Furthermore, upon raising the contact time to 150 min in this range of DO, removal efficiency increased in comparison with 30 min. However, when the amount of DO was raised to 5–6 mg L^{-1}, the removal efficiency of Cd and Hg in both synthetic and real samples unexpectedly decreased. In this phase, the amount of nutrient due to the batch system of the MDC was reduced and the microorganisms were in declining growth and death phases while TEA was great in the cathode chamber. Therefore, the removal efficiency of Cd and Hg decreased especially with elevating the retention time.

If the system of the MDC was continuous, upon increasing the DO in the cathode chamber as well as the retention time and entrance of fresh nutrient and organic matter in the anode chamber, the removal efficiency of the cations may have elevated exponentially, while in this research it increased at first and then decreased. The reduction and elevation of DO decreased the produced electricity. The maximum removal in synthetic (62.63%) and actual (55.80%)

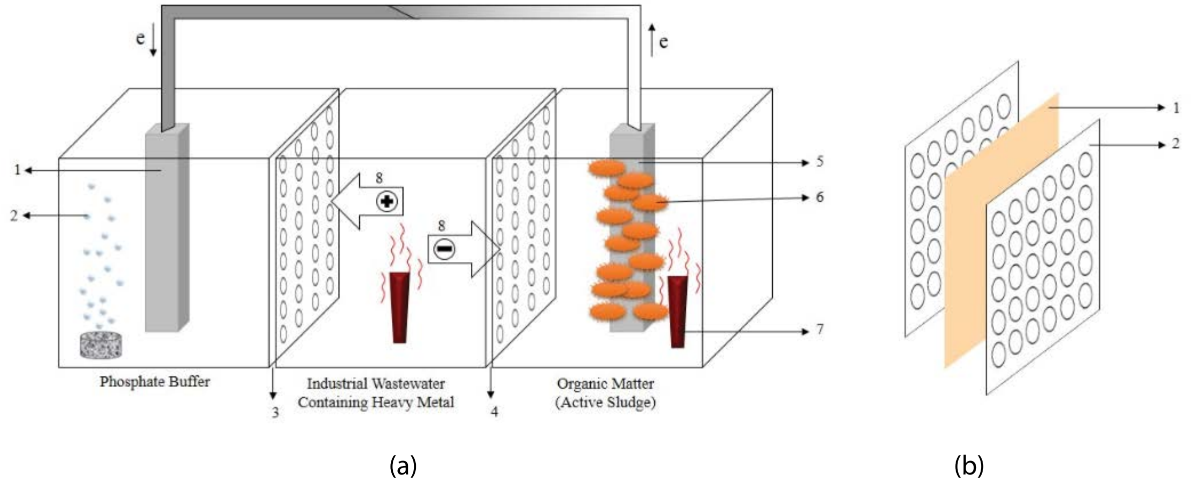

Fig. 2. (a) Schematic view of the MDC: (1) cathode electrode (in the cathode chamber), (2) oxygen bubbles, (3) CEM, (4) AEM, (5) anode electrode (in the anode chamber), (6) exoelectrogenic bacteria, (7) heater, and (8) cations and anions; and (b) anionic and cationic exchange membranes supporting plates: (1) membrane and (2) supporting plate.

Table 1
Quality of the wastewater of Isfahan Steel Company

pH	EC (ds m^{-1})	Turbidity (NTU)	TSS (mg L^{-1})	BOD (mg L^{-1})	COD (mg L^{-1})	SO_4^{2-} (mg L^{-1})	NH_4^+ (mg L^{-1})
7.4	20.7	55	62	97	150	15.68	3.7
VFC (µg L^{-1})	Cu (mg L^{-1})	Zn (mg L^{-1})	Cd (mg L^{-1})	Pb (mg L^{-1})	Ni (mg L^{-1})	Hg (mg L^{-1})	Cr (mg L^{-1})
481	0.06	0.12	0.33	0.02	0.038	0.41	0.1

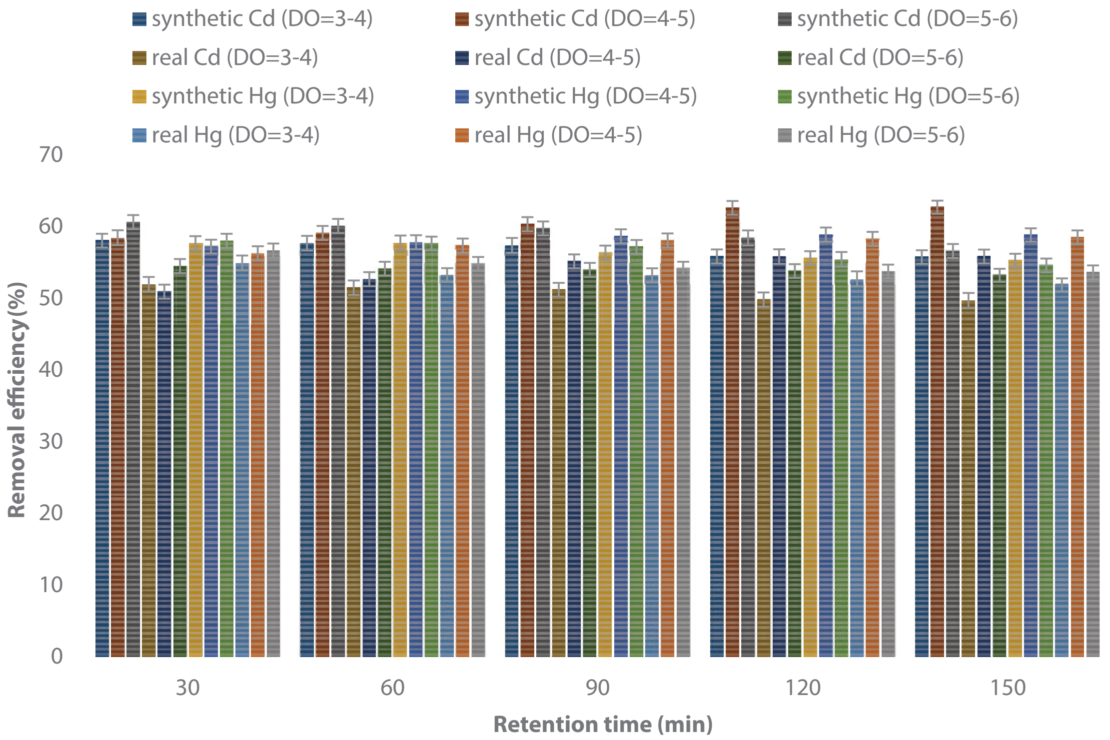

Fig. 3. Effect of DO on Cd and Hg removal efficiency by the MDC in synthetic and actual samples.

samples of Cd was observed in 4.43 and 4.38 mg L^{-1} of DO in 120 min, respectively. The maximum removal of synthetic (58.86%) and real (58.29%) Hg was achieved at 4.43 and 4.46 mg L^{-1} of DO, respectively and 120 min of contact time. Note that the maximum removal efficiency of Cd and Hg was observed in 150 min of retention time but it was not reported because increasing the removal efficiency not only was insignificant but also increased the electricity consumption for aerating. In a similar study in Iran, Malakootian et al. [28] concluded that maximum voltage is produced at the oxygen concentration of 4–5 mg L^{-1} DO. Moreover, Malakootian et al. studied the efficiency of MDC in the removal of arsenic from aqueous solutions, finding that maximum removal efficiency occurred in 120 min [20]. In addition, Clauwaert et al. [30] in Australia and Bergel et al. [29] in Italy found that decreasing DO in the cathode chamber is one of the challenges in MFC and reduced the produced current. These studies are in accordance with the present research considering the range of DO and retention time for maximum removal efficiency.

The results of the effect of temperature on the removal efficiency of Cd and Hg in both synthetic and actual samples by the MDC method with increasing retention time are given in Fig. 4.

Upon increasing the retention time in psychrophilic (0°C–20°C) and thermophilic (40°C–55°C) phases, the removal efficiency of each metal in both synthetic and real samples was decreased. Growth conditions for bacteria were unsuitable and the decomposition of organic matters present in the anode chamber with any range of DO became slow. When the condition moved from the psychrophilic toward the mesophilic (20°C–35°C) phase and with raising retention time, the removal efficiency of cations in 4–5 mg L^{-1} DO elevated. In this stage, the condition was suitable for the metabolism, growth, and proliferation of exoelectrogenic bacteria. The maximum removal of Cd in synthetic (65.02%) and actual (63.23%) samples was observed at 26.25°C and 26.22°C and the maximum removal of Hg in synthetic (63.87%) and actual (54.02%) samples occurred at 26.25°C and 26.51°C in 120 min and 4–5 mg L^{-1} DO, respectively. The maximum removal efficiency of Cd and Hg was observed at the same condition as the effect of DO on Cd and Hg removal efficiency by the MDC in synthetic and actual samples in 150 min of contact time.

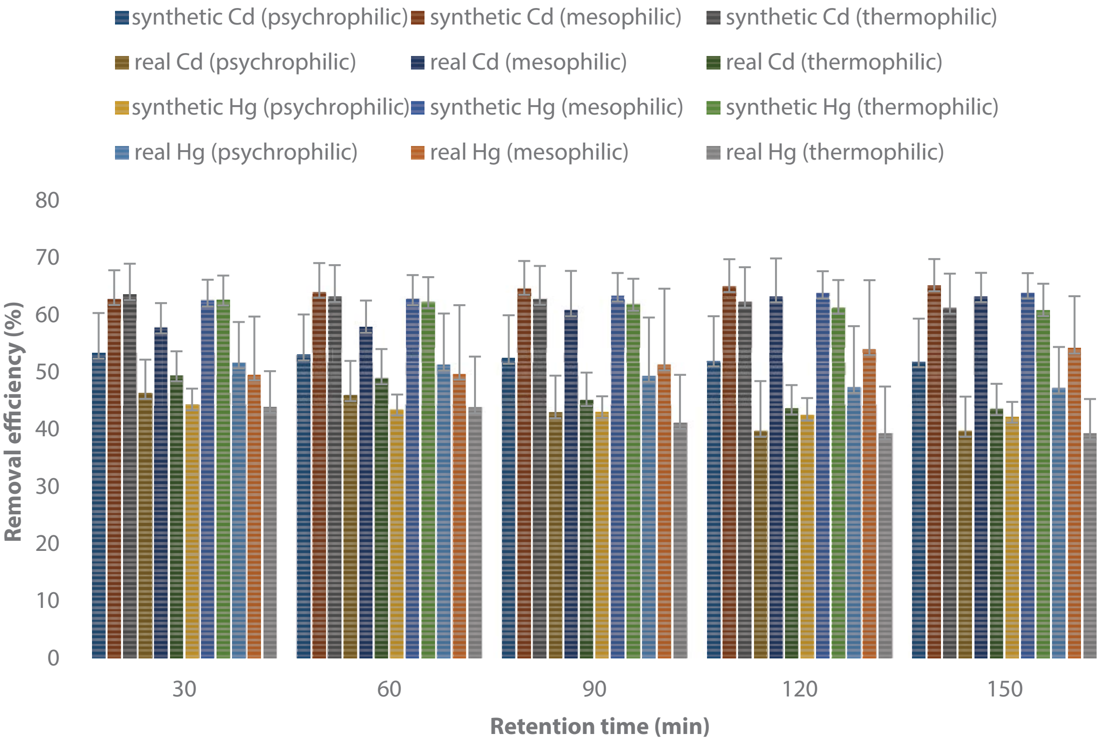

Fig. 4. Effect of retention time on Cd and Hg removal efficiency by the MDC in synthetic and actual samples.

With increasing the temperature (from mesophilic to thermophilic) and contact time, bacterial activity and growth increased to a point and quickly died [31]. It decreased the amount of electricity between two electrodes, and then the removal efficiency of metals was reduced. In a study aimed at evaluating the removal efficiency of nickel and lead from industrial wastewater in Iran using the MDC, Mirzaienia et al. [18] observed that maximum removal efficiency occurred in the mesophilic phase, 120 min, and 4–5 mg L^{-1} DO. Moreover, Brastad and He [32] and Luo et al. [15] in the United States found that, with increasing the retention time in the MDC, electricity production and removal efficiency increased, and a high desalination was observed [15,32]. Furthermore, Lio et al. [33] in the United States, Qu et al. [34] in China, and Werner et al. [35] in Saudi Arabia concluded that decreasing the temperature led to decreased electricity production, and a high voltage was produced at 25°C ± 1°C and 30°C, respectively.

The results of the effect of the initial concentration of Cd and Hg in the synthetic samples on removal efficiency by the MDC method are depicted in Fig. 5.

A direct relationship was observed between different initial concentrations and removal efficiencies of Cd and Hg. With increasing the initial concentration of each synthetic metals at optimum contact time (120 min), their removal efficiency was increased because there were more ions in

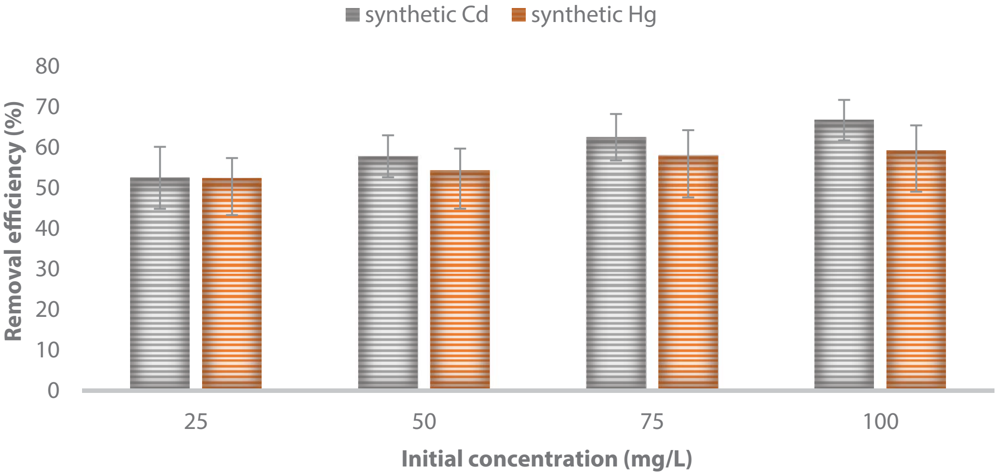

Fig. 5. Effect of initial concentration of Cd and Hg on removal efficiency by the MDC.

Table 2
Comparison of removal efficiency in different studies

Metal	Period	Group	Removal efficiency in synthetic samples (%)	Removal efficiency in real samples (%)	References
Cu	4	IB	79.70	68.37	[19]
Zn	4	IIB	79.60	70.64	[19]
Ni	4	VIIIB	74.50	68.81	[18]
Cd	5	IIB	66.79	69.57	This research
Hg	6	IIB	59.30	67.26	This research
Pb	6	IVA	75.10	70.04	[18]

the middle chamber, thus increasing electrical conductivity and moving electric current and cations and anions toward the cathode and the anode chamber, respectively. The maximum removal efficiency in the synthetic samples of Cd and Hg was 66.79% and 59.30%, respectively, at 100 mg L^{-1} concentration. The removal efficiency of real Cd (0.07 ppm) and Hg (1.12 ppm) was 69.57% and 67.26%, respectively, in optimum conditions (i.e., 4–5 mg L^{-1} DO, mesophilic phase, and 120 min of contact time). Malakootian et al. [19] and Mirzaienia et al. [18] found that maximum removal efficiency occurred at 100 mg L^{-1} concentration. In addition, Lio et al. [33] and Brastad and He [32] in the United States found that a higher number of ions led to greater electrical conductivity.

A comparison of the results of this research and similar studies is given in Table 2.

According to Table 2, with elevating the period (from period 4: Cu, Zn, Ni to period 5: Cd and then to period 6: Hg), removal efficiency was decreased. This relationship is not true in the case of Pb; with increasing the period, the removal efficiency of Pb was raised, because Hg and Pb are in the same period while Hg is a transition metal and Pb is in the main group of elements, and there are some differences between them. The mentioned relationship does not hold in the case of the removal efficiency of actual samples due to the presence of interfering compounds.

4. Conclusion

The removal efficiency of Cd and Hg from the wastewater of Isfahan Steel Company was 69.57% and 67.26%, with a relatively high efficiency. Therefore, the MDC has some advantages in comparison with conventional systems, including saving energy, available exoelectrogenic bacteria and nutrient, and simultaneous treatment of municipal and industrial wastewater. According to these items, the MDC is an effective method for the removal of heavy metals such as Cd and Hg present in industrial wastewater.

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