Removal of salt from the Caspian Sea using a single- and double-layer membrane microbial desalination cell in continuous-mode operation

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ABSTRACT

The Caspian Sea is the enormous source of saline water in northern Iran, which is a potential source for drinking water or for non-potable applications. Therefore, there is an urgent need to develop sustainable desalination methods. Microbial desalination cell (MDC) is an energy-efficient method for saline water desalination. In this study, the performance of single- and double-membrane MDCs were investigated in continuous operation mode flow. Experiments were carried out using water at different initial salt concentrations (5, 20, and 35 g/L). The single- and double-layer membrane MDCs produced a maximum electricity generation of 940 and 970 mV, respectively. However, maximum and minimum salt removing was 63.80% and 49.90% at 5 g/L of the single- and double-layer membrane, respectively. In addition, NaCl removal was increased by increasing hydrological retention time from 1 to 8 h. Moreover, the obtained result shows that recirculation of the catholyte on the anode and cathode increased the electricity generation at the end of the cycle. Totally, these results demonstrate the possibility of using this method as a safe and eco-friendly alternative for Caspian seawater desalination and power production by biodegradable organic matter and bacteria.

Keywords: Microbial desalination cell; Desalination; Bioelectricity; Bioenergy; Caspian Sea; Seawater desalination

1. Introduction

It has been estimated that nearly half of the world's population lives in countries of water scarcity. In the recent decade, freshwater is rapidly depleting due to population growth and water pollution. The vast majority of the water on earth is salty water (96.5%) [1]. It was mentioned that not only community health but also the distribution of vegetation is closely related to water conditions and quality [2–4]. Therefore, lack of fresh water resources has increased the need to desalinate seawater and brackish water, with average salt concentrations of 35 and 5 g/L, respectively [5,6]. The high-energy demands of traditional biological treatment processes such as activated sludge to decrease the salinity of these water sources have raised the need for alternative techniques [7]. Thermal desalination,

reverse osmosis, and electro dialysis (ED) are the most common desalting techniques systems that have recently been used for seawater desalination, but the energy demand of these methods is high [8]. Therefore, using alternative energy efficiency technology is desirable. Microbial fuel cell (MFC) technology is able to oxidize organic compounds of wastewater and generated electrons [9]. MFCs can be modified by adding a desalination compartment in a middle chamber that is known as a microbial desalination cell (MDC). MDC is a newly emerged technology that can use organic compounds of wastewater for producing bioenergy and desalinating saline water simultaneously [10]. MDC is also a green method technology that reduces energy consumption. The MDC process is similar to the ED process, but the current is generated within a MDC. An MDC consists of three chambers in which anode are separate with cathode by middle desalination chamber [11]. These processes are technologies in which ions are separated by migration in different directions

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charged positive and negative electrodes and create the electrical energy. In the system, anode and cathode are charged positive and negative, respectively. Ion-exchange membranes facing electrodes (anode and cathode) control ion flux [10]. Cations ions into the negative electrode and anions ions into the positive electrode are attracted from flow through membranes by two carbon electrodes. Consequently, desalination occurs based on adsorption and desorption processes, and ions are separated from the salty water [12]. It was identified that several factors such as retention time, pH of cathode and anode solution, salinity concentration, recirculation MDC, and number of cell pairs could affect the performance of the MDC system and current generation or desalination of water. Most MDCs have a single structure of ion-exchange membranes [13,14]. It is possible to use more than one membrane consisting of alternating anion-exchange membranes (AEMs) and cation-exchange membranes (CEMs) between membranes. In this type of desalination system, a single electron that is transferred at the electrodes can separate many ion pairs in comparison to a single-membrane system [15]. Much research has been carried out on desalination efficiency and current generation in the application of the different MDCs. Sevda et al. investigated the effect of organic load on salt removal in air cathode up-flow microbial desalination cell (UMDC) in batch mode and chemical oxygen demand removal (95%) was occurred with 48% salt removal [16]. Furthermore, in using of the petroleum refinery wastewater, the maximum desalination efficiency and energy production were 19.9% and 9.5 W h/m3, respectively [17]. Sevda et al. worked on the hydraulically connected osmotic MFC and 48% contained salts were removed from the seawater [18]. In addition, in the other study that have been determined that in utilization of the two chambers UMDC, 2.375 A/m² current density and 72% salt removal were obtained [19]. However, there is no report on the comparison of single- and double-layer MCD efficiency in the continuous-mode operation on salt removal and current generation. Thus, the aim of this research was to compare the efficiency of the single- and double-layer membrane MDCs in continuous-mode operation for salt removal and energy generation also, three different NaCl concentrations (5, 25, and 35 g/L), and real seawater were used in the desalination chamber. The effects of hydrological retention time (HRT) from 1 to 8 h on salt removal and power generation were examined.

2. Materials and methods

2.1. Caspian sea water analyses

Table 1 contains the results of physical-chemical parameters of water samples.

2.2. Area of study

The Caspian Sea, which is 1,200 km long, 320 km wide, and has a surface area of approximately 371,000 km², is the largest completely enclosed body of water on Earth. This landlocked mega-lake, bordered by five countries including Iran, Azerbaijan, Russia, Kazakhstan, and Turkmenistan, contains an abundance of natural resources and diverse wildlife [20]. Chemical and hydrological characteristics of the Caspian Sea are very different. Caspian Sea is affected by some factors such as temperature, wind stress, and discharge

Table 1Characteristics of the Caspian Sea water

Property	Mean ± SD
Calcium (mg/L)	$5,533 \pm 446$
Magnesium (mg/L)	442 ± 50
$SO_4 (mg/L)$	$1{,}500\pm100$
Cl (mg/L)	4,516 + 76
Sodium (mg/L)	$5,100 \pm 70$
Electrical conductivity (ms/cm)	18.36 ± 2.7
pH	8.69 ± 0.9
TDS (total dissolved solids) (mg/L)	$10,400 \pm 200$

of rivers [21]. That is reported that nearly 130 rivers enter the sea. The main sources of fresh water inputs to the Caspian Sea are the Volga (80%–85%), Ural, Emba and Terek Rivers, and Iranian rivers, which is enter to the sea is the Sepidrood River (4%–5%) [22]. The study area in the Farah Abad Region in the north of Iran (Mazandaran Province) is located at approximately N37° 15′ latitude and E50° 23′ longitude [23]. The Caspian Sea with its salinity of about 13,500–16,000 ppm could be an attractive site to test possibilities of producing potable water at a low cost with MDC.

2.3. Water sampling of Caspian Sea

Sample water was provided of the Caspian Sea from May 2017 to August 2017. The basic physicochemical properties of the waters were determined by standard techniques. The sampling was conducted at three sites. Water samples were collected in sterile glass collection vials, and it was acidified by adding 0.1 N HNO₃ to avoid further chemical reactions. The chemical and physical properties of Caspian Sea water are listed in Table 1.

2.4. MDC unit

The MDC was designed based on a cubic-shaped and consist of three polycarbonate an anode, desalination, and cathode chambers. The three compartment cells were physically separated by using an AEM and CEM, which were purchased from Mega Co. (Czech Republic; Fig. 1). The AEM and CEM membranes were soaked in 1 M NaCl for 24 h before use [24]. The cross section area of the chambers was 9 cm², and the inside volumes of an anode, middle desalination, and cathode chambers were 27, 3, and 27 cm³, respectively [25]. Two 7 cm × 7 cm diameter carbons felt without Pt. Catalyst (ELAT LT1400W, USA) were installed as the anode and cathode electrodes. Titanium wire with a diameter of 0.25 mm was assembled in each electrode chamber as current collectors. Prior to use, electrodes and wire were washed for 48 h in 1 M HCl and were rinsed with water to remove trace metals. The external resistance of each separate MDC was 2 Ω at all conditions.

2.5. Standards and operating conditions

The entire analytical grade chemicals used in this study were purchased from the Merck Company, and doubly distilled-deionized water was used in the total experiment process. Artificial wastewater consisted of sodium acetate





(b)

Fig. 1. Three-chamber MDC used for desalination tests: (a) photograph and (b) schematic.

(1.6 g/L) in a nutrient buffer solution containing (per litter in deionized water): 4.4 g KH₂PO₄, 3.4 g K₂HPO₄·3H₂O, 1.5 g NH₄Cl, 0.1 g MgCl₂·6H₂O, 0.1 g CaCl₂·2H₂O, 0.1 g KCl, and 10 ml of trace mineral metals solution was fed in the anode chamber. Anaerobic microorganisms were inoculated in the anode chamber by adding sludge that was collected local municipal wastewater treatment plant (Sari, Iran). Ferricyanide catholyte containing (per litter in deionized water) 16.5 g K₃Fe (CN)₆, 9.0 g KH, PO₄, 8.0 g K, HPO₄, 3H, O was fed on cathode chamber [26]. The middle chamber was filled at different levels of NaCl concentration (5, 20, and 35 g/L) [27]. All of the experiment was carried out by using of single- and double-exchange membrane. Before conducting desalination experiments, the anode chamber was running in MFC mode until the peak voltage was stable at about 600 mV. All experiments were conducted at a room temperature of 22°C ± 2°C. After establishment of biofilm on the anode electrodes, the MDC was evaluated under batch condition. In the batch experiments, solutions in all chambers were replaced after 2 d. On the next step of the experiments, MDC was operated in continuous-mode operation at the different HRT of 1-8 h, respectively. Solutions from individual feed reservoirs (100 ml each) were recalculated through the anode and cathode chambers by using a peristaltic pump at a rate of 5 ml/min (Cole-Parmer Peristaltic Pump; 12VDC/115VAC). Reactors were operated in open-circuit mode as controls for charge transfer in the absence of the current generation. The anolyte was replaced with a fresh feeding solution every 12 h to prevent a drop in pH and provide a sufficient supply of substrate for the bacteria.

2.6. Analytical measurements and calculations

All of the studied parameters in this study were determined following the environmental protection agency method [28]. The produced voltage was recorded by the dual display multimeter system (Victor 8145A). Salt concentrations were evaluated by conductivity measurements using a conductivity meter (SG3-ELK, Mettler Toledo). It was assumed that NaCl concentration would have a linear correlation with solution conductivity in the middle chamber [29]. Thus, salt concentration was measured by conductivity measurements of the salt water in the desalination chamber.

2.7. Statistical analysis

Experiments were conducted based on the mean ± standard deviation. SPSS software was used for statistical analysis (Version 17 for windows, SPSS Inc., Chicago, IL).

3. Results and discussion

3.1. Effect of initial NaCl concentration

Microbial desalination cells were initially operated in continuous-mode until the peak voltage was stable at about 600 mV. Desalination efficiency in the middle chamber with single- and double-layer membranes at each initial salt concentration (5, 25, and 35 g/L) in continuous condition was listed in Figs. 2 and 3. As shown in Figs. 2(a) and 3(a), in both single- and double-membrane MDCs, the maximum desalination was 49.9% and 63.8%, respectively, in the 5 g/L NaCl concentration. On the other hand, when the feed water salinity increased from 5 to 25 g/L, desalination efficiency decreased 26.4% and 25% in single- and double-layer membranes, respectively. While, voltage generation was increased from 889 to 940 mV and from 890 to 1,010 mV in single- and double-layer membranes, respectively. However, fluctuations in the current trend were due to the inactivity of some types of microbes [30]. Obtained result shows that high-concentration salt water was exhibited high voltage generation from 780 mV up to 940 mV in 5 and 35 g/L NaCl, respectively. High electrical conductivity is one of the basic requirements of an electrode material to enhance performance as it may enhance bio electrochemical kinetics of the surface reactions [31]. To explore the effect of salt solution concentration on desalination performance, in the single-membrane MDC, the desalination curve of the 5 g/L NaCl solution achieved the highest extent (49.9%), while the 35 g/L solution reached the lowest level of 36.2% (Fig. 2(a) and (c)). With the double-membrane MDC, NaCl removal efficiency in 5 g/L and 35 g/L is 63.8 and 45.8, respectively, (Figs. 3(a) and (c)). It is attributed that there is lower concentration differences across the ion-exchange membranes in the lower-concentration salt solution, so the liquid junction potential occurs lower and more elements of the electric field force could drive additional desalination activities [32]. Kim and Logan reported that high salinity removals require a large volume of electrolyte especially anolyte to enhance performance otherwise results in partial removal [33].

3.2. Effects of the number of membranes used

To explore the effects of the number of membranes used, the results suggested that the desalination performance of



Fig. 2. Desalination efficiency and energy generation of single membrane MDC in different NaCl concentrations 5 g/L (a) 25 g/L (b), and 35 g/L (c) during 8 h.



Fig. 3. Desalination efficiency and energy generation of double-membrane MDC in different NaCl concentrations 5 g/L (a) 25 g/L (b), and 35 g/L (c) during 8 h.

MDCs could also be improved by using double membranes between the anode and cathode chambers. Double membranes help to improve the charge transfer efficiency, and saline water flows through membrane, prompting more salt removal [34]. Some recent research has referred to using multiple pairs of ion-exchange membranes which have created alternating pairs to increase desalination efficiency in multistage MDCs [35,36], but the new structure MDC which is used in our research without a separated chamber between double membranes recovers more energy and is cost-effective compared with multi-stage MDCs. In addition, an increase in the number of membranes leads to the internal resistance reduction and more efficiency in the desalinating of salty water or reducing the salt content of water. It is demonstrated that intermembrane distance is a key factor in the MDC performance [37]. Ping and He reported that at a smaller intermembrane distance, desalination efficiency increase [38]. Moreover, some studies indicated that, at a minimum intermembrane distance, resistive loss in the ED stack was negligible [30,39,40]. In the MDC with continuous flow mode, substrate was uniformly distributed; hence, desalination efficiency and power production were increased by enhancing current distribution [41,42].

3.3. Effects of (HRT)

Similarly, the HRT is an important factor. The results indicate that in the same initial concentration of salt in both the single- and double-membrane MDCs, HRT has contradictory effects on the desalination efficiency, and NaCl removal was increased by increasing HRT from 1 to 8 h. Moreover, HRT has a significant effect at the higher initial salt concentration removal because more time needed for more salt concentration removal in continues mode operation, while in batch operation mode, the desalination efficiency was decreased during time. Presence of cations, such as Ca²⁺ and Mg²⁺, can

Table 2

Summary of MDC literature findings and recommendations

MDC configuration	Desalination chamber- Salt type /concentration	Key findings	Recommendations for future work	Refs
3 chambers	NaCl, 5, 20, and 35 g/L	 First literature mention of MDCs 90% salt removal even at high concentrations over 24 h period in batch mode 	 Ferricyanide catholyte not acceptable for practice – need research on air cathode Increasing number of chambers will increase efficiency Develop continuous water processing 	[25]
3 chambers	NaCl, 5 and 20 g/L	 First use of air cathode Equal volumes of anolyte and desalination chamber solutions Showed partial desalination; proposed MDC as pretreatment for RO 60% reduction of saline water conductivity 	 Utilize wastewater or other source of organic matter for substrate Power generation will be increased by using wastewater with higher conductivity Recirculation of catholyte to anode chamber to balance charge 	[45]
3 chambers with added voltage (microbial electro dialysis desalination cell, MEDC)		 Applied 0.55 V using external power supply Increased desalination capacity Reduced saline water conductivity by 68% Produced hydrogen gas 	Study variable applied voltage	[46]
Up-flow microbial desalination cell (UMDC)	NaCl, 30 g/L	 99% NaCl removal Saline water HRT = 4 d Max power density = 30.8 W/m² 81%–99% of electrons used for desalination process 	• Optimize system for desalination, WW treatment, or power production – operating conditions will change	[41]
Continuously operated, UMDC	NaCl, 30 g/L	 99% NaCl removal Saline water HRT – 4 d Current production = 62 mA Power density = 30 W/m³ 	Optimize UMDC performance	[42]

cause a decrease in electric conductivity as well as scaling of membrane surface during long-term batch operation mode [43]. It should be noted that in the different initial salt concentration on the continuous operation mode, current generation was not obviously changed because there is sufficient salinity to provide enough ions to transfer charges and current generation [44]. A long HRT was conducive to organics removal, but an overlong HRT decreased treatment capacity of wastewater and might cause in sufficient organics in anode chamber for current generation. Decrease in current production during time perhaps due to organics and salinity removal and mass transfer. Meanwhile, desalination of saline water is scientifically increased in high HRT due to the increase ohmic resistance in middle chambers [13]. In recent years, there has been considerable growth in delivery costs caused by water desalination, so various desalination processes have been developed. Single- and double-layer MDCs have some advantages and disadvantages with compared with previous studies. Table 2 is a comprehensive summary of MDC studies and the key literature findings from recent years.

3.4. Effects of circulation

An experiment was carried out in different circuit time to explore the effect of the recirculation on the produced voltage and desalination. The obtained result shows that the number and time of the circuit are effective factors on the MDC efficiency. The results suggested that produced voltage was increased gradually during the first time of circuit and then it started to decrease. The decrease in the produced voltage can be explained by substrate depletion [47]. Moreover, the effects of pH imbalances inside the cell, blockage membranes, and oxidation organic material and releases protons on cathode chamber should not be ignored [25,33,48,49]. In addition, curve trend on Fig. 4 shows that produced generation under high salt concentrations is more than low salt concentration. It was founded that during circulation the voltage generation was decreased while when it was stopped created a sudden increase in voltage trends. However, when both the CEM and AEM solution were replaced, voltage generation started to increase. It was found that major source of energy source for bacteria is organic matter on the anode chamber which bacteria have used of this source during biodegradation process, so led to a fairly high bioelectricity generation [50].



Fig. 4. Voltage variations from different circulation of anode and cathode catholyte and different initial salt concentrations in the double-layer membrane.

4. Conclusion

It is undeniable that by the innovation of MDC technology can be improved desalination efficiency and power generation. Meanwhile, the configuration of MDCs and operation mode are important factors. Obtained results show that when MDCs were operated with a double-layer membrane under continuous flow mode, desalination, and power generation efficiency were higher than compared with a single-layer membrane. However, the effect of initial salt concentration recirculation cathode/anode solution and HRT is undeniable. In addition, new structure MDC that was used in our research without a separated chamber between double-layer membranes recovers more energy and it is costeffective compared with multi-stage MDCs, which have been used in the previous studies. Authors firmly suggest that further research be carried out based on batch flow mode. Moreover, it seems that observation oxidation and reduction process in the MDC will be interesting.

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