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Effects of inter-membrane distance and hydraulic retention time on the desalination performance of microbial desalination cells

Qingyun Ping, Zhen He*

Department of Civil Engineering and Mechanics, University of Wisconsin-Milwaukee, Milwaukee, WI 53211, USA
Tel. +414 229-5846; Fax: +414 229-6958; email: zhenhe@uwm.edu

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ABSTRACT

Microbial desalination cell (MDC) is a promising technology for simultaneous water desalination and wastewater treatment. To further understand the factors that affect MDC performance, we investigated the complementary roles of inter-membrane distance and hydraulic retention time (HRT) in desalination by a bench-scale MDC. When the inter-membrane distance was changed from 2.5 to 0.3 cm while maintaining the same influent flow rate, the HRT of the salt solution decreased; the desalination efficiency reached a maximum at 0.5 cm distance with 10 g/L salt concentration or at 2.5 cm distance with 30 g/L. The rate of salt removal was clearly improved at a shorter inter-membrane distance. The MDC with an inter-membrane distance of 0.3 cm achieved a specific desalination rate twelve or seven times higher than that with 2.5 cm at an initial salt concentration of 10 or 30 g/L. At the same inter-membrane distance of 1.0 cm, a greater HRT led to better desalination efficiency. While at the same HRT of 6 h, the smaller inter-membrane distances resulted in higher desalination efficiency. In addition to electric current, water osmosis was found to be a major contributor to conductivity reduction. The future design and operation of MDCs should consider the trade-off between inter-membrane distance and HRTs.

Keywords: Microbial desalination cell; Wastewater treatment; Bioelectrochemical; Membrane; Energy

1. Introduction

Microbial desalination cell (MDC) is a novel concept of desalination technology that takes advantage of electrochemically active microorganisms to oxidize organic compounds and accomplishes desalination without a significant requirement of external energy input [1]. A typical MDC consists of three chambers, an anode, middle (salt), and a cathode, separated by an anion exchange membrane (AEM) and a cation exchange membrane (CEM), respectively (Fig. 1). Carbon-based materials are often used as electrodes in the anode and the cathode. Bacteria inhabiting the anode electrode decompose organic matters while releasing electrons and protons. Terminal electron acceptors (e.g. ferricyanide or oxygen) in the cathode are reduced by accepting the electrons through an external circuit. To achieve a charge balance in both the anode and the cathode chambers, cations, like sodium ions, in the middle chamber migrate into the cathode via CEM, and anions, such as chloride ions,

*Corresponding author.

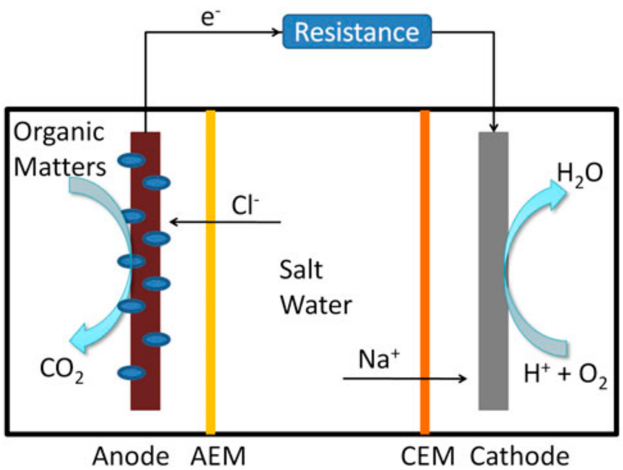

Fig. 1. The schematic of a MDC. AEM: anion exchange membrane; CEM: cation exchange membrane.

move into the anode chamber through AEM. As a result, the salinity in the middle chamber is greatly reduced. MDCs can simultaneously remove organics in wastewater, conduct desalination, and produce electricity [2].

Because of the environmental friendly features, MDC technology has drawn increasing attention from the scientific community and has been advanced through laboratory research. Researchers have replaced ferricyanide catholyte with oxygen that is commonly used as a terminal electron acceptor [3]. Continuously operated MDCs were developed in upflow configuration [4], and the size of the upflow MDC was scaled up to liters [5]. To improve the charge transfer efficiency, stack MDCs were built by using multiple membrane pairs between the electrodes [6,7]. The use of a buffer solution has been an issue in operating bioelectrochemical systems because of the high cost and the potential environmental problems; by recirculating the electrolyte between the anode and the cathode, an MDC was operated buffer-free for a short period of time [8,9]. The high cost of cathode catalysts based on noble metal hinders the future application of MDC technology; to eliminate metal catalysts, an aerobic biocathode developed by using microorganisms was found to be effective in an MDC [10]. When an external voltage was applied, hydrogen gas was produced in the cathode of MDCs [11,12]. In addition to conventional ion-exchange membranes, a bipolar membrane was also employed in MDCs for acid and alkali production [13]. Replacing the AEM with a forward osmosis membrane resulted in an accelerated desalination because of dilution due to water extraction from wastewater [14]. Recent studies applied an ion-exchange resin in the middle chamber of MDCs for treatment of a low-salinity solution [15].

MDCs are operated on a principle similar to electrodialysis (ED) [16,17], except that ED relies on an externally applied voltage while MDCs use the voltage produced internally. Both processes also have a similar reactor configuration. However, the membrane pairs in an ED usually have an inter-membrane distance of less than 1 mm to minimize energy loss [18], while most MDCs reported in the literature have an inter-membrane distance of >5 mm. The smallest inter-membrane distance in an MDC was 1.3 mm, achieved in a stacked configuration [6]. Although a smaller inter-membrane distance is expected to result in a lower internal resistance, it can increase the fabrication complexity and fouling problems in MDCs that contain much fewer membrane pairs than an ED. Larger desalination chambers often create greater ohmic resistance; however, larger chambers can also increase the hydraulic retention time (HRT) of the salt solution, thereby increasing the desalination time that will result in more salt removal. Small inter-membrane distances and long HRTs cannot be achieved simultaneously at a fixed water production rate. Therefore, there could be trade-offs between inter-membrane distances, HRT, and desalination efficiency, which has not been well addressed in the previous studies.

In this study, we have investigated the relationship between the inter-membrane distance and HRT in a bench-scale MDC with different initial salinities or inter-membrane distances. At the same influent flow rate, six different inter-membrane distances ranging from 0.3 to 2.5 cm were tested, resulting in different HRTs but the same water production rate (mL/min). We also analyzed the contributions to conductivity reduction by electric current and water osmosis (water flux into the middle chamber). We studied the effect of different HRTs at the same inter-membrane distances and the effects of different inter-membrane distances at the same HRTs.

2. Materials and methods

2.1. MDC setup

The MDC was a plate-shaped reactor, consisting of three chambers, the anode, the middle (salt), and the cathode (Fig. 1). An AEM (AMI-7001, Membrane International, Inc., Glen Rock, NJ, USA) was used to separate the anode and the middle chambers, while a CEM (CMI-7000, Membrane International, Inc.) was installed between the middle and the cathode chambers, resulting in a liquid volume of 24 mL in the anode or the cathode chamber. The anode and cathode electrodes were made by wrapping carbon cloth (3.0×7.5 cm, Zoltek Companies, Inc., St. Louis, MO, USA) around stainless mesh (3.0×2.5 cm) that was connected to an external circuit via a titanium wire. The cathode electrode contained a catalyst that was prepared by applying a mixture of Pt/C powder with Nafion solution to the surface of the carbon cloth with a final Pt loading rate of 0.3 mg Pt/cm^2. Several layers of rubber gaskets between the AEM and the CEM created the middle chamber and were also used to adjust the inter-membrane distance.

2.2. Operating conditions

The MDC was operated at a room temperature of ~22°C. The anode was inoculated with a mixture of aerobic and anaerobic sludge from local wastewater treatment plants (Jones Island and South Shore Water Reclamation Facilities, Milwaukee, WI, USA). The anode feeding solution (anolyte) was a synthetic wastewater containing acetate as an electron donor (per liter of tap water): sodium acetate, 5 g; NH_4Cl, 0.15 g; NaCl, 0.5 g; $MgSO_4$, 0.015 g; $CaCl_2$, 0.02 g; KH_2PO_4, 0.53 g; K_2HPO_4, 1.07 g; and trace element, 1 mL [18]. The acetate was overly supplied to ensure a sufficient electron supply to drive the desalination. The catholyte was 1 mM phosphate buffer solution (Na_2HPO_4, 91.6 mg/L and $NaH_2PO_4 \cdot H_2O$, 49.0 mg/L). The anode and the cathode chambers were linked to a 500 mL reservoir, respectively, which provided the anolyte or the catholyte that was recirculated between the anode or the cathode chamber and the reservoir at a rate of 24 mL/min. The use of large-sized reservoirs was to ensure a sufficient supply of anolyte and catholyte so that the anode and cathode reactions would not be limiting factors to desalination. The catholyte reservoir was continuously aerated with the air to provide adequate dissolved oxygen. The middle chamber was continuously supplied with NaCl solution from a 500 mL reservoir at different flow rates, controlled by a peristaltic pump. The anode and cathode electrodes were connected through an external circuit over an external resistor of 10 Ω. All tests were carried out in the same MDC by modifying the inter-membrane distance between the AEM and the CEM. The data were collected every 24 h, and that was designated as one operating cycle.

2.3. Analysis and calculation

The MDC voltage was recorded every 3 min by a digital multimeter (2,700, Keithley Instruments, Inc., Cleveland, OH, USA). The pH was measured using a Benchtop pH meter (Oakton Instruments, Vernon Hills, IL, USA). The conductivity was measured by a Benchtop conductivity meter (Mettler-Toledo, Columbus, OH, USA). The polarization curve was performed after open circuiting for 6 h by a potentiostat (Reference 600, Gamry Instruments, Warminster, PA, USA) at a scan rate of 0.2 mV/s. The maximum power density was calculated based on the anode liquid volume. The charge transfer efficiency was calculated as the theoretical amount of coulombs required to remove the NaCl divided by the coulombs harvested from the electric current, assuming that one mole of NaCl removal will require one mole of electrons. The desalination efficiency was determined as the percentage of salt solution conductivity decreased over 24 h. The specific desalination rate (SDR) was calculated as the total salt removed from the salt solution per day per liquid volume of the middle chamber (desalination chamber) [7]. The amount of water flux due to osmosis was determined by measuring the difference in the water volume between the salt water influent and its effluent over 24 h. The electric field strength was calculated by dividing the MDC voltage by the distance between the anode and the cathode electrodes.

3. Results and discussion

3.1. Different inter-membrane distances or HRTs at the same influent flow rate

At the same influent flow rate of 0.02 mL/min, six inter-membrane distances, from 2.5 to 0.3 cm, were tested, resulting in decreasing salt water HRTs from 50 to 6 h. Each distance was performed with at least three cycles. Two initial salt concentrations, 10 and 30 g NaCl/L, were examined. The desalination efficiency (conductivity reduction) exhibited a peak of ~40% at 0.5–1.0 cm when fed with 10 g/L (Fig. 2(A)), while it decreased with the decreasing inter-membrane distance with 30 g/L (Fig. 2(B)). At 10 g/L, the highest desalination efficiency of $40.7 \pm 1.0\%$ was achieved at the inter-membrane distance of 0.5 cm (HRT 10 h) and the lowest desalination efficiency of $31.9 \pm 2.1\%$ was obtained at 2.5 cm (HRT 50 h). When the initial salt concentration was increased to 30 g/L, the largest inter-membrane distance of 2.5 cm produced the highest desalination efficiency of $35.6 \pm 1.5\%$, while the smallest inter-membrane distance of 0.3 cm (HRT 6 h) resulted in the lowest desalination efficiency of $30.2 \pm 1.1\%$. Those results indicate that when the water production rate (or the influent feeding rate) is kept the same, the inter-membrane distance and the HRT have contradictory effects on the desalination efficiency in

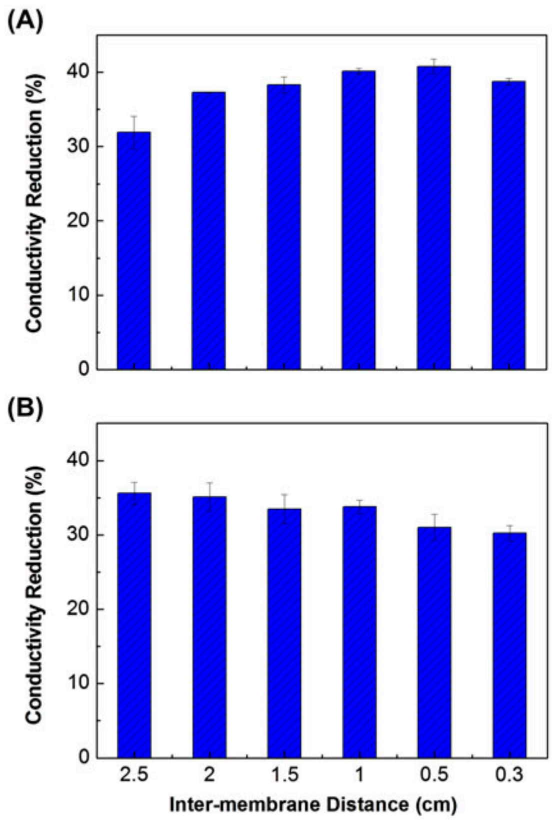

Fig. 2. The conductivity reduction of the salt solution at different inter-membrane distances at two different initial salt concentrations: (A) 10 g/L; and (B) 30 g/L.

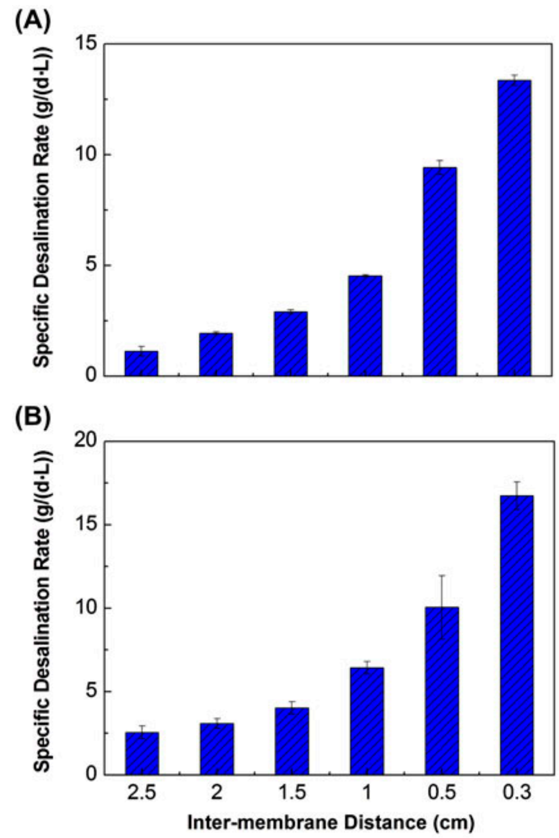

Fig. 3. The SDR at different inter-membrane distances at two different initial salt concentrations: (A) 10 g/L; and (B) 30 g/L.

an MDC. The smallest distance of 0.3 cm did not perform better than larger distances in terms of overall desalination efficiency due to a short HRT. The effect of the HRT is more significant at a higher initial salt concentration because more salts require more time to be removed. Therefore, it is reasonable to conclude that HRT has a greater effect than the inter-membrane distance at a higher initial salt concentration, and a larger distance will result in more salt removal at a fixed influent flow rate.

However, when we included the factor of salt chamber volume in the evaluation by using a SDR, we observed a significantly increasing trend along the decreasing inter-membrane distance with both initial salt concentrations (Fig. 3). At the initial salt concentration of 10 g/L, the highest SDR was 13.4 ± 0.2 g/(d-L) at the inter-membrane distance of 0.3 cm, which was twelve times of the lowest SDR of 1.1 ± 0.2 g/(d-L) at 2.5 cm. With 30 g/L, the highest SDR of 16.7 ± 0.8 g/(d-L) was also obtaine at the inter-membrane distance of 0.3 cm and was about seven times of the lowest SDR, 2.6 ± 0.4 g/(d-L) at 2.5 cm. The advantage of the 0.3 cm distance became less significant compared with the 2.5 cm distance at a higher initial salt concentration. At the same inter-membrane distance, the SDR improved by 1.1–2.3 times at 30 g/L compared with those at 10 g/L. The reduced effect of the smaller inter-membrane distance and the elevated SDR at a higher initial salt concentration were likely due to a greater conductivity of the salt solution when more salts were supplied to the middle chamber. We also observed increasing electric field strength along the decreased inter-membrane distance, confirming that the driving force of desalination in an MDC is the electric field (Fig. 4).

The calculated charge transfer efficiency (the relationship between the salt removal and the electric current) was above 100%, indicating that electric current contributed to a part of salt removal, and conductivity reduction in the middle chamber was also caused by other factors such as water osmosis, dialysis, or ion exchange [3,5]. The theoretical analysis shows that with the initial salinity of 10 g/L, the electric current contributed to $75.0 \pm 5.4\%$ of the conductivity reduction at an inter-membrane distance of 0.3–2.0 cm; at 2.5 cm, we observed a higher contribution from the electric current that reduced

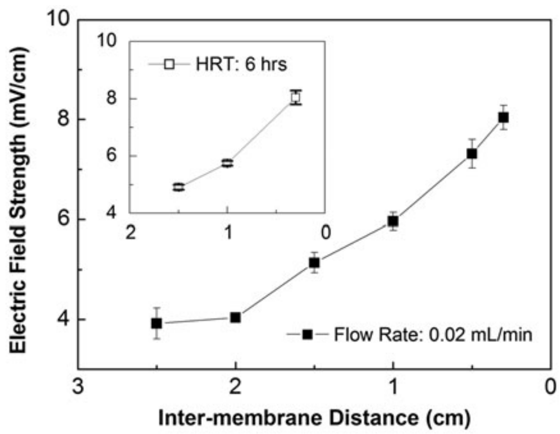

Fig. 4. The electric field strength at different inter-mem brane distances when the salt feeding rate was same at 0.02 mL/min. Inset figure: the electric field strength with 0.3, 1.0, and 1.5 cm inter-membrane distance at the same HRT of 6 h. Note: electric field strength was calculated based on the distance between the anode and the cathode electrodes, instead of the inter-membrane distance.

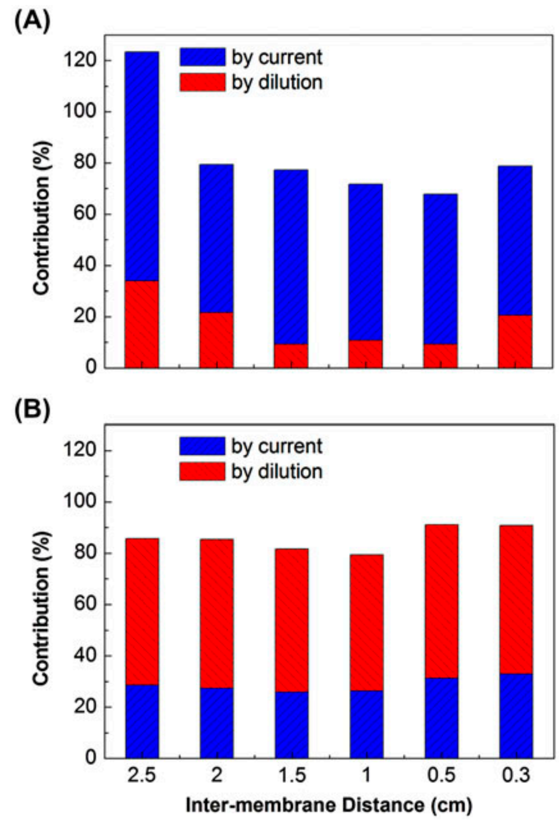

Fig. 5. The contributions to conductivity reduction by electric current (blue) and dilution (red) at two different initial salt concentrations: (A) 10 g/L; and (B) 30 g/L. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

82.3 ± 11.7% of the conductivity. Water osmosis was identified as another major contributor to the conductivity reduction via dilution. An inconsistent trend of the dilution effect was found; for instance, at the inter-membrane distances of 0.5, 1.0, and 1.5 cm, the dilution contributed to about 10% of the conductivity reduction, and at 0.3, 2.0, and 2.5 cm, the dilution resulted in 20.6 ± 1.0, 21.6 ± 3.3, and 33.9 ± 15.7% of the desalination, respectively (Fig. 5(A)). The higher initial salt concentration intensified the dilution effect because of the stronger water osmosis due to a higher salinity gradient between the middle chamber and the anode/cathode chambers. At an initial salt concentration of 30 g/L, it was found that over 50% of the conductivity reduction was caused by dilution; thus, dilution became a major mechanism of desalination over the electric current that contributed to about 30% of conductivity reduction (Fig. 5(B)).

The overall electricity generation was evaluated by using polarization curves. At the initial salt concentration of 10 g/L, we did not observe obvious difference in electricity generation among the three inter-membrane distances of 1.5, 1.0, and 0.3 cm. The maximum power normalized by the anode chamber volume was close to 40 W/m^3 and the maximum current density was about 67 A/m^3. The indistinguishable polarization behaviors at the three different distances indicated that varying inter-membrane distances within the tested range did not significantly change the internal resistance of the MDC. It should be noted that this occurred under the condition of the same salt influent flow rate, which supplied the same amount of salt into the MDC with different inter-membrane distances. The internal resistance was governed by the electrolyte conductivity; although different inter-membrane distances created different travel distances for ions, the sufficient salinity (due to a low performance of desalination) provided enough ions to transfer charges to meet the need of the current generation. As a result, current generation was not obviously changed by varying inter-membrane distances. However, the internal resistance of the MDC would be varying under other testing conditions that are addressed in the following sections.

3.2. *Different inter-membrane distances at the same HRT*

The previous tests contained two variables, the inter-membrane distance and the HRT. Because the HRT can greatly affect the desalination efficiency, we used a fixed HRT of 6 h and examined the influence of the inter-membrane distance on the MDC performance. Three inter-membrane distances, 1.5, 1.0, and 0.3 cm, were selected for the test and the initial salt concentration was 10 g/L NaCl. As shown in Fig. 7(A), the desalination efficiency increased with decreasing inter-membrane distance and the three distances (from large to small) achieved a conductivity reduction of 9.6 ± 0.6, 12.2 ± 0.7, and 38.7 ± 0.4%. Likewise, the SDR increased from 2.4 ± 0.5 g/(d-L) with 1.5 cm to 13.4 ± 0.2 g/(d-L) with 0.3 cm and the electric field strength also increased with decreasing inter-membrane distance (the inset of Fig. 4).

The internal resistance of the MDC with 0.3 cm inter-membrane distance was 342 Ω, much higher than that of the MDC with 1.0 cm (222 Ω) or 1.5 cm (214 Ω) distance. The difference in the internal resistance was related to electrolyte conductivity or salinity of salt solution in the middle chamber. At the same HRT, the smaller inter-membrane distance resulted in much less salt solution and salts fed into the middle chamber. Consequently, the electrolyte conductivity with 0.3 cm inter-membrane distance was lower than those with larger distances. As previously addressed, the internal resistance of the MDC was largely affected by electrolyte conductivity. Therefore, the MDC with smaller inter-membrane distance had a larger internal resistance than that with larger distance. However, the generation of electric current was similar at the three distances and varied around 1.46 mA, which was not expected. The polarization curves with those three distances exhibited significant difference in current and power production (Fig. 6). We found that a current of 1.46 mA was within a low current zone (power curve had not reached its maximum) and accidently the three distances had similar performance in this zone. Once current generation was beyond 2 mA, the difference became much more significant, and 1.5 cm distance had the best performance while 0.3 cm distance showed the lowest, which matched their internal resistance. The power curve with 1.0 cm distance showed an overshoot at its high-current zone, which is related to the measurement procedure [19].

However, the amount of water transported into the middle chamber via osmosis became larger at a bigger distance (Fig. 7(A)). When the inter-membrane distance increased from 0.3 to 1.0 cm, the amount of transported water increased from 2.5 to 5.8 mL. When we further increased the inter-membrane distance to 1.5 cm, there was an additional 7.2 mL of water in the middle chamber. The higher water osmosis at the larger distance was caused by the faster influent flow rate. To maintain an HRT of 6 h, the influent flow rate at the 0.3 cm distance was 0.02 mL/min, while 1.5 cm distance had a flow rate of 0.1 mL/min. The faster influent flow rate brought in more salt per unit time and thus created a larger salt gradient across the ion-exchange membrane, thereby accelerating the water osmosis. The additional water contributed to conductivity reduction, and in general the generation of electric current accounted for more than 50% of

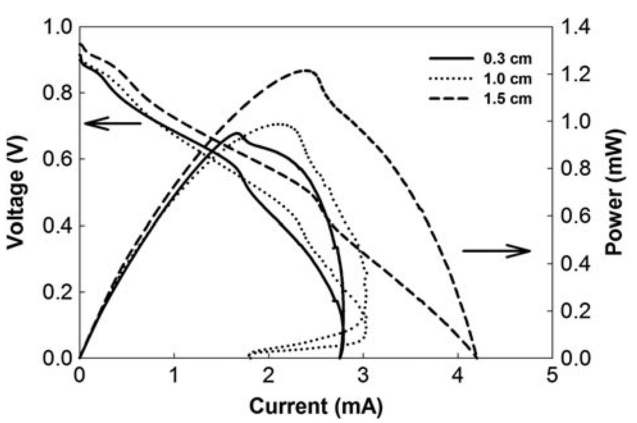

Fig. 6. The polarization curves (power and voltage vs. current) of the MDC with 0.3, 1.0, and 1.5 cm inter-membrane distance at the same HRT of 6 h.

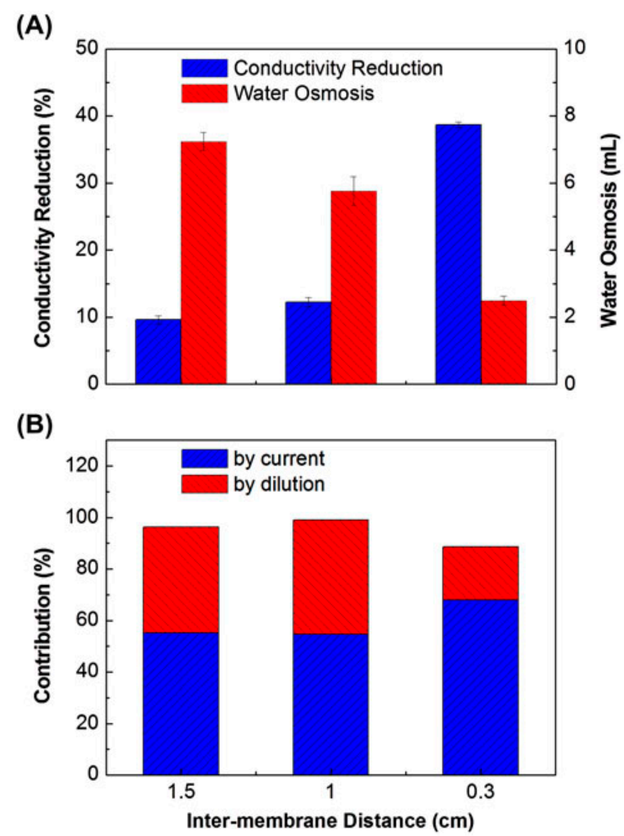

Fig. 7. The desalination performance of the MDC with different inter-membrane distances at the same HRT of 6 h: (A) conductivity reduction (blue) and the amount of water osmosis (red); and (B) The contributions to conductivity reduction by electric current (blue) and dilution (red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

desalination at 1.0 and 1.5 cm distances or more than 60% at 0.3 cm distance (Fig. 7(B)). Although the 0.3 cm distance resulted in the best desalination performance, it produced the least amount of water because of the smallest salt chamber and the slowest influent flow rate.

3.3. Different HRTs at the same inter-membrane distance

We have also examined different HRTs (6 and 20 h) at the same inter-membrane distance (1.0 cm). The results showed that the conductivity decreased by 12.2 ± 0.7% at HRT of 6 h and 35.2 ± 0.1% at HRT of 20 h (Fig. 8(A)), confirming that a longer HRT benefits desalination in an MDC. The internal resistance of the MDC at HRT 20 h was 339 Ω, higher than 222 Ω at HRT 6 h, because a longer HRT had less salt influent fed into the middle chamber and thus a lower salt flux, resulting in lower electrolyte conductivity. A shorter HRT, on the other hand, resulted in more water osmosis that had 5.8 mL additional water in the middle chamber at HRT 6 h, higher than 2.9 mL at HRT 20 h. It was found that electricity generation contributed to 68% of the desalination at HRT 20 h, higher than 55% at HRT 6 h (Fig. 8(B)). The results confirmed that at electrolyte conductivity, electric current plays a more important role in reducing conductivity than dilution effect.

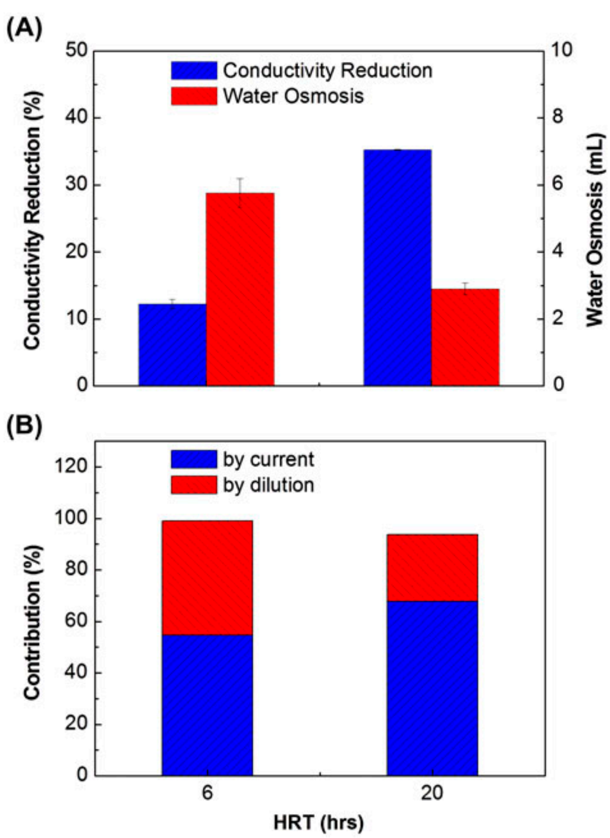

Fig. 8. The desalination performance of the MDC with same inter-membrane distance of 1 cm at two different HRTs of 6 and 20 h: (A) conductivity reduction (blue) and the amount of water osmosis (red); and (B) The contributions to conductivity reduction by electric current (blue) and dilution (red). (For interpretation of the references to color in this figure legend,the reader is referred to the web version of this article.)

3.4. SDR and inter-membrane distances

We summarized the SDR in the literature and plotted the data against the inter-membrane distance (Fig. 9). There is not an obvious trend that SDR increases with decreasing distance; however, high SDRs were generally obtained with small inter-membrane distances. In addition to inter-membrane distances, factors such as the initial salt concentration, the number of desalination chambers, the electrolyte conductivity, and the reactor configuration affect the SDR. The highest SDR was 61.0 g/(d-L) at an inter-membrane distance of 1.0 cm and an initial salinity of 20 g/L; the SDR decreased when more desalination chambers were added in the same study [7]. The lowest SDR in the literature was 0.2 g/(d-L) with an inter-membrane distance of 1.0 cm and an initial salinity of 0.7 g/L [15]. The significant difference in SDRs at the same inter-membrane distance between the two studies was likely due to the extremely low salt concentration in the latter in which, only a small amount of salt was provided for the ion exchange and resulted in very low conductivity of the electrolyte for electricity generation. The smallest inter-membrane distance reported was 0.13 cm, with an SDR of 4.7 g/(d-L) at an initial salinity of 35 g/L [6]. The largest

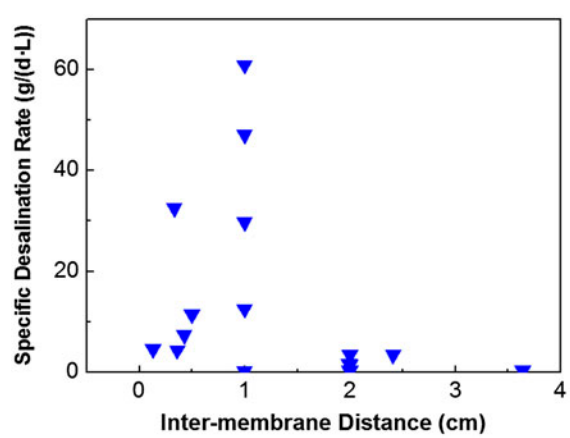

Fig. 9. SDR vs. inter-membrane distances in the MDC studies (the data were analyzed from the previous literature).

inter-membrane distance in the previous studies was 3.6 cm, and an SDR of 0.4 g/(d-L) was achieved using actual wastewater as a carbon source in the anode [2]. Because there is limited literature on MDCs and a significant difference in MDC configuration and operation among different studies, we were not able to extract enough information to conclude a clear relationship between SDR and inter-membrane distance, but smaller distances seem to be generally beneficial.

4. Conclusions

Our results show that a small inter-membrane distance results in a higher specific desalination rate; however, a larger inter-membrane distance does not negatively affect the desalination efficiency at the same influent flow rate, because the increased HRT improves the desalination performance. At the same HRT, a smaller inter-membrane distance improves the desalination efficiency, but a low water production rate remained as a drawback. Internal resistance was significantly affected by electrolyte conductivity. In conclusion, the inter-membrane distance and the HRT have complementary effects on desalination performance in MDCs, and future design and operation of MDCs need to consider the trade-off between these two factors.

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