



## Effect of the organic load on salt removal efficiency of microbial desalination cell

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### ABSTRACT

Microbial desalination cell presents a novel concept for wastewater treatment, salt removal and bioelectricity generation in a single system. In this study, the effect of organic load on salt removal in air-cathode up-flow microbial desalination cell (UMDC) was investigated in batch mode. Four different organic concentrations were fed to the anodic chamber of microbial desalination cell. The initial chemical oxygen demand in the anodic chamber was 2,024; 4,048; 6,074 and 8,096 mg/L. Real seawater was used in the desalination chamber of UMDC. The obtained results showed that the organic load 3 (initial COD of 6,074 mg/L) produced a maximum percentage of salt removal compared with other organic loads tested. The maximum power density production (1,769 mW/m<sup>2</sup>), the maximum percentage COD removal (95%) occurred with 48% salt removal in the microbial desalination cell using the organic load of 6,074 mg/L. The obtained results showed that microbial desalination cell is a unique system that provide wastewater treatment and salt removal in a single system. Organic load is a very important factor for the operation of UMDC, there is an optimum value that achieves the best performance of the UMDC system.

*Keywords:* Microbial desalination cell; Seawater desalination; Wastewater treatment; Bioelectricity generation

### 1. Introduction

In recent years, the world energy consumption increases as the population increases. Due to the increase in population, drinking water demands also increase, but current resources are insufficient for these demands. Energy consumption is very high in conventional wastewater treatment and water desalination, so there is a need to develop a new system that needs less energy with higher output. In the last decade, bioelectrochemical systems (BES) emerged for wastewater treatment and energy production and other functions [1,2]. Conventional drinking water treatment processes such as reverse osmosis and electrodialysis need very high energy and they only separate the pollutants that need further treatment. The development of microbial desalination cell (MDC) provides a unique solution for seawater desalination and

wastewater treatment in a single system. MDCs are developed from the microbial fuel cell (MFC) concept in which one additional chamber is added namely desalination chamber between anodic and cathodic chambers [3–6]. In the anodic chamber organic substrate such as acetate rich solution, synthetic wastewater or real wastewater are used as substrate. The anodic chamber is considered as powerhouse for the MDC, in this chamber, microbes donate their electrons to the anode electrode and these electrons later pass to the cathode electrode through an external circuit. The final reaction occurs at the cathode electrode in the presence of oxygen. Recently air cathode is used in MDC; this provides more sufficient and effective system for reducing the overall cost. Recently, this system attracted attention of several investigators [7,8].

In general, MDC has three different components, anodic chamber, desalination chamber and cathodic chamber [9]. Over the time, different MDC designs are proposed by different researchers to enhance the performance of the process.

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These designs include stack MDC, tubular MDC and air-cathode MDC [9–14]. Stacked MDC system has more desalination chambers that can remove more salts. In stacked MDC system, due to increase in number of chambers, the total desalination rate increases as the charge transfer efficiency increases (more pairs of ions separated from the salt solution while one electron pass through the external resistance). In general, the total desalination rate can be increased with increasing the number of desalination chambers and reducing the external resistance. In stack MDC, to reduce the cost, air-cathode MDC provides a suitable system for removal of COD and salt. In air-cathode MDC system, there is no separate cathode chamber and cathode electrode is directly exposed to the air. In this system, catholyte is recycled and fed from the upper part of the cathode. In this work, the effect of different anode organic loads on the performance of air-cathode up-flow MDC (UMDC) was investigated. In the desalination chamber, seawater was used. Four different organic loads were investigated, and their effects on the COD removal, salt removal efficiency and power generation were reported.

## 2. Materials and methods

### 2.1. Construction and operation of UMDC

The UMDC was constructed using tubular anion and cation exchange membranes. The two chambers were separated by an anion-exchange membrane (AMI-7001, Membranes International, Inc., USA) between the middle chamber and the anodic chamber, and a cation exchange membrane (CMI-7000, Membranes International, Inc., USA) between the middle chamber and the air cathode. Carbon cloth was used as cathode electrode and it is coated with 5% Pt catalyst on one side. Fig. 1 shows a schematic diagram of the used up-flow MDC. A carbon fibre brush with a titanium wire (Gordon Brush, USA) was used as anode electrode. Both the anode and desalination chamber held a liquid volume of 300 and 100 mL, respectively. The cathode used in this UMDC system was air cathode. In this work, four different organic loads of synthetic wastewater and natural seawater were used. The organic loads used were referred to as organic load 1 (2,024 mg/L), organic load 2 (4,048 mg/L), organic load 3 (6,074 mg/L) and organic load 4 (8,096 mg/L). The UMDC was operated in batch mode of operation.

### 2.2. Medium and microorganisms

In all experiments, synthetic wastewater was used. The synthetic wastewater (glucose X g/L, yeast extract 0.34 g/L, ammonium chloride 0.84 g/L, potassium dihydrogen phosphate 0.136 g/L, di-potassium hydrogen phosphate 0.234 g/L, magnesium chloride hexahydrate 0.084 g/L, ferric chloride 0.05 g/L, sodium thioglycolate 0.1 g/L) was used in the anodic chamber. The concentration of glucose varied to obtain higher COD values. The pH of the anolyte was kept  $6.9 \pm 0.1$ . This wastewater has COD values of 2,024; 4,048; 6,074 and 8,096 mg/L. High COD wastewater can be found in food industry such as dairy wastewater. The wastewater samples were characterized for general wastewater parameters. Real seawater was used in the desalination chamber.

The seawater was collected from Al Khor, Doha, Qatar. This collected seawater has a salinity of 37.5 psu, which is higher than the average salinity of global sea of 35.5 psu, due to the nature of the Arabian Gulf and the high evaporation rate. The initial conductivity of real seawater was 64 mS/cm. The cathode chamber was fed with acidic water (adjusted with sulphuric acid solutions at pH of 2). The anodic chamber was inoculated from a previously running MDC that has been in operation for 8 months with synthetic wastewater as an anodic substrate. The previously enriched bioanode consortium was initially inoculated with anaerobic sludge collected from local wastewater treatment plant (Doha, Qatar) and was running in batch mode. All the UMDCs running experiments were operated at room temperature in the range of  $25^\circ\text{C} \pm 2^\circ\text{C}$ . All the UMDC experiments were conducted in batch mode of operation.

### 2.3. Measurements and analysis

The electric output of each MDC was measured in millivolts (mV) against time using a Fluke 289 multimeter. The voltage was recorded every 20 min against respective external resistance of 1 ohm and 100 ohm. The current  $I$ , in mill amperes (mA) was calculated using Ohm's law,  $I = V/R$ , where  $R$  is the known external resistance and  $V$  is the measured voltage in millivolts (mV). The power density was calculated as  $P = (I \times V)/A$ , where  $A$  is the surface area of the cathode electrode. The volumetric power density was calculated as  $P_{\text{vol}} = P/V$ , where  $P$  is the power output and  $V$  represent the volume of the anodic chamber. The conductivity, total dissolved solids and pH were measured by benchtop pH/conductivity meter (Orion Star A215, Thermo Scientific, MA, USA). The wastewater was characterized according to the standard method (COD was determined using a COD testing kit (DR LANGE). Electrochemical measurements were conducted using a potentiostat SP-150 (Bio-logic SPS, France).

## 3. Results and discussion

### 3.1. Effect of organic load on UMDC power generation

The UMDC was operated for three consecutive cycles using each of the four organic loads used, each cycle last for 10 d. After 10 d, the feed was changed in both anodic and desalination chamber of the UMDC. The UMDC was connected with 850-ohm external resistance for each load. Cell voltage across the 850 ohm resistance was continuously recorded by a potentiostat every 20 min interval. This external resistance was determined by polarization curve. Fig. 2 shows the behaviour of power density (with respect to the surface area of electrode) at different organic loads.

The maximum power density of each day was calculated and plotted vs. time for each feed. Fig. 2 shows that in the first cycle, during the initial 2 d, using all the organic loads, the profile is the same, as all microbes were in the lag phase. After lag phase, difference in power production was observed after day 3. Power density increases rapidly in period between the 3rd to 7th day with all organic loads. After eight days, a rapid decrease in power density was observed due to the depletion of substrate in the anodic chamber of the UMDC. After the 10th day, the media were replaced in the anodic and

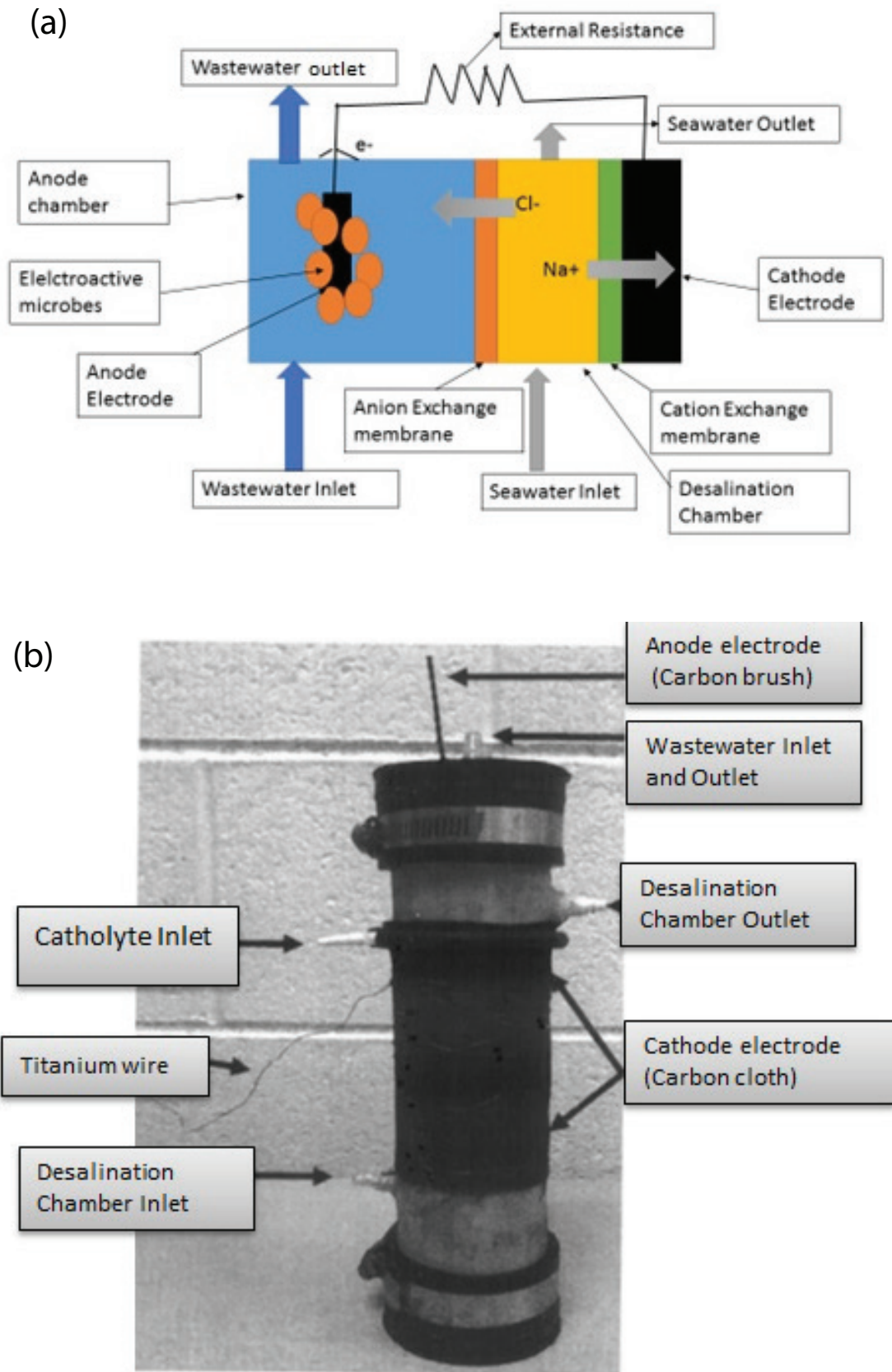


Fig. 1. (a) A schematic diagram of up-flow microbial desalination cell (UMDC). (b) Real UMDC operated in the lab.

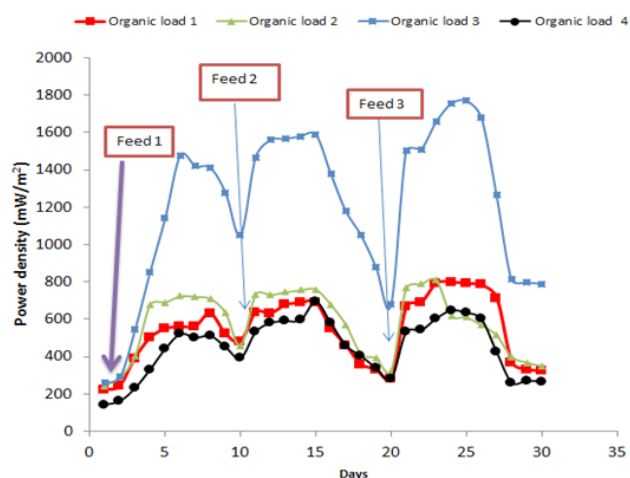


Fig. 2. The power density behaviour for the three different feed cycles at four different organic loads in the UMDC.

desalination chamber of the MDC. Since microbes were active for the last 10 d, they made biofilm on the anode electrode and sudden increase in power density was observed after addition of the new feed in the anodic chamber. The power density was suddenly increased and stayed stable up to day 15. After 15 d, again power density starts to decrease, this time decrease in power started before 2 d as compared with the first cycle. Possible explanation is that in the second cycle microbes were more active and electroactive microbes were enriched in the anodic chamber. After 20 d, the feed again replaced with new media, a sudden increase in power density was observed. In this UMDC, the third cycle produced higher maximum power compared with the first two cycles. As shown in Fig. 2, similar behaviour was observed in each cycle, but the lag time was reduced after the first cycle due to the improved activity of electroactive microbes in the anodic chamber. In all the four organic loads tested, organic load 3 produces maximum power density in the system. The maximum power density in the first cycle for organic loads 1, 2, 3 and 4 was 630, 723, 1,478 and 520  $\text{mW/m}^2$ , respectively. These results showed that at very high organic load, bacteria did not produce higher power due to substrate inhibition [1]. The maximum power density obtained in the second cycle using organic loads 1, 2, 3 and 4 was 690; 760; 1,589 and 693  $\text{mW/m}^2$ , respectively. So clearly, more power was produced in the second cycle compared with the first one. A possible explanation for this behaviour is that lag time was reduced for the microbes and more electroactive bacteria were acclimatized in the anodic chamber [7,8]. The power production in the third cycle using organic loads 1, 2, 3 and 4 was 798; 809; 1,769 and 647  $\text{mW/m}^2$ , respectively. The trend of power density change with time was similar in all the three cycles, even the power density increased in every cycle. The maximum power density was achieved with organic load 3 (6,072  $\text{mg/L}$ ). At very high organics concentration, the obtained power production was less. This might be due to inhibition at very high concentration of substrate occurred to the electroactive bacteria and also some methanogens might be active at these conditions [6].

The maximum current density of each day was calculated and plotted vs. time for each feed. Fig. 3 shows that

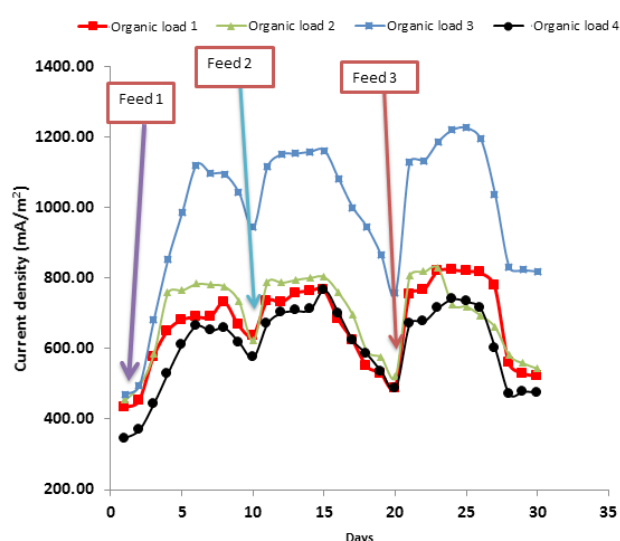


Fig. 3. The current density behaviour for the three different feed cycles at four different organic loads in the UMDC.

in the first cycle, during the initial 2 d for all the organic loads, the profile is the same, as all microbes were in the lag phase. After lag phase, difference in current generation was observed after day 3. Current density increased rapidly in the period between 3rd and 7th day with all organic loads. After 8 d, a rapid decrease in current density was observed due to the depletion of substrate in the anodic chamber of the UMDC. After the 10th day, the media were replaced in the anodic and desalination chamber. Since microbes were active for the last 10 d, they made biofilm on the anode electrode and sudden increase in current density was observed after addition of the new feed in the anodic chamber. The current density was suddenly increased and stayed stable up to day 15. After 15 d, again current density starts to decrease, this time decrease in current started before 2 d as compared with the first cycle. Possible explanation is that in the second cycle microbes were more active and electroactive microbes were enriched in the anodic chamber. After 20 d, the feed again replaced with new media, a sudden increase in current density was observed. In this UMDC, the third cycle produced higher maximum current compared with the first two cycles. As shown in Fig. 3, similar behaviour was observed in each cycle, but the lag time was reduced after the first cycle due to the improved activity of electroactive microbes in the anodic chamber. In all the four organic loads tested, organic load 3 produced maximum current density in the system. The maximum current density in the first cycle for organic loads 1, 2, 3 and 4 was 731; 783; 1,120 and 664  $\text{mA/m}^2$ , respectively. These results showed that at very high organic load, bacteria did not produce higher current density probably due to substrate inhibition [1]. The maximum current density obtained in the second cycle using organic loads 1, 2, 3 and 4 was 765; 803; 1,162 and 767  $\text{mA/m}^2$ , respectively. The maximum current density obtained in the third cycle using organic loads 1, 2, 3 and 4 was 823; 818; 1,226 and 742  $\text{mA/m}^2$ , respectively.

The growth condition of electroactive microbes and methane producing bacteria is almost similar and the inoculum was also collected from anaerobic digestion plant.

If the methanogens are active they caused electron loss and less number of electrons are available for bioelectricity production. Therefore controlling of methanogenesis can result in an increase in Coulombic efficiency [15].

### 3.2. Effect of organic load on COD removal

In BES system, the cell's powerhouse is the anodic chamber media, which provide food to microbes and then these microbes deliver electron to the anode electrode. The performance of UMDC is also affected by the anolyte media used. In this study, four different organic loads were used to find out the optimum organic load and its effect on salt removal in the UMDC. Based on the produced electrons, salt removal efficiency is determined. Fig. 3 shows the percentage COD removal with time using the four different organic loads in the anodic chamber. The feed was changed every 10 d. COD samples were collected each day for all four different organic loads.

In the first cycle, the COD reduction rates were slow for the first 5 d in all the organic loads. The slow removal in COD at the starting time can be explained as microbes were in the lag phase and COD reduction rate was lower. As explained above in this period, power density was also lower compared with other periods. After 5 d, the % COD reduction rate was accelerated and it was maximum on day 9 for cycle 1. The maximum % COD removal rate for cycle 1 using organic loads 1, 2, 3 and 4 was 84%, 86%, 92% and 32%, respectively. This COD reduction behaviour is comparable with power production in the same periods. At the highest organic load used, the % COD reduction rate was lower compared with the lower organic loads. For the organic loads 1, 2 and 3, the % COD reduction increases as the influent COD concentration increases. At these conditions, electroactive microbes are most active in the anodic chamber of UMDC. The power generation also increases as the % COD reduction increases, but using organic loading 4, % COD reduction rate decreased due to the high initial organic load and the time period is only 10 d. At higher concentration, methanogen bacteria may be also active and due to that all electrons were not transferred to the anode electrode hence the overall power production starts to decrease. However, the amount of COD reduction is almost similar, but due to the high organic load, in 10 d it is difficult to reduce that much of COD.

Compared with the power generation profile (Fig. 2), at the highest % COD removal level, low power was obtained. One possible reason is that may be at this high concentration of substrate, methanogen bacteria might be active and hence the electroactive microbe's activity was reduced [15]. After the first 10 d, the second cycle was started; all feeds were replaced in the system. Due to the shorter lag phase, the % COD reductions were higher compared with the first cycle. The maximum % COD reduction for cycle 2 using organic loads 1, 2, 3 and 4 was 86%, 88%, 94% and 46%, respectively. In the second cycle, more COD was removed in all the feeds.

The highest % COD reduction was obtained using organic load 3 (6,074 mg/L). In this case, the % COD reduction was very fast in the initial 3 d, around 50% COD was removed in the first 3 d in all the four organic loads. After 20 d, the third cycle started. As Fig. 4 shows, after changing the feed, the % COD removal increased drastically and it was

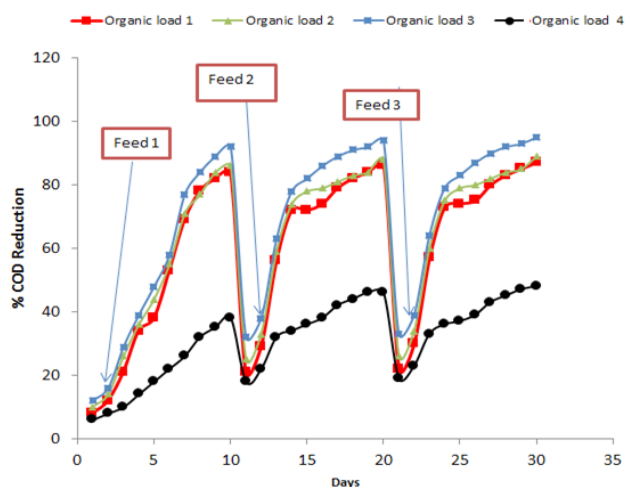


Fig. 4. Percentage of COD reduction at various organic loads in the up-flow microbial desalination cell.

much higher compared with the first cycle. The maximum % COD reduction for the third cycle using organic loads 1, 2, 3 and 4 was 87%, 89%, 95% and 48%, respectively. In the third cycle, the maximum % COD reduction was obtained using organic load 3. Compared with the second cycle, in the third cycle, faster COD reduction was observed due to the higher activity of microbes.

### 3.3. Effect of organic load on salt removal

The main advantage of MDC is that it can treat wastewater in the anodic chamber and simultaneously remove salt from the desalination chamber. The anodic substrate works as an electron donor to the cathode electrode. Also due to the salt removal from the desalination chamber (due to passage of ions to the anodic chamber) conductivity of anolyte was increased. This will help increase the low conductivity of wastewater such as groundwater and municipal water. By increasing the conductivity, more electrons were transferred to the anode electrode, which is the key for desalination. Four different organic loads to the anodic chamber were tested with a constant composition of desalination chamber (seawater). Fig. 4 shows the behaviour of salt removal from seawater with time in UMDC using different organic loads.

Fig. 5 shows the percentage of salt removal behaviour in the UMDC at different organic loads. In this work, the salt removal was determined as reduction in conductivity of seawater. As shown in Fig. 5, the pattern is almost similar to that of the % COD reduction in the anodic chamber. During the desalination process, chloride ions were transferred to the anodic chamber while sodium ions were transferred to the cathodic chamber. The catholyte pH increases because of the reaction of oxygen with electrons and protons. The initial pH of catholyte was 2 and after 2 d of operation, it reached to 5, therefore, every 2 d the catholyte was changed during the experiments.

In the first cycle, for the initial 6 d, the percentage salt removal was small but in the 7th to 8th days it was higher. Since in the initial period not much electrons were available

due to the lag phase of the microbes. After 10 d, all the media were replaced with new one to start the second cycle. The maximum percentage of salt reduction for cycle 1 using organic loads 1, 2, 3 and 4 was 42%, 43%, 46% and 19%, respectively. These results demonstrate that low organic load for the initial cycle did not have much effect on the salt removal. As the obtained results showed that for the first three organic loads, the % salt removal was in range of 42% to 46%. Here also at the highest organic load, less salt was removed, as previously explained. At this organic load, less % COD was reduced due to the presence of methanogens in the anodic chamber. After 10 d of operation, all the feeds were changed.

The second cycle was conducted from the 11th to 20th day. As compared with the first cycle, salt removal was faster due to the short lag phase of the microbes. The highest % salt removal was obtained after 14 d. The maximum % salt reduction for cycle 2 using organic loads 1, 2, 3 and 4 was 43%, 44%, 48% and 23%, respectively. Using organic load 3, the highest % of salt removal was achieved. After the 20th day, the third cycle was started, again all the feed was replaced in the UMDC. The % salt removal for the first 3 d was faster than that of cycle 2. On the 23rd day, the maximum % salt removal was achieved. The maximum % salt reduction for cycle 3 using organic loads 1, 2, 3 and 4 was 43.5%, 44.5%, 48% and 26%, respectively. These results show that there is

no significant difference between the organic load 1 and 2 in terms of salt removal although the second organic load was double than that of the first organic load. The highest % salt removal was achieved with the organic load 3. Here also at the highest organic load, the lowest % of salts was removed but as a value, it was higher compared with that of cycle 1 and cycle 2.

### 3.4. Discussions

In this work, the effect of organic load on salt removal from the desalination chamber of UMDC was investigated. The main theme for this work is how the UMDC work with different organic loads in the anodic chamber. The obtained results confirm that there is a huge difference in power generation at different substrate organic loads. The same behaviour was also seen in terms of the percentage of COD reduction. As the anolyte media has anaerobic bacteria with an electroactive type, they have the ability to donate their electrons to the anode electrode. In this work, the produced power density was increased using organic loads 1 to 3, but it was decreased using organic load 4. Nam et al. [16] reported the effect of organic load in a single chamber MFC. When organic load increased to 3.84 mg/L, the maximum produced power density increased to 2.98 W/m<sup>2</sup> from 1.88 W/m<sup>2</sup> [16]. These results showed that at higher organic loads, there might be a chance for the presence of methanogenic bacteria, therefore not enough electrons transferred to the anode electrode. In general, when electroactive bacteria consume the organic content, the final terminal electron acceptor is oxygen and it reacts at the cathode electrode. When both electroactive and methanogenic microbes are present in the anodic chamber, then the net electron transfer to the cathode electrode decreases. For methanogens, the final electron acceptor is carbon dioxide. Therefore, it is very important to control the pH of the anolyte and organic load of anodic medium to suppress the growth of methanogenic bacteria. In this case, electrons are transferred to the final electron acceptor, carbon dioxide. Kim et al. [17] investigated the effect of organic load in a single chamber MFC. They showed that the produced power increases as the organic load increases [17]. Luo et al. [10] operated an MDC for a period of 8 months, their observation showed that MDC performance decreases over time. In their work, the desalination efficiency, current density and Columbic efficiency decreased by 27%, 47% and 46%, respectively [10]. Based on the power generation, similar behaviour was also obtained with % COD reduction. As the flow rate increases more power was generated and more

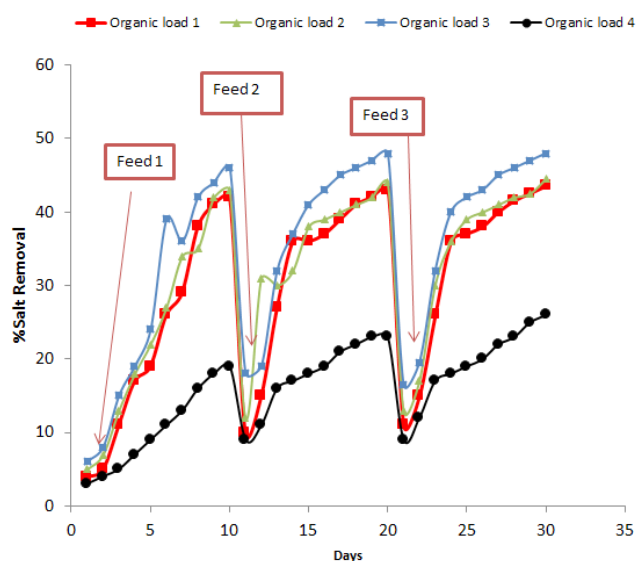


Fig. 5. Percentage salt removal in the desalination chamber of UMDC at different organic loads in the anodic chamber.

Table 1  
Performance of the UMDC in the last cycle of operation (cycle 3)

Organic load No.	Organic load (mg/L)	Maximum power density (mW/m <sup>2</sup> )	Maximum COD removal (%)	Maximum salt removal (%)
1	2,024	798	87	43.5
2	4,048	809	89	44.5
3	6,074	1,769	95	48
4	8,096	647	48	26

% COD removal was achieved. Tamilarasan et al. [18] investigated the effect of organic load on bioelectricity production in up-flow anaerobic MFC treating surgical cotton industry. In their work, they showed that increasing the loading rate increased the power production [18]. A maximum power production of 2.2 W/m<sup>3</sup> was obtained with organic loading of 1.9 g COD/L with total COD removal of 79% using up-flow MFC [17]. In our work, there was not much difference in salt removal in the first three organic loads in the third cycle (Table 1). In addition, almost equal percentage of salt was removed in the first and second cycles as compared with the third cycle using the first three organic loads. This is because the UMDC was operated at higher power generation mode. At the higher power production mode, less salt is removed compared with the high current generation mode [8].

#### 4. Conclusions

In this work, the effect of organic load on the percentage salt removal was investigated in an UMDC. Four different organic loads were tested in the anodic chamber with constant natural seawater in the desalination chamber of air-cathode UMDC. The organic load showed to have significant effect on the % COD reduction. UMDC performs better with organic loads of 4,048 and 6,074 mg/L in the anodic chamber. At very low organic load, there are not enough electrons available for redox reaction at the cathode chamber. At very high organic load (8,096 mg/L) there was a chance for the presence of methanogens that suppress the electroactive microbial activity. Using an organic load of 6,074 mg/L, the highest power density, % COD reduction and % salt removal achieved were 1,769 mW/m<sup>2</sup>, 95% and 48%, respectively. The UMDC shows a better solution for wastewater treatment, seawater desalination in a single chamber microbial desalination cell. An optimum organic load exists that maximize the performance of UMDC.

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