Journal of Environmental Chemical Engineering 5 (2017) 5961–5967

Contents lists available at ScienceDirect

Journal of Environmental Chemical Engineering

journal homepage: www.elsevier.com/locate/jece

Reduction of organic compounds in petro-chemical industry effluent and desalination using *Scenedesmus abundans* algal microbial desalination cell

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ARTICLE INFO

Keywords:
Desalination
Bio-cathode
Scenedesmus abundans
Bioelectricity

ABSTRACT

Water and energy shortages are considered to be very serious global issue. As a consequence, it is necessary to develop an alternative water resource such as desalination and wastewater treatment etc. Microbial desalination cell (MDC) is the new, green and environment–friendly technology that desalinates seawater and produces bioelectricity. This study integrate desalination with wastewater treatment and bioelectricity production in MDC by utilizing microalgae as bio-cathode and the results were compared with the chemical cathode. Recently, microalgae have received a high attention for application in bio-electrochemical systems due to its potential to be used for oxygen generation and biodiesel production. The study utilizes petroleum wastewater in the anode chamber and microalgae *Scenedesmus abundans* in the cathode chamber. The results have highlighted that microalgae bio-cathodes perform better than chemical cathode and increases COD removal and produce a considerable amount of bioelectricity. It also proved that high initial salt concentration 35 g Nacl L^{-1}desalinates well when compared with low initial salt concentration 20 g NaCl L^{-1} .The maximum voltage and desalination efficiency of MDC 1 (35 g NaCl L^{-1}) and MDC 2(20 g NaCl L^{-1}) is found to be 654 mv and 55.3%; 506 mv and 42.6% with the volumetric ratio of 1:0.5:1(anode, desalination chamber and microalgae cathode). Thus the experimental results reveal that utilizing bio-cathode MDC proved to be a promising platform for effective desalination of sea water.

1. Introduction

The Social and economic development are searching for the sustainable supply of water and energy. Especially, water security is an urgent global issue faced by many regions of the world and or expected to face the shortage in supply. More than one in six people globally are water stressed, in that they do not have access to safe drinking water. The hydrological cycle greatly depends on climate change and it is expected that climate change increases the global temperature and precipitation levels which in turn affects the availability and quality of water. About 3% of earth's surface is covered by fresh water and rest 97% is covered by sea water. The removal of salt by innovative techniques helps to reduce the fresh water crisis.

Desalination is found to be efficient and promising technology in processing salt water to nearly fresh water. But the energy demands and high operating cost pose challenges in their application. Desalination process requires high-grade electricity for the production of fresh water which results in the emission of excessive waste heat, greenhouse gas (GHG) and high concentration of brine to the environment. Many steps have been taken in past years and succeeded in reducing the capital cost yet the high energy requirement remain a concern. It is important to obtain energy and water from a renewable source. This led to the development of the new technology called Bio-Electrochemical systems (BESs).

The new type of BES called Microbial Desalination Cell (MDC) was developed by the integration of microbial fuel cell and electro-dialysis cell for desalination and wastewater treatment. Cao et al. [1] reported first Microbial Desalination technology for desalination purpose in 2009. Simultaneous organic and salt expulsion along with power production can be done with the help of MDC. The above process can also be combined with reverse osmosis process as a pretreatment process for decreasing the salinity of feed solution and lower the power consumption rate. [2]. The other important aspect include microbial production of electricity called "bio-energy" offers extraction of electric current from wide range of soluble and dissolved complex form of organic wastes and renewable biomass. The major substrates used in treatment process are artificial and real wastewater and lignocellulosic biomass [3]. Improvement in knowledge and innovation in substrate utilizing technology can substantially increase the conversion of biomass into useful energy. MDC also eliminates GHGs which are a backlog

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https://doi.org/10.1016/j.jece.2017.11.017
Received 18 August 2017; Received in revised form 30 October 2017; Accepted 3 November 2017
Available online 04 November 2017

in conventional processes.

MDC comprises of three chambers that are separated by anion exchange membrane and cation exchange membrane. Wastewater containing organic matter is placed in the anode chamber where the biofilms are formed by the multiplication of exoelectrogens and leads to the generation of bioelectricity [4,5]. The biofilms adhere to the anode surface area and start the bio-pollutant oxidation process to releases protons and electrons. The electrons are passed to the cathode via a resistor. At cathode chamber, oxygen is usually used as an electron acceptor that combines with electrons and protons to forms clean water. Thus electrode potential generated across cathode and anode leads to the movement of anions and cations present in the middle desalination chamber through anion exchange membrane and cation exchange membrane resulting in desalination process [6]. Thus the process leads to simultaneous wastewater treatment, desalination and bioelectricity generation. The rate of desalination is the key factor in MDC function. It depends on the initial salt concentration of the sample to be desalinated. MDC is primarily appropriate for desalinating water with high salt content. This may lower the ohmic resistance and result in higher current production and a higher desalination rate (DR).

At the anode:

$$(CH_2O)_n + nH_2O \rightarrow nCO_2 + 4ne^- + 4nH^+. \quad (1)$$

At the cathode:

$$O_2 + 4ne^- + 4nH^+ \rightarrow 2H_2O. \quad (2)$$

The cathode plays an important role i.e., clean electron acceptors in cathode chamber is the key issue of bio-electro chemical systems. Chemical cathodes such as Ferricyanides were used as a catholyte in the first attempt but had a negative effect on water quality. Though it had been used widely, it had some drawbacks such as replacement of catholyte for a particular period of time, expensive and toxic to the environment [1]. Thus an air cathode may be used in which Oxygen is considered to be terminal electron acceptor among other electron acceptors because of its high redox potential, availability and cost effectiveness. It is not toxic when compared with other chemicals. The usage of air cathode is decreasing because of its slow redox kinetics in ambient conditions [7,8]. In order to reduce the activation over potential for oxygen reduction, expensive catalysts such as platinum should be used. Another drawback associated with air cathode is high energy requirement for maintaining dissolved oxygen concentration in the cathode [9].Therefore the application of bio-cathodes is the best alternative to overcome the above drawbacks.

Bio-cathodes called biological cathodes utilize microorganisms to catalyze the reduction reactions. Microalgae and cyanobacteria are considered to be potential bio-cathodes. Especially algae are considered to be a renewable source that can generate green energy [10]. They produce almost 40–50% of oxygen in earth's atmosphere and considered to be primary producers in the ocean that cover 71% of earth's surface [11]. Oxygenic photosynthesis is carried out that eliminates the usage of mechanical aeration. Oxygen availability at cathode depends on oxygenic photosynthesis that requires three membrane-bound proteins such as PSI, PSII and cytochrome b6F complex that helps in electron transport from water to NADP [12].The electrons produced are transferred between protein complexes by small mobile molecules such as plastoquinone and plastocyanin that carries through long distance and plays a significant role in photosynthetic energy conversion. The advantages of bio-cathode includes 1) Enhanced desalination of water 2) Sustainable and self-generating 3) Reduction in start-up time [7]. Further innovation in application of bio-cathodes advanced as electricity driven production technology for chemicals from easily available feedstock carbon dioxide using biocatalysts. The low value carbon dioxide, on long term reduction at the bio-cathode produced acetate and other compounds like ethanol and butyrate. For continuous biochemical separation process, improvement in wide range of biomass retention strategies should be checked thoroughly [13]. Detailed review on advancement in bioelectrochemical approaches of carbon dioxide transformation in terms of process design and biocatalyst development has been elucidated by Bajracharya, et al. [14].

Chlorella and Scenedesmus species are tested in previous research in wastewater treatment and stated that it reduced the cost of operation and maintained [15,16]. Scenedesmus growth is successfully seen in the saline water as it consumes salts and makes use of them in metabolism. The species proved promising results and that paved the development of desalination process up to 95% removal efficiency. The major drawback of using algae is harvesting the biomass [17].

In this paper, the performance of bio-cathode microbial desalination cell was studied by using microalgae, *Scenedesmus abundans* as bio-cathode. The desalination performance and power generation efficiency of bio-cathode was compared with chemical cathode and the current study also assessed the bio-cathode activity with different initial salt concentrations.

2. Materials and methods

2.1. Bio-cathode microbial desalination cell construction

Experiments were carried out in a three-chambered MDC made of low-cost acrylic sheets as shown in Fig. 1. The liquid volumes of three chambers include 500 ml, 250 ml, 500 ml (anode, middle desalination chamber, and cathode) respectively in the ratio of 1:0.5:1. Graphite rods with the surface area of 127 cm^2 were used as anode and cathode electrodes. The chambers were separated by ion exchange membranes. Anode and desalination chamber separated by anion exchange membrane (AEM) (AMI 7001, Membrane International) and cation exchange membrane (CEM) (CMI 7000, Membrane International) separated cathode and desalination chamber. In the present study, the distance between AEM and CEM is fixed as 3 cm. The electrodes were dissolved in 1 M HCl solution for two days and rinsed with Millipore water to eliminate trace materials. Fixed resistance of 1 KΩ was applied between anode and cathode electrodes in order to follow closed circuit. No external aeration or mixing was given.

2.2. MDC operation

Three microbial desalination cells were tested. One MDC with chemical cathode of saline concentration 35 g Nacl L^{-1} was operated as control. Other two MDC with microalgae bio-cathode of different saline concentration i.e., 35 g Nacl L^{-1} (MDC 1) and 20 g Nacl L^{-1} (MDC 2) was carried out. Internal resistance loss is a key factor that effects the power production in MDCs. The reduction of power density and desalination performance with decreasing the salt concentration might be due to increasing the internal resistance of reactor in low salt concentration. Hence in the present Study, desalination performance and electricity generation using higher and middle variant salt concentration have been analyzed in bio-cathode MDC exempting low salt

Fig. 1. Fabricated microalgae bio-cathode desalination cell.

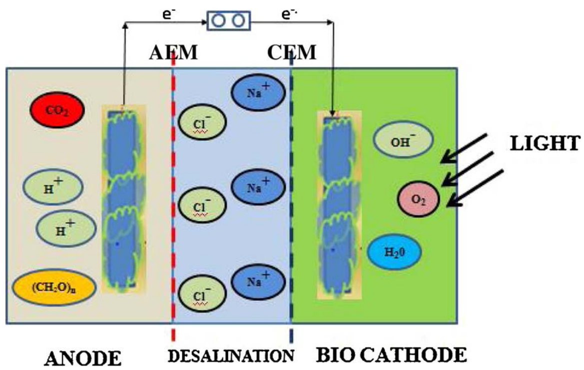

Fig. 2. Schematic representation of microalgae biocathode microbial desalination cell.

concentration.

The set up was operated initially with an open circuit in order to obtain the stability. The anode solution was changed once in a while when the voltage drops to 50 mV. The saline solution and catholyte remain constant. The MDC was operated for 22 days and desalination rates were measured periodically in duplicates. The chemical cathode MDC is taken as the reference for experimentation purpose. Fig. 2 describes the schematic representation of microalgae bio-cathode microbial desalination cell.

2.3. Medium

The anode chamber was fed with real time petroleum wastewater (CPCL, Chennai, India) containing mixed bacterial consortia. It consists of hydrocarbons, ammonia, chlorides, sulfides, bicarbonates, phenol and other constituents in minute concentration. The Chemical Oxygen Demand (COD) of the wastewater is found to be 650 mg L^{-1}.

In chemical cathode, 15 mM concentration of potassium hexaferrricyanide was used. In microalgae biocathode, Scenedesmus abundans grown in the BG-11 medium was fed. BG 11 nutrient solutions contain Sodium nitrate (1.500 g L^{-1}), Dipotassium hydrogen phosphate (0.0314 g L^{-1}), Magnesium sulphate (0.036 g L^{-1}), Calcium chloride dihydrate (0.0367 g L^{-1}), Sodium carbonate (0.020 g L^{-1}), Disodium magnesium EDTA (0.001 g L^{-1}), Citric acid (0.0056 g L^{-1}), and Ferric ammonium citrate (0.006 g L^{-1}). Final pH is maintained as 7. Sodium chloride and Vitamin B12 in respective quantities was added since *Scenedesmus abundans* is a marine species [18].

The desalination chamber was filled with Nacl concentration of 35 g L^{-1} (MDC 1) and 20 g L^{-1} (MDC 2).

2.4. Experimental procedure

All the experiments were conducted in ambient temperature approximately equivalent to 30 °C ± 2 °C. Electrical conductivity, Nacl concentration, Total Dissolved solids (TDS) concentrations and pH of the saline solution were analyzed by water analysis kit (Eutech Instruments, Cyber scan series 600). Algae growth was monitored by measuring optical density at 680 nm by UV-3200 double beam spectrophotometer. The COD of the anolyte solution was measured by the standard methods [19]. The voltage over 1 KΩ resistor was measured continuously by the digital multimeter (VC 97). Current (I) and power (P) were determined according to $V = R_{ext} \times I$ and $P = V \times I$.

3. Results and discussion

3.1. Desalination performance in microalgae bio-cathode MDC

The desalination performances were evaluated based on electrical conductivity, Nacl removal and TDS removal. The chemical cathode (35 g NaCl L^{-1}) performance is compared with biocathode MDC 1 containing 35 g NaCl L^{-1} concentration and also the MDC performance of two different initial salt concentrations ie., MDC 1(35 g NaCl L^{-1}) and MDC 2 (20 g NaCl L^{-1}) with bio-cathode is analyzed.

The initial electrical conductivity and TDS concentration of chemical cathode and microalgae bio-cathode (MDC 1) were found to be 49.65 mS/cm and 27.01 g L^{-1}. After twenty two days of operation, the initial saline concentration of 35 g NaCl L^{-1} reduced to 23.56 g NaCl L^{-1} for the chemical cathode and 15.62 g NaCl L^{-1} for microalgae bio-cathode. The percentage removal efficiency was found to be 32.7% for Chemical cathode and 55.3% for microalgae bio-cathode. Though desalination occurs in chemical cathode, the efficiency was predicted to be low when compared with microalgae bio-cathode. The higher salt removal efficiency is obtained in bio-cathode due to the high electrical potential developed between cathode and anode that had driven a large

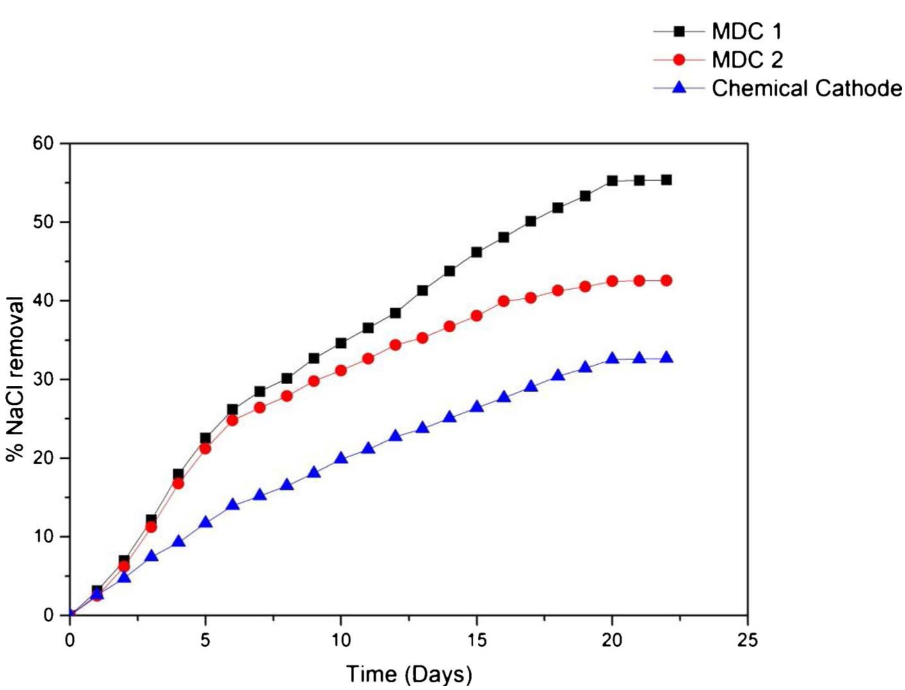

Fig. 3. NaCl removal percentage with chemical cathode and MDC with different initial salt concentration.

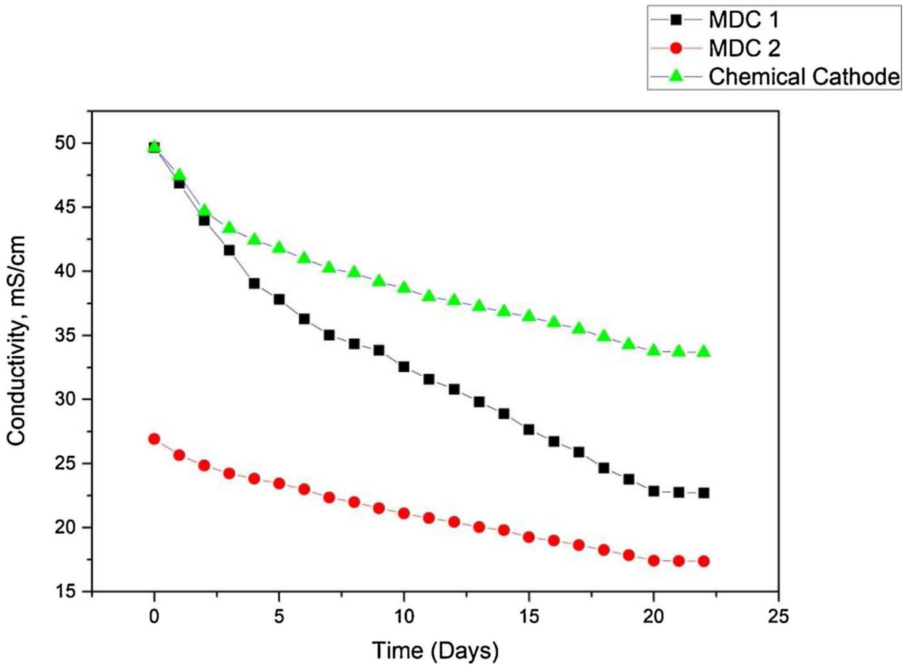

Fig. 4. Electrical conductivity variation with chemical cathode and MDC with different initial salt concentration.

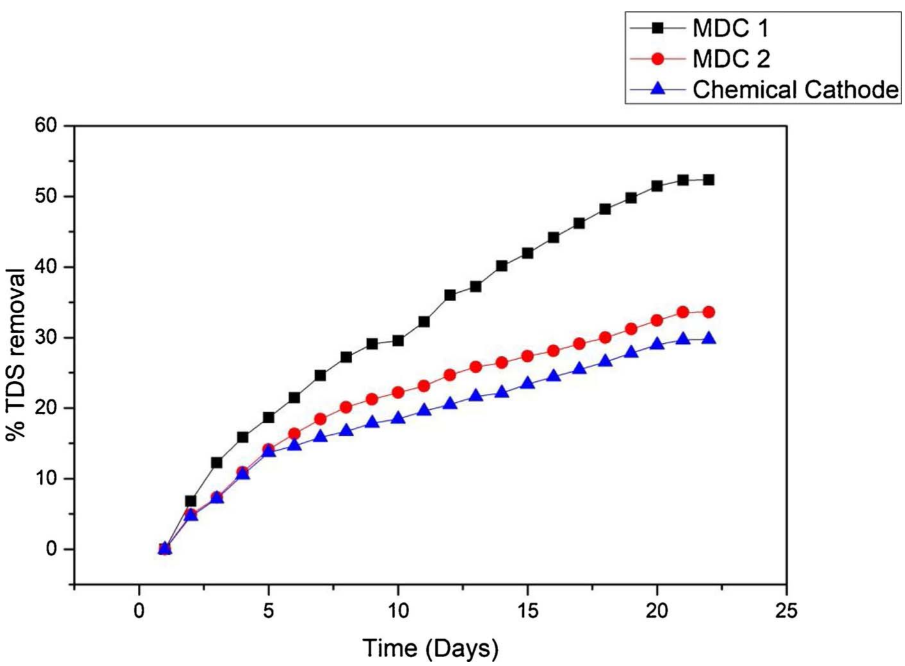

Fig. 5. TDS removal percentage with chemical cathode and MDC with different initial salt concentration.

quantity of migration of ions from middle chamber to anode and cathode chamber. The same scenario had been observed by Wen et al. [20] and Bahareh Kokabian et al. [8].

In case of MDC 2 (20 g NaCl L^{-1}) the electrical conductivity and TDS concentration were found to be 26.90 mS/cm and 14.87 g L^{-1} respectively. The salinity removal percentage was found to be 42.6% .The rate of desalination depends on the initial salt concentration of the sample to be desalinated. In the present work, high desalination rate is observed in MDC 1with higher salt concentration of 35 g NaCl L^{-1} compared with MDC of low salt concentration of 20 g Nacl L^{-1}. MDC is primarily appropriate for desalinating water with high salt content. This may lower the ohmic resistance and result in higher current production and a higher desalination rate (DR) [21]. Hence in our study, MDC with higher salt concentration was found to have higher removal efficiency. Our results matches with the ideology of Atieh Ebrahimi et al. [22] i.e., the accelerated desalination in higher initial salinity was due to higher current and power generation which drove desalination. Fig. 3 illustrates Nacl percentage removal efficiency of the chemical cathode, MDC 1 and MDC 2 respectively.

Figs. 4 and 5 elucidate the conductivity change and TDS removal efficiency of the chemical cathode, MDC 1 and MDC 2 respectively. Gradual decrease in electrical conductivity was seen in chemical cathode and MDC with different initial salt concentration. MDC 1 with higher salt concentration showed higher electrical conductivity variation when compared with chemical cathode and MDC 2. In case of TDS removal efficiency, MDC 1 containing higher salt concentration showed prominent results.

The electrical conductivity of anode and bio-cathode compartments was analyzed at the initial and at the day of replacement of anode. The initial and final ionic conductivity of anolyte for 20 g NaCl L^{-1} and 35 g

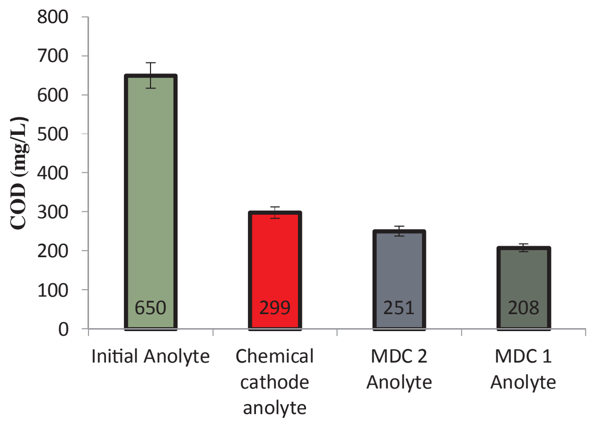

Fig. 6. COD removal in chemical cathode and MDC with different initial salt concentration.

NaCl L^{-1} were 1.291 mS/cm and 4.045 mS/cm; 1.291 mS and 6.542mS/cm at the end of first cycle. The gradual increase in conductivity of the anolyte solution during desalination may be due to the migration of chloride anions from the middle desalination chamber to the anode chamber. The initial and final ionic conductivity of catholyte for 20 g NaCl L^{-1} and 35 g NaCl L^{-1} were2.121 mS/cm and 5.542mS/cm; 2.121 mS and 7.425 mS/cm. The increase in ionic conductivity may be attributed to the migration of sodium ions from middle chamber to cathode chamber. Hence the above changes indicate that the transfer of ions occurred due to concentration difference across the membranes by the diffusion process.

MDC with higher salt concentration showed an increase in pH change when compared to MDC with low salt concentration. Anolyte pH decreased from 6.99 to 5.31 for MDC 1 and for MDC 2 the value decreased slightly i.e., from 6.74 to 6.08 over the fed batch period. The same pattern was followed in case of anolyte replacement till the completion of desalination process. Accumulation of protons and chloride ions forms hydrochloric acid that may result in lowering of anolyte pH. In contradiction, due to increase in hydroxyl and sodium ions (sodium hydroxide formation), the catholyte pH increased when compared with anode pH. In higher salt concentration the pH shifted from 7.98 to 9.78 in microalgae bio-cathode of 35 g NaCl L^{-1} whereas in 20 g NaCl L^{-1} the pH ranged from 8.06 to 9.06. The increase in pH of bio-cathode may alter or lower the algae growth. Hence the data predicted that the pH variation is solely dependent on initial salt concentration.

3.2. COD removal

The catalytic activity of bacteria, substrate utilization and formation of metabolites plays vital role in forming electrochemical gradient that establish the rate of desalination process. Chemical oxygen demand (COD) determines the substrate removal or utilization by the bacteria. Increase in breakdown of organic matter increases the COD removal efficiency which indicates increases in electron production and power generation. The initial COD of petroleum wastewater was found to be 650 mg/L. The COD removal percentages for Chemical cathode, MDC 1 and MDC 2 over a fed batch cycle were 54%, 61.3%, and 68%. Respectively and are shown in Fig. 6. From this study, it can be reported that COD removal efficiency of microalgae bio-cathode MDC is high when compared to the chemical cathode. Increase in activity of bio-film during degradation of organic matter produces large quantity of carbon dioxide that can be used by microalgae bio-cathode for efficient algae growth and oxygen generation [23]. Apparently, most of the MDCs in the study showed average COD removal. [22]

3.3. Power production performance

Figs. 7 and 8 show the voltage profile and power density profile for chemical cathode, MDC 1 and MDC 2. The MDC was operated one month prior to experimentation in order to obtain stable voltage. The voltage for chemical cathode increased slowly due to the lag phase of microorganisms and to establish bio-film formation in the electrodes [24,25]. Shorter lag phase is seen in the microalgae bio-cathode due to oxygen generation by algae that increased the electron mobility. The oxygen generation through photosynthesis can be given as

$$\text{Carbon dioxide} + \text{Water} \xrightarrow{\text{Algae, light}} \text{Sugar} + \text{Oxygen}$$

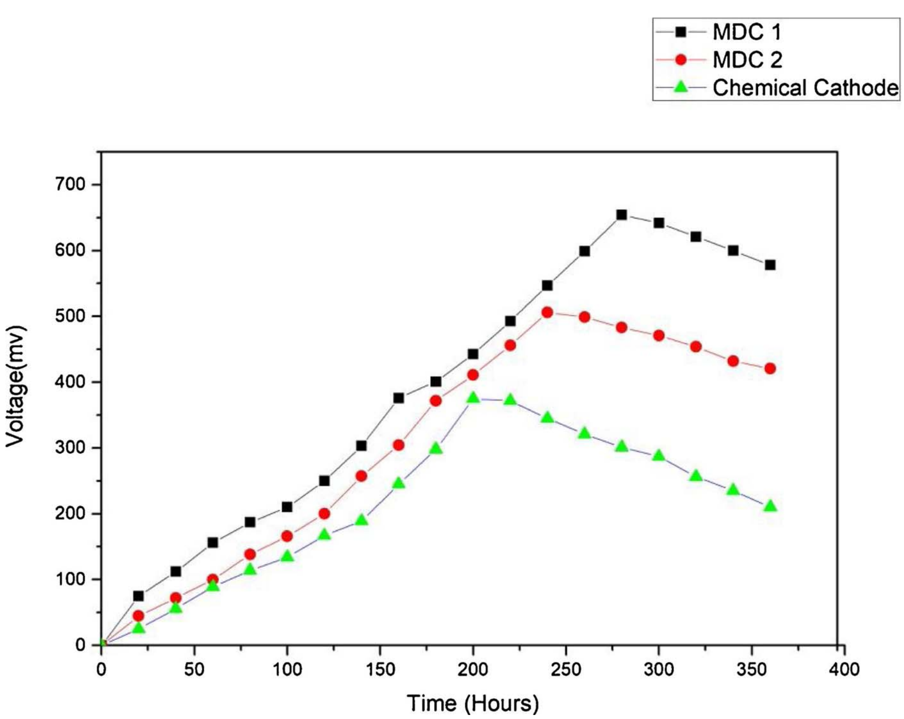

Fig. 7. Voltage profile for chemical cathode and MDC with different initial salt concentration over a batch cycle.

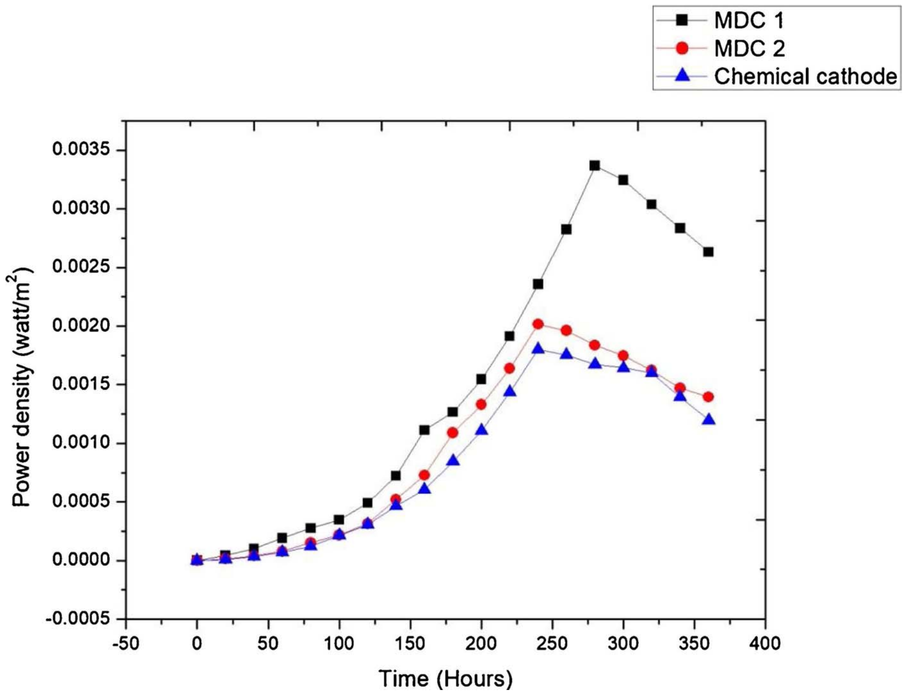

Fig. 8. Power density profile for chemical cathode and MDC with different initial salt concentration over a batch cycle.

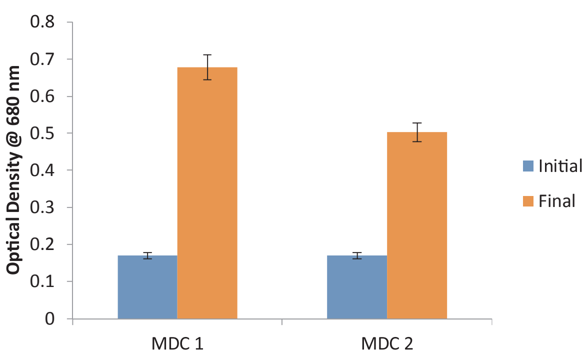

Fig. 9. Optical densities of biomass in MDC 1 and MDC 2.

The maximum voltage produced in a closed circuit for MDC 1 is 654 mv which is higher when compared with MDC 2 (506 mV) and chemical cathode (375 mV) across 1 KΩ resistor. The power produced for the respective voltages includes 427.7 mwatt (MDC 1), 256 mwatt (MDC 2) and 140.6 mWatt (Chemical cathode). Further, power density is calculated from power generated by surface area of the electrode. The power densities of chemical cathode, MDC 1, MDC 2 are 1.79 mWatt/m^2; 3.368 mWatt/m^2 and 2.10 mWatt/m^2. The high voltage produced by microalgae bio-cathode in this study may be due to the reduction of electrons and protons that occurs in aerobic catalyzed microbes [26,27]. Steady output voltages can be obtained from microbial catalyzed bio-cathodes when compared with other catholytes such as ferricyanide. When comparing with the different initial salt concentrations, conductivity plays a major role in voltage generation. Since, conductivity reduction in the middle chamber resulted in an increase in internal resistance that caused reduction in voltage production. MDC with salt concentration of 35 g NaCl L^{-1} produced high voltage than MDC with salt concentration of 20 g NaCl L^{-1}. The above observation was noted by Atieh Ebrahimi et al. [22] wherein he stated that internal resistance loss was the important factor that affects power generation in MDCs.

3.4. Biomass concentration

Microalgae growth rate played an important role in electrochemical desalination process. High power production was observed in MDC due to the migration of ions to the cathode that affected the algae growth positively. Algae require more electrons for its metabolism and new cells synthesis as shown in our study. The considerable amount of biomass is obtained at the end of 22th day. The variation in biomass identified through optical density observation at 680 nm. The optical density for MDC 1increased from 0.17 to 0.678 and for MDC 2 increased from 0.17 to 0.502 as shown in Fig. 9.

4. Conclusion

The study establishes MDC using microalgae as bio-cathode to be effective when compared with chemical cathode in simultaneous desalination, wastewater treatment and power generation performance. In this research it is discovered that petroleum wastewater in the anode compartment and *Scenedesmus abundans* algae in the cathode compartment can efficiently transfer electrons and ions across the membranes and desalinate water with small amount of bioelectricity production. It can be demonstrated that algae can serve as an insitu oxygen generator. The higher salt removal rate was achieved in MDC 1 with the salt concentration of 35 g NaCl L^{-1} i.e., 55.3% when compared with MDC 2 containing 20 g NaCl L^{-1} salt concentration is 42.6%. Thus the effect of high conductivity in the desalination chamber determines the system efficiency. The bioelectricity generation of 654 mv in MDC 1 also proved that high salt concentration with microalgae bio-cathode works effectively. The system produced algal biomass considered to be valuable feedstock and also be used for biodiesel production. The study enables a step towards green, renewable and environmental friendly photosynthetic microbial desalination cell.

Further development in reactor design, configuration of electrode material, ideas to decrease the internal resistance can dramatically influence the power production. With the help of recent trends in molecular biology and bioinformatics techniques, better bacterial and algae species can be identified, modified and utilized in increasing the overall performance of the Photosynthetic Microbial desalination cell.

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