Microbial Desalination Cell Performance Indicators and Limiting Factors

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Microbial desalination cells (MDCs) are promising bioelectrochemical systems for desalination using the bacteria-generated electricity from the biodegradation of organic wastes contained in the wastewater. Instead of being a sustainable and eco-friendly desalination technology, the large-scale application of MDC was limited due to the high installation cost of the metal-catalyst-coated cathode electrode and the poor performance of the cathode in long-term operation due to catalyst fouling.

Keywords: microbial desalination cell ; air-cathode ; cathodic limitations

1. Introduction

Globally, the rise of freshwater scarcity issues has become more evident due to the expanding population growth, industrialization, and climate change [1][2]. To minimize freshwater demands, different nations installed their first desalination initiatives in the late 1950s [3]. Nowadays, the number of worldwide desalination plants reached almost 16,000, where 68.7% are reverse osmosis (RO) membrane-based technology plants [3]. The conventional reverse osmosis (RO) method for desalinating saline water consumes excessive electrical and mechanical energy [4]. For instance, the energy used to purify 1 m^3 of seawater is approximately 0.36–0.47 kWh [5]. Seawater desalination also uses thermal technologies, such as multieffect desalination (MED) and multistage flash (MSF). However, they have several disadvantages from a climate change perspective [6].

Recently, numerous studies have focused on developing desalination processes powered by renewable energies, such as wind, solar, geothermal, and bioenergy [7]. However, renewable energy-powered desalination processes are more expensive than conventional processes [8]. Therefore, developing a sustainable, inexpensive, and efficient desalination technology was necessary. Microbial desalination cells (MDCs) are a promising bioelectrochemical technology for desalinating seawater, requiring little or no energy (**Figure 1**) [9][10]. The sustainability of MDC is attributed to their use of wastewater organics as the driving force to desalinate saline or brackish water via biological wastewater treatment and power generation [11]. Thus, integrating wastewater treatment with desalination can maximize the environmental benefits as the energy contents are extracted from the wastewater before being released into nature [12].

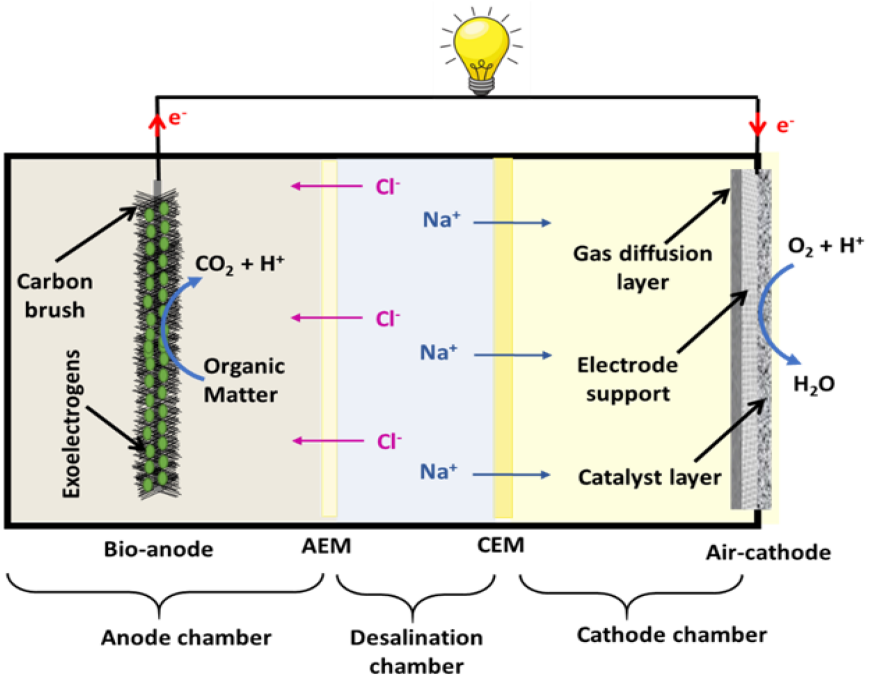

Figure 1. Schematic diagram of typical MDC.

Many publications have been published in the last decade on various MDC reactor configurations [13][14][15]. Investigated reactors include biocathode MDC [16][17][18], air cathode MDC [8][19][20], etc. The three-chamber microbial desalination cell has been the focus of most researchers. The conventional three-chamber MDC reactors consist of anodic and cathodic chambers with a central desalination chamber separated by ion-exchange membranes [19].

The desalination mechanism in MDCs is a spontaneous process that needs no external power [20][21]. It is only dependent on the electricity produced by exoelectrogenic bacteria [22]. The organic-rich wastewater is utilized as a metabolic substrate for the exo-electrogens (e.g., *Shewanella*, *Geobacter*, etc.) that grow on the carbonaceous anode in the anode compartment [23]. The attached anodic bacteria oxidize the organic contents of wastewater by the metabolic processes that produce electrons that move to the anode by the bacterial nanowire piles, cytochrome C, etc. [9][24][25]. Then, the biogenerated electrons are sent to the cathode through an external circuit and load, where they are reduced by electron-acceptors/oxidizing-agents (e.g., O_2) present in the catholyte. Hence, a potential gradient between the electrodes is created by the electrical current flow from the bioanode to the cathode for the cathodic reaction. As a result, the ions (anions and cations) in the central compartment (desalination chamber) are forced to flow through the membranes to the surrounding chambers by the current potential. The saltwater is desalinated as a result of this phenomenon. The charge balance is maintained by ions migrating across ion exchange membranes. Through a cation exchange membrane (CEM), cations (Na^+) flow towards the cathode, whereas anions (Cl^-) migrate towards the anode, passing through the anion exchange membrane (AEM). Generally, Equations (1) and (2) represent the redox reactions in MDC [20].

$$\text{At the anode: Substrate} + nH_2O \rightarrow nCO_2 + 4ne^- + 4nH^+ \quad (1)$$

$$\text{At the cathode: } O_2 + 4ne^- + 4nH^+ \rightarrow 2H_2O \quad (2)$$

For a decade, microbial desalination cell scale-up initiatives focused on desalination limitations, such as limited power output and high internal resistance caused by the MDC components [26]. Several factors, including reactor configuration, electrode materials, electrolyte conductivity, ion exchange membrane fouling, biofilm inhibition, and operational circumstances, have contributed to these limitations [14]. Electrode materials, as an illustration, affect both the MDC system performance and cost efficiency due to internal losses (e.g., electrode overpotential) [27]. High-performance electrode materials with stable structures and a large surface area must be employed in MDC to increase the power-output performance at low internal resistance [24].

Furthermore, MDC performance is influenced significantly by anodic and cathodic reactions. However, cathodic reduction is a limiting factor for stable and effective MDC performance [25]. Generally, cathodes in microbial desalination cells can be divided into cathodes submerged in liquid electrolytes and air cathodes exposed to open air [28]. Reagents are consumed as electron acceptors in the liquid-cathode MDC, which must be treated before reuse or disposal. On the other hand, air-cathode MDC uses oxygen as an electron acceptor. Thus, it can be seen as an ecofriendly and commercially scalable alternative compared to liquid-cathode MDC due to the abundance of O_2 [13]. Hence, most MDC research utilizes atmospheric oxygen as an oxidant species [29][30]. Although, limited cathodic performance was recorded due to the high

overpotential of oxygen reduction reaction that reduces cathode reaction kinetics and the system's efficiency [31][32]. Accordingly, effective electrocatalysis is required to boost the oxygen reduction reaction (ORR) performance [13].

2. MDC Performance Indicators and Limiting Factors

The performance and efficiency of the MDCs system are indicated and measured by considering the various parameters listed in **Table 1**. However, any technology's overall performance and efficiency are determined and controlled by the magnitude and conditions of specific factors before, during, and after the operation [33]. The MDC system's efficiency and output are affected significantly by the configuration of the reactor constructed, including the material, dimensions, and electrode materials used. It is also influenced by the membrane (IEMS), substrate (electrolytes), and operational conditions, i.e., temperature, hydraulic retention time (HRT), and pH imbalance, as shown in **Figure 2**.

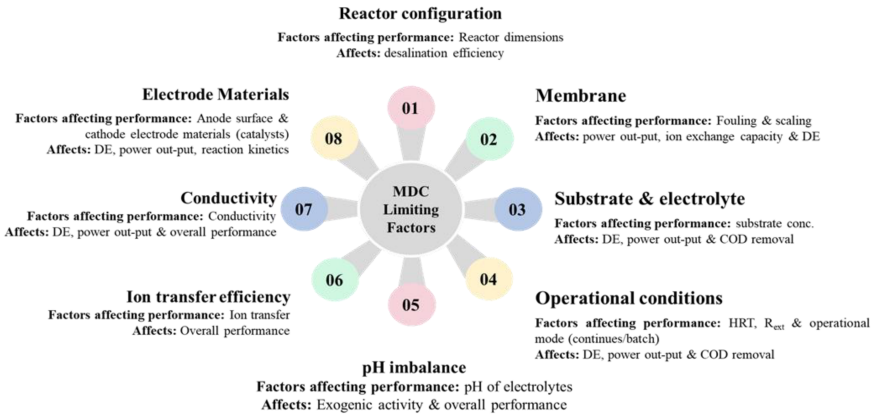

Figure 2. Limiting factors of MDC performance.

Table 1. MDC performance and efficiency indications.

Performance Indicator	Remarks	Mathematical Expression	References
Desalination Efficiency (DE)	**Reduction percentage in saltwater's conductivity.**	DE=(Salt conc. i−Salt conc. f)(Salt conc. i)	[34]
Desalination Rate (TDR)	**Amount of salt removed per unit of time.**	DR=Total salt removedTotal desalination time	[35]
COD Removal	**The amount of organic matter removed via microbial metabolism.**	CODRE=(Conc.i−Conc.f)(Conc.i)	[36]
Current Efficiency	**The ratio of produced current to the chamber's working volume or the number of ions separated per electron transferred at both electrodes.**	ηi=FZVΔcNcp∫idt	[37]
Power Density	**Power generation in a cell-based on the projected surface area of electrodes or electrolytes volume.**	PD= Power produced Electrolyt's volume	[38]
Coulombic Efficiency	**The ratio of actual charge produced to the available charge is theoretically calculated based on the reduction in COD.**	CE=(MO2∫idt)(neFVanΔCOD)	[39][40]
Ion-exchange Efficiency	**The exchange efficiency of IEMs to allow the number of produced ions by anodic and cathodic reaction in MDC**	-	[41]

Precisely, the reactor dimension, such as the volumetric ratio utilized in constructing and processing the MDC system, has shown a notable impact on its performance. Jingyu et al. (2017) revealed that a lower volume of anolyte and catholyte chambers accompanied lower reactor efficiency [33]. Cao et al. (2009) and Meng et al. (2014) used a volumetric ratio (Vanode:Vcathode:Vdesalination) of (3:3:1) and (100:33:1) for the desalination of a 5 g/L synthetic seawater (NaCl) [41][42]. Meng et al. (2014) achieved a high desalination efficiency (DE) of 90% and a power output of 31 W/m^3 compared to Cao et al. (2009), who recorded a DE of 46.37% [41][42]. In addition, constructing an MDC with a shallower-depth desalination compartment and reducing the gap between electrodes could be beneficial in overcoming the high internal resistance and optimizing the electromotive potential and power generation.

The membranes employed in MDC reactors can segregate ionic species in a solution using electrical current, resulting in a bipolar process. During reactor operation, membranes can cause the dissociation of water, which significantly reduces power output due to the loss of electrons for the dissociation process. The findings of various studies revealed that different types of membranes with high surface areas and great ion-exchange capacity could improve the removal of the salts, enhancing the desalinization efficiency by 50–63% [8][41]. However, fouling and scaling are issues that appear due to IEMs used in bioelectrochemical systems (BESs) for long-term operations, imposing extra resistance to salt removal [43].

Moreover, electrolytes have a remarkable impact on BES performance as they are considered a reservoir for ion species, a source of organic matter, and a medium for pH fluctuations [44][45][46]. Therefore, high salinity removal demands a large volume of electrolytes, especially anolytes [47]. In addition, the properties of substrates used to feed biofilms in an anode chamber influence the MDC efficiency. As an illustration, Kuichang et al. (2016) improved the performance of M-MDC by increasing the concentration of anolytes through adding more glucose, resulting in a maximum performance of high DE (47.3%), COD (40.2%), and a current output of 0.6 mA [48].

Further, the modes of operating MDC (batch or continuous) can significantly influence MDC stability and performance. For instance, in a batch mode, the conductivity of the electrolytes decreases with time, resulting in high internal resistance and a reduction in the overall performance [23]. In continuous mode, the reactor chambers are fed with a solution circulating continuously in cycles under specific HRT. Consider the comparative performance of UMDCs operated in batch mode and continuous mode. High performance with a maximum power density of 38 W/m^3, current production (62 mA), salt removal (>99%), total dissolved-solids removal rate (7.50 g $TDS \cdot L^{-1} \cdot d^{-1}$), and efficiency of charge (98.6%) was achieved by operating UMDC under continuous mode [8]. On the contrary, Jafary et al. (2020) MDC reactor operated under batch mode recorded energy production of 8 mW, current generation (43 mA), and a desalination rate of 24.3 mg/h [11]. However, one promising future recommendation for METs is to be operated by a recirculation batch mode. Jafary et al. (2017) revealed that MFC performed in recirculation in batch mode gave a high performance of maximum power density of 38 W/m^3 compared to batch and continuous modes, respectively [49].

Aside from modes of operation, hydraulic retention time plays an essential role in overall desalination efficiency. To illustrate, Jacobson et al. (2011) investigated the UMDC under HRT on days one and four using the continuous mode of operation. The results showed a maximum boost in their system's performance by increasing the HRT from 1 to 4 days. It was evident that the HRT of salt solution significantly influences the TDS since the longer hydraulic retention time allows more involvement of salt in the current generation, and thus it being removed [8].

Equally important, the external resistance (R_{ext}) is another factor that may majorly affect the MDC efficiency outputs. Numerous publications reveal that MDC performance (current production) increases by decreasing the R_{ext} [45][50][51]. A constructed upflow-stacked microbial desalination cell (USMDC) reactor operated under applied external resistance (R_{ext}, 1000 Ω–1 Ω) recorded a high desalination ratio of 91.9% when R_{ext} was 1.5 Ω [51]. The USMDC's current production increased as the R_{ext} was reduced from 500 Ω. Nevertheless, Wang et al. (2020) reported reductions in the current generation when R_{ext} continued to decrease, proving the fact that there is an optimal external resistance in which the current production can be maximized (1 Ω–5 Ω) [51]. In previous studies, the optimum external resistance values of the MDC reactors were approximately 10 Ω [52].

Furthermore, the released protons react with anions transported from the desalination chamber, producing internal biofilm acidification due to the metabolic process. As the desalination process operates over time, protons accumulate due to microbial respiration, decreasing pH in the anode compartment. Hydroxides accumulate in the cathode compartment because of oxygen reactions and rising pH values. The pH of an anode impacted the anode's potential of many METs and was the main cause of desalination efficiency reduction in MDC. Accordingly, controlling pH fluctuation has become a critical factor in optimizing the performance of BES, including MDC, especially in terms of power production and water desalination. Luo et al. (2012) introduced anolyte's continuous recirculation between a feed container and an anode compartment. This operational mode helped mitigate the anode chamber's pH reduction and raised the current output by 61% compared to results obtained under batch mode [53]. Additionally, Jafary et al. (2020) established a proof-of-concept study for a two-chamber UMDC, using a new arrangement of anion-exchange membrane and cation exchange membrane that resulted in a self-generated pH control approach [11].

To drive a desalination process in an MDC reactor, several electrons are needed to remove an equal quantity of salt from saline water. However, this number of electrons could be lost while running other reactions instead of salt removal, affecting the overall system's efficiency. The loss of electrons is caused by back diffusion and membrane resistance [33]. Further, required electrons for saltwater desalination may be influenced by other electron acceptors, such as O_2 diffusion into the anode compartment [54]. Under those circumstances, more organic oxidation reactions are needed to drive the

desalination process [8]. The split of large pairs of ions due to electron transport through the systems enhances the charge transfer efficiency, improving desalination [55]. More studies on optimizing charge transfer efficiency are needed.

The concentration of ionic content in an electrolyte, which can transmit electrical charges, is referred to as conductivity, which remarkably affects MDC performance. High conductivity values resulting from high saltwater concentrations create a higher concentration gradient between chambers. If the concentration of electrolytes is lower than that of the saline water, the MDC's desalination performance is enhanced by dialysis. In addition, desalination performance is influenced by some ionic species and impurities that prohibit the overall performance. Ion transport efficiency is affected by salt solution concentration and composition. For instance, seawater contains various ions and impurities, such as Ca_2^+, Mg_2^+, K^+, Br^-, SO_4^{-2}, clay, and silica, which decrease the conductivity and result in a higher internal resistance during MDC operation [56].

Finally, the anode surface contributes highly to the activation losses, despite the amount of gained energy by biofilm metabolism activities [57][58]. Therefore, electrodes should be created from materials with a larger surface area, so that a larger mass of biofilm can adhere to the anodic oxidation process. The adhesion of large biomass onto anode electrodes can consequently enhance the current generation [23]. Accordingly, most studies used an extensive surface area of the material. For example, carbon-based products, such as carbon felt, fiber brushes, cloths, graphite granules, graphite plates, parous graphite, activated carbon (AC), and 3D carbon nanotube (CNT) matrices [14]. However, each type of these electrodes exhibits different performances and output efficiencies because of the variations in their effective surface areas, structures, operational conditions, and metal-catalyst coatings. As an illustration, the large surface area of AC results in high electron transfer in anode compartments, as it enables microbial growth, which leads to more significant bacterial cultures [59]. In addition, AC electrodes utilized in MDC achieved a complete NH_3-N removal (99%), high removal of COD (96.9%), and removal of total PO_4 (98.3%) [60]. In comparison, applying rough surface graphite (RSG) catalysts to MDC electrodes led to a remarkablely high power output of 10.8 W/m^3 [61].

Cathode electrode materials also play a crucial role in retrieving electrons and in their consequent use in the reduction reaction [62]. In cathodes, the suitable terminal electron acceptors (TEA) are reduced by electrons coming from the anode, completing the cathodic half-cell reaction [63]. Further, the performance of METs significantly depends on the cathode's electron harvesting efficiency. For instance, MDC was investigated with several cathodic electron acceptors, such as oxygen, hypochlorite, permanganate, and dichromate [64]. However, cathode effectiveness is determined by TEA's reduction reaction kinetics, which is one of the critical things in microbial electrochemical technologies (MET) [65][66].

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