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Review

Pushing microbial desalination cells towards field application: Prevailing challenges, potential mitigation strategies, and future prospects

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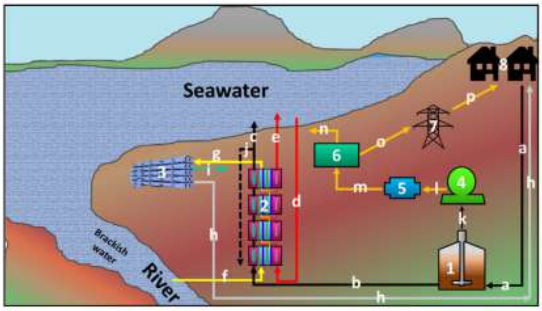
HIGHLIGHTS

- Recent standalone, assembled, coupled, and scaled-up MDCs are reviewed and discussed.
- The performance of assembled, coupled, and scaled-up MDCs are comparatively analysed.
- Prevailing challenges and potential solutions for scaling-up MDCs are discussed.
- Integration of MDCs with conventional technologies as prospects are proposed.
- Recommendations for future MDCs scaling-up studies are provided.

GRAPHICAL ABSTRACT



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ABSTRACT

Microbial desalination cells (MDCs) have been experimentally proven as a versatile bioelectrochemical system (BES). They have the potential to alleviate environmental pollution, reduce water scarcity and save energy and operational costs. However, MDCs alone are inadequate to realise a complete wastewater and desalination treatment at a high-efficiency performance. The assembly of identical MDC units that hydraulically and electrically connected can improve the performance better than standalone MDCs. In the same manner, the coupling of MDCs with other BES or conventional water reclamation technology has also exhibits a promising performance. However, the scaling-up effort has been slowly progressing, leading to a lack of knowledge for guiding MDC technology into practicality. Many challenges remain unsolved and should be mitigated before MDCs can be fully implemented in real applications. Here, we aim to provide a comprehensive chronological-based review that covers technological limitations and mitigation strategies, which have been developed for standalone MDCs. We extend our discussion on how assembled, coupled and scaled-up MDCs have improved in comparison with standalone and lab-scale MDC systems. This review also outlines the prevailing challenges and potential mitigation strategies for scaling-up based on large-scale specifications and evaluates the prospects of selected MDC systems to be integrated with conventional anaerobic digestion (AD) and reverse osmosis (RO). This review offers several recommendations to promote up-scaling studies guided by the pilot scale BES and existing water reclamation technologies.

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Contents

1. Introduction

Approximately 30% of the people living around the world are suffering from the lack of clean water, while 60%–70% of the world's population may experience severe water shortage due to increasing population, natural resource reduction, global warming and environmental pollution (Macedonio et al., 2012). Although the readily available technology of water reclamation through conventional wastewater treatment and desalination processes has been established, it requires a considerably high operational cost. A study has estimated that the aerobic activated sludge processes used in domestic wastewater treatment plants require ~0.6 kWh m^{-3} of wastewater (McCarty et al., 2011). Conventional high-pressure-membrane desalination and thermal-desalination technologies consume 3.7–650 kWh energy per m^3 of water desalination (Mehanna et al., 2010a). Wastewater contains a high quantity of organic carbon sources, which are composed of approximately 17.8–28.7 kJ g^{-1} of chemical oxygen demand (COD) in the form of stored energy (Heidrich et al., 2011). Attractively, an abundant source of energy from high-strength wastewater can be harvested through anaerobic digestion with a positive net energy balance (McCarty et al., 2011).

For the past decades, bioelectrochemical systems (BESs), such as microbial fuel cells (MFCs), microbial electrosynthesis cells (MECs) and MDCs, have proven their ability to capture energy from wastewater by using electrochemically active bacteria (EAB) (Pant et al., 2011; Wang et al., 2015; Yuan and He, 2015). However, at the present stage, the operational cost estimated for MFC (i.e. \$0.05 kg^{-1} COD) is slightly higher than that for conventional anaerobic digestion, which costs at \$0.048 kg^{-1} COD, whereas MEC is significantly higher than AD by \$0.11 kg^{-1} COD (Sleutels et al., 2012). In our perspective, the energy requirement for solution recirculation used in MFCs may be approximately equivalent to the energy required for stirring used in a conventional anaerobic digester. Moreover, an MEC system requires power input for electrolysis or synthesis to occur, thus adding up operational costs. In terms of capital density, MDCs can reach approximately \$6773, which is slightly more expensive than that of MFC and MEC and contributed by the additional membrane of AEM (Zhang and Angelidaki, 2016). Despite high cost and energy consumption issues, BESs offer a potential in energy recovery in various forms, including power generation, hydrogen evolution, hydrogen peroxide, methanol and ethanol (Wang et al., 2015; Yuan and He, 2015). BESs are also capable of minimising the sludge production to about 65%–71% lower than that of activated sludge treatment (Zhang and Angelidaki, 2014); consequently, the budget for sludge disposal is reduced. Through energy optimisation, a positive net energy production can potentially offset the capital cost invested for the system.

MDCs show a great potential for applications that generate electricity or fuel (e.g. hydrogen) from a low-energy wastewater treatment with a concurrent novel desalination or nutrient recovery process depending on feed stream sources (Li et al., 2014). The pioneering design of MDCs was in the form of plate stacking with an installed ion exchange membrane (IEM) to allow the removal of salts from saline water (e.g. seawater and brackish water) (Cao et al., 2009; Mehanna et al., 2010a, b). Since then, various MDC designs and configurations have been investigated to accommodate different functions (Fig. 1). For example, a value-added chemical of acids and bases can be synthesised concurrently with desalination process by adding a bipolar membrane next to an anode chamber in a typical MDC design (Chen et al., 2012b; Liu et al., 2015). Salinity-gradient power can be generated beside the electron generation from bioelectrochemical oxidation in an anode chamber by combining a reverse electrodialysis cell in between anodes and cathodes of a typical MFC system; as a result, the energy input for

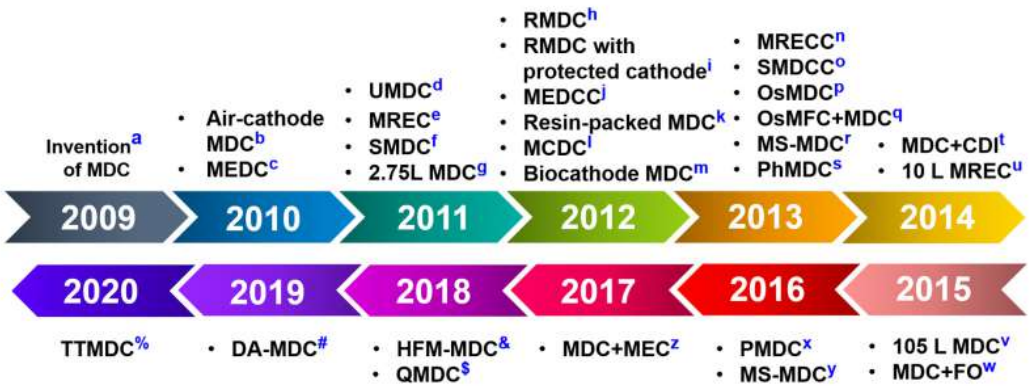


Fig. 1. The timeline of the MDCs design development: [a](Cao et al., 2009), [b](Mehanna et al., 2010b), [c](Luo et al., 2010; Mehanna et al., 2010a), [d](Jacobson et al., 2011a), [e](Kim and Logan, 2011b,c), [f](Chen et al., 2011), [g](Jacobson et al., 2011b), [h](Qu et al., 2012), [i](Chen et al., 2012c), [j](Chen et al., 2012a), [k](Zhang et al., 2012), [l](Yuan et al., 2012), [m](Wen et al., 2012), [n](Luo et al., 2013), [o](Zhang and Angelidaki, 2013), [p](Werner et al., 2013), [q](Zhang and He, 2013), [r](Qu et al., 2013), [s](Kokabian and Gude, 2013), [t](Qu et al., 2013), [u](Zuo et al., 2014), [v](Zhang and He, 2015), [w](Yuan et al., 2015), [x](Liang et al., 2016; Yuan and He, 2015), [y](Zuo et al., 2016b), [z](Li et al., 2017b), [&](Zuo et al., 2018), [$](Ebrahimi et al., 2018), [#](Liu et al., 2019c). [%](Jafary et al., 2020). Please refer to the list of abbreviations provided in this review paper.

pumping is offset (Kim and Logan, 2011b). Typical MDC designs have also been reconfigured by placing IEM in contact with bulk saline groundwater in submersible MDC (SubMDC) to conduct in situ nitrate removal (Zhang and Angelidaki, 2013).

Although MDCs have been proven as multifunctional tools, they are limited by several drawbacks: 1) pH imbalance between anode and cathode chambers (Luo et al., 2010); 2) high internal resistance; 3) different solutions required for anode, desalination and cathode chambers, thereby complicating the operation (Zuo et al., 2016a); 4) desalination rate reduction due to diluted saline solution (Mehanna et al., 2010b); 5) low desalination rate (Chen et al., 2012b); 6) poor effluent quality of <85% (Davis et al., 2013; Luo et al., 2012c; Ping et al., 2015; Wen et al., 2012); 7) water flux into the middle chamber, thus decreasing the conductivity of saline water (Jacobson et al., 2011b); and 8) ineffective desalination of high-concentration saline solution in a single MDC (Lu et al., 2015).

Several studies have explored the connection of identical MDC units forming an assembled MDC system (Kim and Logan, 2011c; Qu et al., 2013; Zuo et al., 2016a) or the combination of MDCs with other BES/existing water reclamation technologies creating a coupled MDC system (Dong et al., 2017; Li et al., 2017b; Wen et al., 2014; Yuan et al., 2015; Zhang and He, 2013). Although a lab-scale prototype of MDCs has been consistently improved, the pace of scaling-up efforts is rather slow, consequently impeding the materialization of these applications. To date, only tubular and stacked MDC designs have been scaled-up into several litre scales (>2.75 L to >100 L) (Jacobson et al., 2011b; Zhang and He, 2015; Zuo et al., 2014).

Although many review articles related to MDCs have been provided (Al-Mamun et al., 2018; Saeed et al., 2015; Sevda et al., 2015; Yang et al., 2019; Zhang et al., 2019), current literature surveys show that few cell configurations addressing the unresolved issues in MDC systems have not been reviewed. These issues include the utilisation of a large amount of anolytes relative to saline water in CMDCs, which have been tackled by the invention of a quadripartite MDC (QMDC) (Ebrahimi et al., 2018). Furthermore, issues associated with the concentration polarisation of saline solutions and long hydraulic retention time (HRT) in hollow-fibre-membrane MDC (HFM-MDC) have been addressed by the development of a dual-anode MDC (DA-MDC) (Liu et al., 2019c; Zhang and He, 2012b). Moreover, a two-chamber tubular MDC is workable to alleviate the pH imbalance between anodes and cathodes in a more practical way than in conventional designs (Jafary et al., 2020). A detailed discussion on these challenges and potential mitigation strategies are provided in the next section.

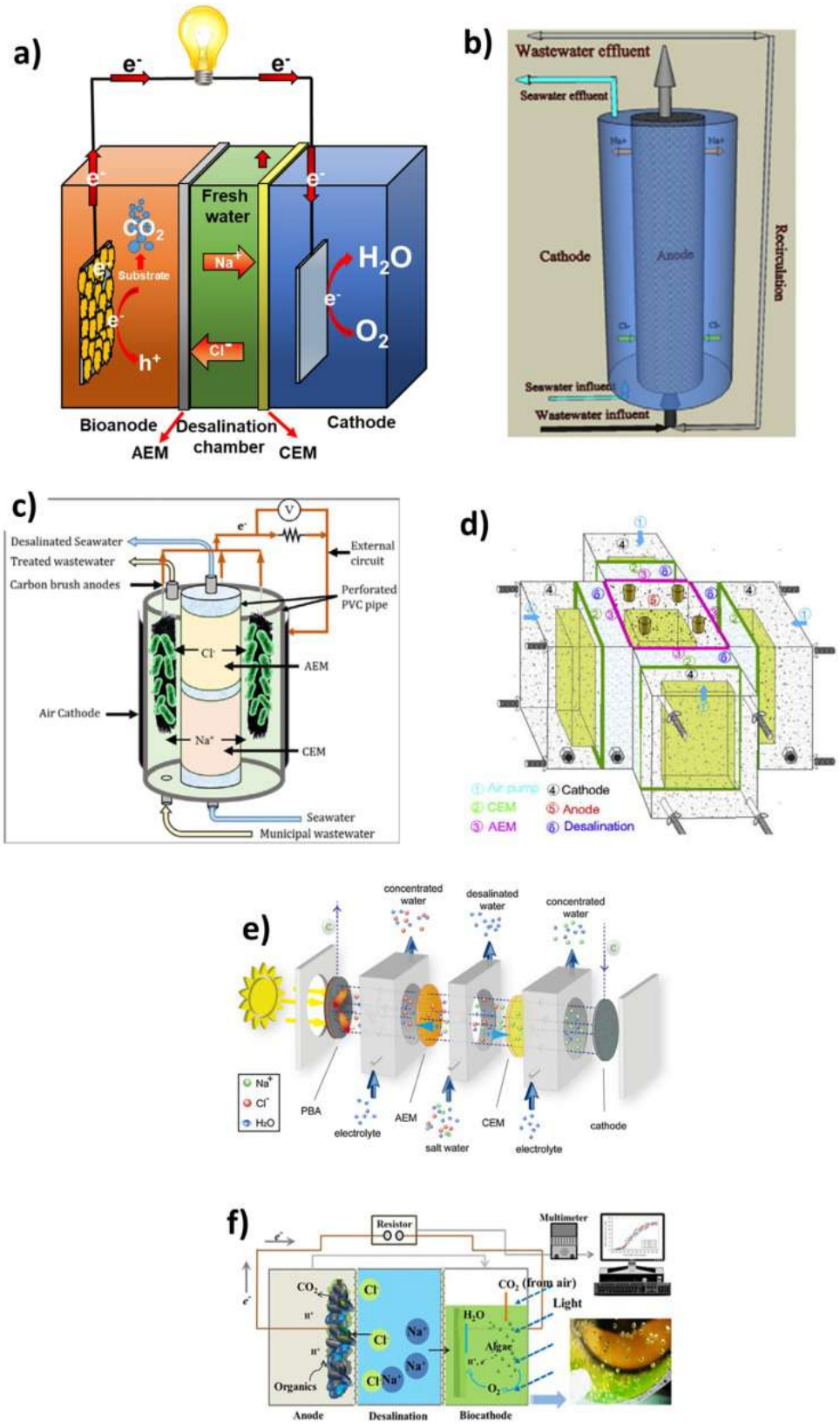
Regarding the importance of scaling-up studies on MDCs, the recommendation and guidance for scaling-up MDCs have not been discussed in depth. Therefore, in the present review, recommendations for large-scale studies on MDCs are proposed on the basis of lab-scale advancements with guidance from other large or pilot-scale BESs and existing desalination technologies. The development of different cell configurations to accommodate different novel applications is initially discussed by chronologically outlining their pros, cons and mitigation strategies. Then, the progress of assembled, coupled and scaled-up MDC systems is comprehensively described. Single MDCs, assembled MDCs, coupled MDC systems, lab-scale systems and large-scale systems are subjected to a comparative analysis. Furthermore, the prevailing challenges related to scale-up and potential mitigation strategies are presented. Several MDCs are selected as the prospects for the potential integration with the existing technology of anaerobic digestion (AD) and reverse osmosis (RO). These prospects are preliminarily assessed on the basis of the performance reported in the literature. Finally, concluding remarks and recommendations for future studies are provided.

2. MDCs, configurations and applications

MDCs have been remarkably improved to accommodate novel multipurpose reactions related to environmental, water source and energy issues. The emergence of different MDC designs in the literature is classified based on the types, orientations and shapes of membranes and membrane-based processing. With the installation of ion exchange membranes (IEMs), namely, anion exchange membranes (AEMs) and cationic exchange membranes (CEMs), a three-chambered cell of a conventional MDC can be produced (Al-Mamun et al., 2018; Jacobson et al., 2011b; Jafary et al., 2018; Luo et al., 2010), a forward osmosis (FO) membrane and a CEM can be used to design an osmotic MDCs (OsMDCs) (Ismail and Ibrahim, 2015; Zhang and He, 2012b), and a bipolar membrane (BPM) combined with an IEM can be applied to develop microbial electrodialysis and chemical production cells (MEDCCs) (Chen et al., 2012a; Luo et al., 2017; Ye et al., 2017). The orientation of IEMs in direct contact with a bulk waste/wastewater source has been applied to develop a submersible MDC (Zhang and Angelidaki, 2013, 2015b). Membrane-based processing such as reverse electrodialysis (RED) has been utilised to construct a microbial reverse-electrodialysis cell (MREC) (Cusick et al., 2013; Kim and Logan, 2011a, b). Membrane capacitive deionisation (CDI) has been employed in an MDC that functions as microbial capacitive desalination cells (MCDCs) (Feng et al., 2013; Won et al., 2014; Yuan et al., 2012). Under the membrane shape category, a hollow fibre membrane (HFM) has been used to create a recently developed hollow fibre membrane (HFM-MDC) (Liu et al., 2019a,c; Zuo et al., 2018).

Table 1
Summarised advantages, challenges and solutions of standalone MDC systems.

System	Advantages	Challenges	Solution
MDCs	• Simultaneously treat wastewater, desalination and electricity generation • Negligible toxic effects of oxygen. • Cost-effectiveness • Salinity removal of ~70% (Jacobson et al., 2011b) • Reduction of total dissolved solids (TDS) of ~43% (Jacobson et al., 2011b)	• pH imbalance between anode and cathode chambers (Luo et al., 2010) • Acidic environment inhibiting microbial growth (Al-Mamun et al., 2018) • Possible bacterial growth on the cathode surface when RMDC is employed (Qu et al., 2012) • Low desalination rate • Increased ohmic resistance • Reduction of conductivity because of the dilution effect, so a high concentration of saline water is preferable • Poor quality of effluent (<85%) • Complicated by the different kinds of a solution in each MDC chamber • Large anolyte consumption	• Recirculating the anolyte into the cathode via recirculation MDC (RMDC) (Qu et al., 2012) • Introducing a two-tubular MDC (TTMDC) to provide a direct proton transfer from a bioanode to an air cathode (Jafary et al., 2020) • Protecting the cathode with a carbon cloth and a glass fibre separator (Chen et al., 2012c) • Designing a tubular up-flow MDC (UMDC) with an increased membrane surface area for a convenient ion exchange (Jacobson et al., 2011a) • Installing multiple desalination chambers (Chen et al., 2011) • Installing thin desalination stacks (Kim and Logan, 2011c) • Developing MDC-microbial capacitive deionisation (MCDI) (Wen et al., 2014). • Designing MDC as pretreatment for RO (Mehanna et al., 2010b) • Using a single water source in a hollow fibre microfiltration membrane (HFM)-MDC (Zuo et al., 2018) • Inventing the quadripartite MDC (QMDC) (Ebrahimi et al., 2018)
MEDCs	• Simultaneously treat wastewater, desalination and hydrogen production • Better control on charge transportation across membrane (e.g. Na^+ and Cl^-) • Potential as a self-powered system due to hydrogen generation as fuel (Mehanna et al., 2010a)	• High internal resistance because of a large inter-membrane distance • pH imbalance between anode and cathode chambers (Luo et al., 2010) • EAB viability can be inhibited by the accumulation of protons and chloride ions in the anodic chamber (Luo et al., 2010).	• Installing a bipolar membrane (BPM) next to the anode chamber to create microbial electrolysis desalination and chemical production cells (MEDCCs)
MEDCCs	• Simultaneous wastewater treatment, desalination and chemical production (Chen et al., 2012a) • Simultaneous production of acid and alkali (Chen et al., 2012a) • Desalination occurs without a large pH variation (Chen et al., 2012a) • Increased desalination rates and reduced pH imbalance (Chen et al., 2012a)	• Low desalination rate (Chen et al., 2012b) • Low acid and alkali production rates (Chen et al., 2012b) • EAB are sensitive to a high electrical potential, resulting in the deactivation of MDC operation (Ye et al., 2017).	• Installing four desalination chambers (AEM–CEM stack membrane structure) to improve the desalination rate (Chen et al., 2012b) • Installing two desalination chambers with stacked membrane structure of the BPM–AEM–CEM (Chen et al., 2012b) • Selecting durable EAB under a high electrical potential and increasing the surface area of the anode (Ye et al., 2017)
MCDCs	• Simultaneous electricity generation, desalination and dewatered sludge degradation • Continuous/intermittent removal of salts deposited on electrodes • Less contamination on the electrode. • Facilitated ion migration across desalination to anodes/cathodes • Approximately 70% of salinity is removed (Feng et al., 2013).	• Limited to a low salt concentration • High internal ohmic resistance, thus reducing the desalination rate (Feng et al., 2013; Yuan et al., 2012) • High energy supply to run the MCDC system	• Employing a coupled system of MDC + CDI containing a high salt concentration (Wen et al., 2014) • Combining the MFC stack (in serial or parallel connection or both) and the CDI with an optimum circuit connection (Liang et al., 2015)
OsMDCs	• Water extracted from anolytes increases water production and dilutes saline water (Zhang and He, 2012b). • The discharge of wastewater effluent decreases and becomes beneficial with water reuse (Zhang and He, 2012b). • Improved current generation because of active proton transport due to water flux (Zhang et al., 2011).	• The FO membrane impedes salt transportation, thereby removing a low amount of salt (Zhang and He, 2012b). • Membrane biofouling	• Improving desalination by hydraulically connecting OsMFCs and MDCs (Zhang and He, 2013). • Fabricating an FO membrane with an overlayer of a silver nanoparticle-doped polydopamine to mitigate biofouling and increase power generation (Pardeshi and Mungray, 2013)
MRECs	• Enhanced power generation because of the salinity-driven potential from a RED cell stack (Kim and Logan, 2011b) • RED improves the catalytic activity of EAB; thus, the power of MRECs is generated (Cusick et al., 2013). • The potential of RED alone can drive HER (Kim and Logan, 2011a). • The produced chemical solution by using solely salinity-gradient potential (Luo et al., 2013).	• pH reduction due to chloride ion accumulation in the anode chamber • The number of RED stack cells does not proportionally increase with power generation.	• Replacing an AEM with a BPM next to the anode chamber turning into microbial reverse-electrodialysis chemical production cells (MRECCs) (Luo et al., 2013) • Seven cell pairs are required to recover energy (Cusick et al., 2013). • Applying patterned IEMs to reduce the internal resistance of the RED stack, thus improving the power density (Liu et al., 2014).
SubMDCs	• Concurrent wastewater treatment, nitrate removal from groundwater and bioelectricity generation (Zhang and Angelidaki, 2013) • Simultaneous ammonia recovery and electricity production from anaerobic reactors containing high levels of ammonia (Zhang and Angelidaki, 2015b) • Simpler cell design in contrast to CMDC	• Low-quality purified wastewater and low NH_4^+ and PO_4^{3-} retrieval from SMDDCs (Zhang and Angelidaki, 2013).	• Introducing an advanced microbial nutrient recovery cell (AMNRC), which is equipped with two desalination chambers and three recovery chambers between an anode and a cathode to obtain a high charge transfer efficiency (Chen et al., 2017)
HFM-MDCs	• Membrane fouling problem is alleviated because of a low organic content and aeration flushing (Zuo et al., 2018). • Wastewater is the only water source from which energy can be recovered to drive desalination.	• Desalination efficiency decreases when real wastewater and seawater are used (Jacobson et al., 2011b). • Low wastewater treatment capacity in HFM-MDC	• Improving the desalination performance despite the real wastewater in a dual-anode MDC (DA-MDC) (Liu et al., 2019c). • Investigating the HRT to improve wastewater treatment capacity in DA-MDC


a)
e-
e-
e-
e-
Fresh water
CO2
Substrate
e-
h+
Na+
Cl-
H2O
e-
O2
Bioanode
AEM
Desalination chamber
CEM
Cathode
b)
Wastewater effluent
Seawater effluent
Cathode
Anode
Recirculation
Seawater influent
Wastewater influent
c)
V
e-
Desalinated Seawater
Treated wastewater
Carbon brush anodes
External circuit
Perforated PVC pipe
Cl-
AEM
Air Cathode
Na+
CEM
Seawater
Municipal wastewater
d)
① Air pump
② CEM
③ AEM
④ Cathode
⑤ Anode
⑥ Desalination
e)
concentrated water
desalinated water
concentrated water
Na+
Cl-
H2O
PBA
electrolyte
AEM
salt water
CEM
electrolyte
cathode
f)
Resistor
Multimeter
CO2
e-
Cl-
Na+
Organics
CO2 (from air)
Light
H2O
Algae
O2
Anode
Desalination
Biocathode

MDCs can simultaneously carry out wastewater treatment, desalination and various redox activities either in cathodes or anodes. A redox activity is a reduction–oxidation reaction that participates in electron transfer between two species. In oxidation, electrons are lost, or the oxidation state of ions or atoms in a molecule increases. In reduction, electrons are accepted, or the oxidation state of ions or atoms decreases. A redox activity in an MDC system may involve electricity generation (Chen et al., 2011; Jacobson et al., 2011a; Zhu et al., 2014), hydrogen production (Luo et al., 2010; Mehanna et al., 2010a; Zhu et al., 2014), chemical production (Chen et al., 2012a,b; Liu et al., 2015), nutrient recovery (Chen et al., 2017; Zhang and Angelidaki, 2013), metal reduction (An et al., 2014a,b; Shaohua and Fang, 2013) and biomonitoring (Jin et al., 2016).

The basic principle of each MDC is explained and their technological advancements are reviewed on the basis of the development of various cell configurations in addressing challenges attributed to a conventional MDC design. The discussion is then expanded to highlight the advantage, challenges and reported mitigation approaches in enhancing the performance of MDCs, as summarised in Table 1.

2.1. Microbial desalination cells (MDCs)

In contrast to dual-chamber MFCs, conventional MDCs (CMDCs) are equipped with an extra chamber generally placed in between anode and cathode chambers and designated for desalination. A desalination chamber is created because of the physical separation of the facing anode and cathode of AEMs and CEMs, respectively (Fig. 2a). Like in any type of BESs, electroactive bacteria (EAB) are the main components of MDCs responsible for recovering energy from wastewater. In general, EAB are microbes reliable of transporting electrons through biological membranes into their extracellular environment or vice versa (Rabaey, 2009). The liberated electrons are reactive towards extracellular electron acceptor or donor species. In anodes, electroactive bacteria (EAB) oxidise biodegradable substrates, generating electrons and liberating protons into anolytes. Electrons then extracellularly move to the anode and ferry to the cathode through an external circuit to reduce the terminal electron acceptor present in the cathode chamber. The anion and cation present in the saline solution in the desalination chamber must move across AEMs and CEMs into the anode and cathode chambers, respectively, because of different polarities and imbalanced charge quantities between anodes and cathodes. From this point forward, MDCs are used to name the system in general, whereas CMDCs represent conventional three-chamber MDC designs.

MDCs eliminate the need for using water pressurisation and drawing solutions, which consume 0.98 kPa (Cheng et al., 2014) and 0.276 kWh m^{-3} (Iskander et al., 2017), respectively. Although a direct applied voltage between anodes and cathodes is unnecessary in a conventional MDC system, a considerable amount of energy input is required for pumping the influent into anodes, cathodes, desalination chambers or RED cells. In certain MDC designs and functions, energy is supplied for the aeration of catholytes to drive the oxygen reduction reaction (ORR) or reduce the overpotential for the hydrogen evolution reaction (HER).

In a lab-scale MREC system (a type of MDC), the estimated energy for pumping the concentrate and diluate into RED cells at 0.85 mL min^{-1} only accounts for <2% of the maximum power production (3.6 W m^{-2}) (Kim and Logan, 2011b). The energy for pumping is determined by estimating the head loss required to generate the flow rate through RED cells. In a large-scale MDC, the supplied potential has remarkably reduced the energy consumption per kilogram TDS removed (0.37 kWh $kg\ TDS^{-1}$) relative to that of the system without applied voltage (1.32 kWh $kg\ TDS^{-1}$; (Zhang and He, 2015). This result demonstrates that an additional power input can contribute to energy benefits with an increased desalination efficiency. In terms of capital density, the capital density of MDCs can range from 7031 to 12,305 $ m^{-3} which is higher than those of MFCs and MECs (Zhang and Angelidaki, 2016). It is expected that a tubular MDC may exhibit a comparable capital density with those of MEDCCs and MRECs due to the utilisation of a large membrane.

Nevertheless, MDCs are advantageous BESs due to the negligible toxic effect of the produced oxygen on anaerobes, low sludge production and low carbon footprint (Mehanna et al., 2010a,b). A proof-of-concept study has demonstrated that 90% of salt is removed, corresponding to 31 W m^{-3} power density generation by using the CMDC design assisted with a ferro-ferricyanide cathodic reaction (Cao et al., 2009). A comparative study has suggested that seawater desalination can be better implemented with a ferro-ferricyanide redox mediator as a catholyte, whereas brackish water is better to be desalinated via the air cathode method (Ramírez-Moreno et al., 2019). However, the use of costly ferrocyanide as a catholyte is impractical in large-scale applications because scaling-up is the ultimate focus.

An air cathode has been the most practical approach of a cathodic reaction in MDCs because it has a simple set up and eliminates the need for aeration for ORR. However, the drawback of using air cathodes in a MDC system is the poor power generation to drive wastewater and desalination. The issue is addressed with sodium hypochlorite as a catholyte, which has been proven as a compatible alternative to ORR with an enhanced performance attributed to its fast kinetics (Borjas et al., 2017). However, with a prolonged time, sodium hypochlorite can deteriorate the membrane surface and trigger environmental issues associated with chlorinated by-products.

When external power is supplied to an MDC system, hydrogen can be produced on a cathode with a better control of potentials between electrodes. This system is known as a microbial electrodialysis cell (MEDC; Luo et al., 2010; Mehanna et al., 2010a). The generated hydrogen can be used to self-power the system or stimulate additional processes, given that more than twice the amount of hydrogen gas can be recovered compared with that of the energy consumed to run the system (Mehanna et al., 2010a). Luo et al. (2010) found that applied voltage and cathode buffer capacity are the two main factors affecting H_2 production. This result suggests that the influence of an external power supply on the microbial activity in cathodes is greater than that in anodes (Luo et al., 2010).

One of the drawbacks that hamper the progress on CMDCs is the poor quality of desalinated water to a level of the freshwater standard. Mehanna et al. (2010b) proposed that MDCs are suitable for pre-desalination devices of RO mainly because of the lower energy needed to process pre-desalinated saline solutions. A distinct design of a tubular up-flow MDC (UMDC) is invented to improve the desalination rate (Fig. 2b), realising >99% of NaCl at HRT 4 h, corresponding to twice that achieved in another study (Luo et al., 2010). In other words, HRT is the average duration that a solution/compound remains in a chamber/bioreactor. It is expressed as the volume of the chamber divided by the flow rate of the influent resulting in a unit time. Another limitation of CMDCs is the unequal pH between anolytes and catholytes, leading to highly acidic and basic conditions in anode and cathode chambers, respectively. These conditions disfavour the growth of bacteria in the anode chamber, whereas potential losses occur in the cathode chamber. This issue has been encountered by expanding the volume of anolytes (Cao et al., 2009) or by introducing acids or alkali (Cao et al., 2009; Chen et al., 2011; Jacobson et al., 2011a). The approaches can increase operational costs to an extent, and safety measures should be considered during the large-scale handling of highly concentrated acids or bases. Therefore, Qu et al. (2012) proposed that this issue can

Fig. 2. a) The illustration of typical CMDC, b) UMDC, reprinted with permission from Jacobson et al. (2011a), c) TIMDC with different membrane configuration, reprinted with permission from Jafary et al. (2020), d) QMDC with 9 chambers in a single cell, reprinted with permission from Ebrahimi et al. (2018), e) PMDC with photobioanode, reprinted with permission from Liang et al. (2016), f) PhMDC with photobiocathode, reprinted with permission from Kokabian and Gude (2013).

be alleviated by merely recirculating the anolyte into the cathode chamber, resulting in a balanced pH between electrodes. The development of this system is called as a recirculation MDC (RMDC). However, this approach may induce microbial growth on the cathode's surface. Chen et al. (2012c) responded to this issue by clamping a piece of glass fibre wool on the carbon cloth in contact with water (i.e. the air cathode approach).

A recent approach in alleviating the pH imbalance issue has been achieved by inventing a novel two-chamber tubular MDC (TTMDC), which is installed with inner and outer cylinders (Jafary et al., 2020). In contrast to UMDCs (Jacobson et al., 2011a), bioanodes are contained in the outer cylinder, making a close contact with the air cathode. The saline water flowing in the inner cylinder wrapped with an AEM (top) and a CEM (bottom), as shown in Fig. 2c. Allowing the direct transfer of proton from anode to cathode without membrane separation has improved the overall MDC performance because of the balanced pH of the electrolyte solution even with a non-buffered domestic wastewater. However, the conventional tubular up-flow MDC (UMDCs) and TTMDCs have yet to be comparatively analysed.

Another issue to tackle is the large volume of anolytes used in CMDCs, which are needed to desalinate a small volume of saline water. The smaller ratio of the volume of anolytes to the volume of desalinated water is more economical in terms of anolyte consumption. To mitigate this issue, Ebrahimi et al. (2018) invented a quadripartite MDC (QMDC). QMDCs consist of one anode chamber, which is shared with four sets of desalination and cathode chambers, as depicted in Fig. 2d. A proof-of-concept study has suggested that a comparable desalination efficiency (72.8%) is obtained with a three-chamber CMDC (78%) with a lower ratio of an MDC system (Ebrahimi et al., 2018). Thus, its operational cost is expected to be lower than that of an MDC design with a high ratio.

Photo-MDCs (PMDCs) have been an attractive way to enhance the power output of MDCs by converting abundant sunlight energy into electrical energy without increasing the operational cost of a system. Liang et al. (2016) developed a new composite photobioanode (PBA) made of a nanostructured semiconductor hematite (α-Fe_2O_3) (Fig. 2e) to harness sunlight for powering PMDCs. The semiconductor is coupled with graphite as a anode carrier, given that it has an excellent biocompatibility. In addition, hematite itself is characterised as a good light absorber between ultraviolet and visible light spectra (Morrish et al., 2011). A study has shown that a PMDC produces the highest achievable current density of 8.8 A m^{-2}, which corresponds to two-fold enhancement compared with that obtained with an MDC. This result indicates that PMDCs have the potential to generate concurrent electricity and perform desalination assisted with an abundant source of solar energy. However, photocatalytic materials are prone to photocorrosion, affecting the electrode durability over a long time period.

Another promising approach is the use of a photosynthetic biocathode with the presence of algae and abundant solar energy in MDCs (PhMDC) for the simultaneous wastewater treatment and desalination (Fig. 2f) (Kokabian and Gude, 2013). In biocathode–PMDC, as algae grow, carbon dioxide is consumed during the day to produce metabolic oxygen, which acts as the terminal electron acceptor. Interestingly, the metabolic CO_2 produced by EAB under anaerobic conditions in an anode chamber can be channelled to the cathode chamber for photosynthesis, creating in situ synergism between EAB and microalgae. The algal biomass is believed as precious bioproducts, which are in contrast to the formation of sludge in other biological systems. A study has revealed that non-stop illumination is impractical to be implemented in a large-scale application; thus, a periodic cycle of light and dark is rather workable. Additionally, biomass proliferation is influenced by the quality and quantity of light exposure in which the maximum growth is achieved under a clear light (Blair et al., 2014).

The common issue of accumulated protons in anodes as a result of the installation of AEMs has not been investigated in a large scale. However, a recent report has proposed a more practical way of alleviating pH imbalance by providing a direct transfer of the generated protons in anolytes to catholytes in TTMDCs (Jafary et al., 2020). The design also shortens the distance between anodes and cathodes, reducing the internal ohmic resistance of a TTMDC system. In terms of tubular shape, a large-scale TTMDC may outperform desalination and wastewater treatment performed in UMDCs.

2.2. Microbial electrolysis desalination and chemical production cell (MEDCC)

In addition to the problem faced by CMDCs and MEDCs aforementioned, the accumulation of Cl^- in anolytes can occur and be increased remarkably because of the transfer of Cl^- from a desalination chamber. Its transfer is in response to the balance of H^+ production by EAB in anodes. However, the built-up Cl^- in anolytes is toxic to microbial viability. Kim and Logan (2013) verified that the exoelectrogenic activity is retarded by the high concentration of anions produced in the anode chamber containing a high load of organic matter. In a MFC system, the maximum power generation is reduced by 12% when chloride concentration reaches 500 mM (Oh and Logan, 2006). In addition, the presence of excessive salt in the anode chamber can only provide a more habitable condition for specific types of bacteria than others in a community (Mehanna et al., 2010a). The use of phosphate buffer saline creates another problem of phosphate ion transport to the desalination chamber across an AEM. The accumulation of phosphate ions may lead to the excessive deposition of Ca^{2+} and Mg^{2+} in the desalination chamber if actual seawater is used. In the first attempt to mitigate these issues, a bipolar membrane (BPM) is installed next to an anode chamber (facing anode), in addition to AEMs and CEMs (facing cathode), which are placed in between the anode and the cathode (Chen et al., 2012a). The obtained cell is known as a microbial electrolysis desalination and chemical production cell (MEDCC), which comprises four chambers, namely, an anode chamber, an acid production chamber, a desalination chamber and a cathode chamber (Fig. 3a).

In principle, acids and bases are produced through the transport of H^+ and OH^- derived from water dissociation in the presence of a BPM. In this case, H^+ migrates into an acid production chamber and couples with Cl^- that migrates from the desalination chamber to produce an acid (e.g. HCl). Conversely, OH^- moves across an AEM into an anode chamber to neutralise anolytes, a favourable condition for microbial growth. Concurrently, cations (e.g. Na^+) migrate to the cathode chamber and couple with the generated OH^- through an ORR to finally yield an alkali solution (e.g. NaOH). However, an increase in pH in the cathode likely increases the cathode overpotential, which incurs an additional 0.059 V pH^{-1} to the system. Therefore, the smart way to reduce the cathodic pH is to channel the produced acid into the cathode chamber. These simultaneous processes desalinate the saline solution in the middle chamber without phosphate leakage; thus, problems related to pH imbalance between anodes and cathodes are resolved, and a valuable chemical compound in MEDCCs is produced (Chen et al., 2012a).

Although MEDCCs have been used for wastewater treatment, desalination and chemical production, the acid and alkali production rates should be improved further. Motivated by the excellent desalination performance of five desalination-stacked chambers (Kim and Logan, 2011c), Chen et al. (2012b) proposed a modified design of stacked MEDCCs to enhance the chemical production and desalination rate (Fig. 3b). They demonstrated that the maximal desalination rate can be achieved in four pairs of AEM–CEM stacked MEDCC structures with 1.5 mm membrane spacing, and this rate is 43% higher than that of the standard MEDCC. As a result, a simultaneous reaction of desalination and chemical production is achieved at its maximum with a two-desalination-chamber MEDCC in which the BPM–AEM–CEM stacked structure and the membrane distance of 3 mm are installed. In another investigation, seven pairs of membrane stacks are needed to cater to the potential required for H_2 evolution and desalination (Zhu et al., 2014). The application of MEDCCs is not limited to the production of HCl and

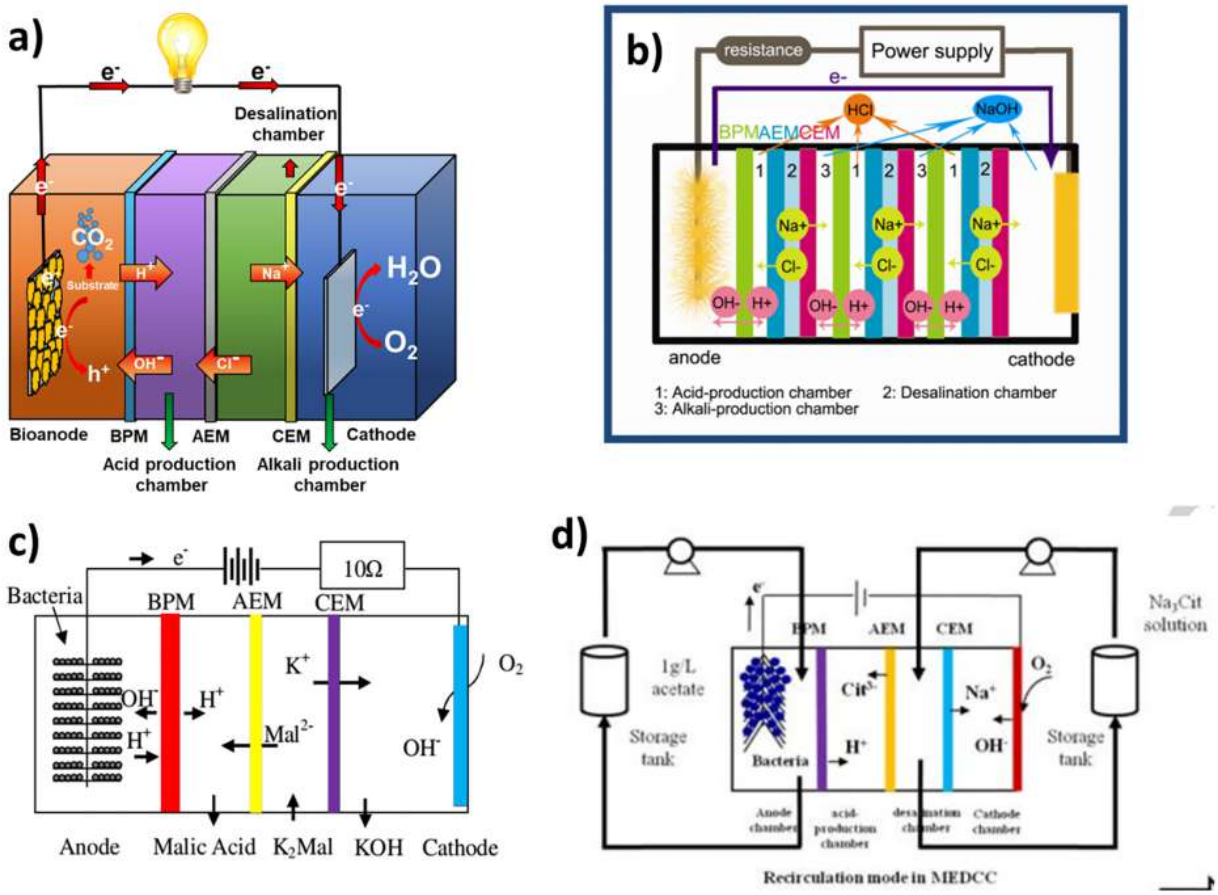


Fig. 3. a) The illustration of typical MEDCC, b) MEDCC for acid and alkali production, reprinted with permission from Chen et al. (2012b), c) stacked MEDCC, reprinted with permission from Chen et al. (2012b), d) MEDCC for malic acid production, reprinted with permission from Liu et al. (2015), e) MEDCC for citric acid production, reprinted with permission from Luo et al. (2017).

NaOH, but its application is diversified to produce malic acid (Liu et al., 2015) (Fig. 3c) and citric acid (Luo et al., 2017) (Fig. 3d) by manipulating the order and type of membranes (Liu et al., 2015; Luo et al., 2017).

Advancements in MEDCCs have demonstrated that the rate of acidic and basic solutions and desalination can be enhanced by configuring the repeated membrane set consisting of BPMs, AEMs and CEMs (Chen et al., 2012b). However, the production of a caustic solution from MEDCCs requires an energy input that adds up to operational costs. If a chemical is the targeted product, it should be highly pure and marketable. Although its purity and marketability are ensured, its price must be comparable with its market price. Revenues should offset the cost for energy input. Alternatively, the produced acidic and basic solution can be used to balance the pH in the anode and cathode chambers with proper control. However, the former approach is more favourable, considering the cost of energy supply that needs to be compensated. Future research should focus on the durability of membrane towards extremely low and high pH if a large scale is to be realised.

2.3. Microbial capacitive desalination cell (MCDC)

Another approach to address the migration and accumulation of salt ions in the anode and cathode chambers in CMDCs is by adapting the capacitive deionisation (CDI) concept into MDC systems (Forrestal et al., 2012b; Yuan et al., 2012). The integration of CDI into CMDCs as a single unit cell is known as MCDCs (Fig. 4a). The migration of salt ions towards oppositely charged electrodes during desalination can be adsorbed by a double-layer capacitor created on the electrode's surface. The adsorbed ions can be retrieved by removing the electrochemical potential while the CDI membrane is immersed into a liquid to produce a concentrate for salt reclamation. In this way, ions are not accumulated in anolytes and catholytes, thereby avoiding the pH imbalance in the system.

MCDCs are the result of the alteration of capacitive deionisation (CDI) by incorporating anion and cation exchange membranes on anode and cathode surfaces, respectively. CDI is considered a simple technology to remove salts from brackish water or seawater for water reclamation (Oren, 2008). The mechanism of CDI is based on electrical double-layer (EDL) theory, which involves adsorption and desorption steps. During adsorption, counter ions present in solutions move towards and are held onto electrodes under an electrostatic force when an external voltage is applied between electrodes. In desorption, when a reverse voltage is applied between two electrodes, the adsorbed ions become repelled away from electrodes and carried away by the flushing stream; the two electrodes can be regenerated by shorting the two electrodes (Forrestal et al., 2012a).

A pioneering study has demonstrated the ability to simultaneously generate power and conduct desalination without poisoning anolytes and catholytes. For example, Forrestal et al. (2012a,b) utilised an activated carbon cloth (ACC) as an adsorptive electrode material and used built-up capacitive double layers for electrochemical ion adsorption. They installed two CDI membrane assemblies of CEM/ACC/Ni-Cu into MDC cells, where each Ni—Cu layer faces the desalination chamber, thereby forming three chambers resembling a traditional MDC

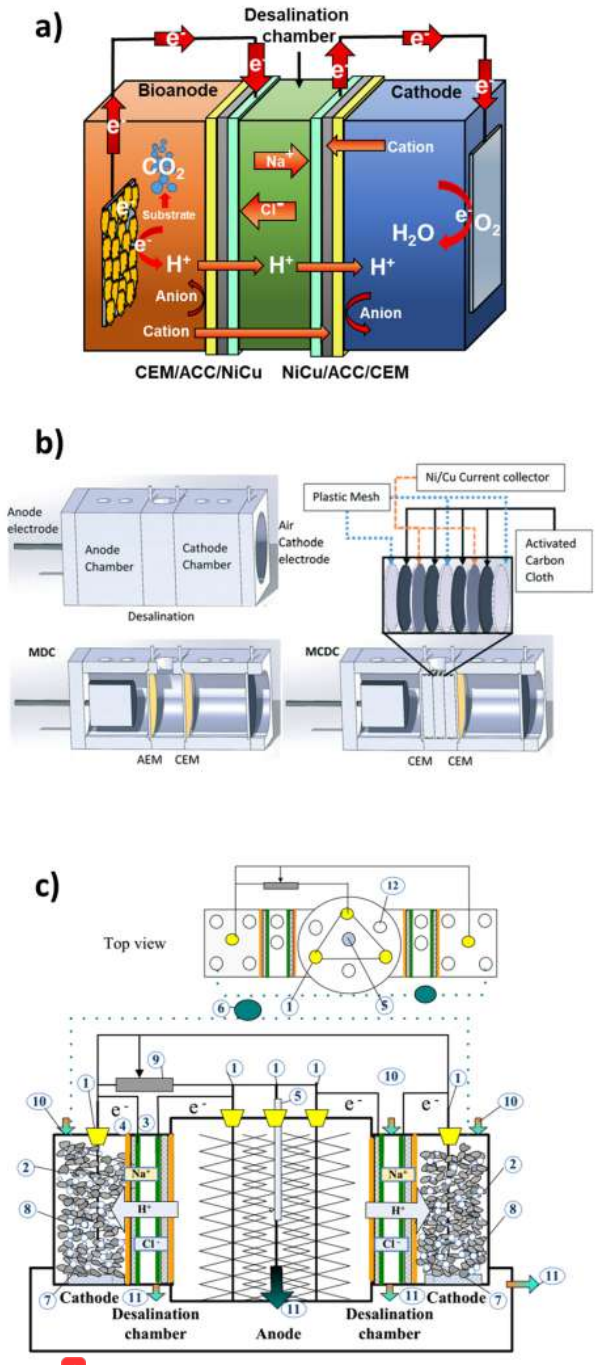


Fig. 4. a) The illustration of pioneered MCDC with CDI membrane of CEM/ACC/NiCu|NiCu/ACC/CEM, reproduced with permission from Forrestal et al. (2012b), b) MCDC with CDI membrane module of consecutive PM/ACC/Ni-Cu, reprinted with permission from Forrestal et al. (2015), c) five chambers MCDC with CDI membrane of ACC/Ni foam/CEM|AEM/Ni foam/ACC, reproduced with permission (Meng et al., 2017).

(Fig. 4a) (Forrestal et al., 2012b). This MCDC configuration achieves the desalination efficiency of about 7-fold to 25-fold higher than that measured in conventional CDI (Forrestal et al., 2012b). In another series of studies, two different electrode arrays of ACC/Ni-Cu/AEM and CEM/Ni-Cu/ACC are applied to anodes and cathodes, respectively (Forrestal et al., 2012a). After one batch of cycle, MCDCs discard 69.4% of salt from the desalination chamber through electrode adsorption, corresponding to the adsorption of 61–82 mg of TDS in 1 g of ACC electrode (Forrestal et al., 2012a).

MCDCs can be used to simultaneously remove organic pollutants, salt from shale gas-produced water and electricity generation (Fig. 4b) (Forrestal et al., 2015). An MCDC system has three modules containing a plastic mesh (PM)/ACC/Ni-Cu, which is arranged consecutively in each module between AEMs and CEMs, and it can remove 2760 mg of TDS L^{-1} h^{-1} and 170 mg COD L^{-1} h^{-1}, which are equivalent to 18- and 5-fold quicker than that of CMDCs, respectively (Forrestal et al., 2015). Continuous investigations on MCDCs have shown that the feasibility of preventing salt ion migration into anode and cathode chambers is limited by the slow culture enrichment and an unstable long-term operation ascribed to the use of a liquid anodic substrate and domestic wastewater (Forrestal et al., 2012b). The issues related to slow culture enrichment and salt accumulation in the anode are alleviated using five-chamber biocathode MCDCs, which are equipped with four sets of CDI membranes and dewatered sludge as an anodic fuel (Fig. 4c) (Meng et al., 2017).

Laboratory testing has shown that MCDCs have been used to desalinate low-salinity water and unconventional natural gas-produced water. The prime factors affecting desalination and charge transfer in MCDCs include the internal resistance of MDCs, the internal resistance and capacitance of CDI membranes and the open circuit voltage. These factors can be more challenging during scale-up. For scaling-up purposes, inexpensive adsorptive materials, such as modified titanium and silica, should be employed. A large reactor design should be equipped with an improved adsorption ability, an enhanced diffusion rate and a high ACC loading. Moreover, a large reactor should be equipped with a modular stack and a tuneable operation scheme to increase the incorporation of assembly regeneration and desalination in several units to improve salt removal management and optimise water reclamation. MCDCs render a complex configuration. Although MCDCs have been proven successful in lab-scale testing, whether MCDCs can be scaled up at a low capital cost has remained unclear.

2.4. Osmotic MDCs (OsMDC)

In principle, FO is a spontaneous process transporting water across a semipermeable membrane in response to the osmotic pressure gradient from a high water content (low solute concentration) to a low water content (high solute concentration) (Zou et al., 2017). With natural osmotic pressure and substrate oxidation in anode chambers, the concept of FO has gained considerable attention for wastewater treatment through BESs to recover freshwater and improve power generation.

The replacement of IEM by FO membranes in MFC or MDC reactors creates an FO–BES-based system known as an osmotic microbial fuel cell (OsMFC) or an OsMDC. Both integrated systems have been investigated, but few studies have been conducted for OsMDCs (Ismail and Ibrahim, 2015; Werner et al., 2013; Zhang and He, 2012b). The installation of FO membranes drives water diffusion from anodes to desalination chambers, diluting saline water with significant water increment via water flux (Fig. 5). This process impedes the migration of salt ions from the desalination chamber into the anode.

The occurrence of water flux promotes better proton/ionic transportation across a FO membrane compared with that of conventional MDCs; consequently, electricity generation is enhanced (Sumikura et al., 2008; Werner et al., 2013; Zhu et al., 2015). Studies have shown that saltwater dilution and overall desalination performance can be improved via OsMDCs, which benefit from the rejection of most of the solids dissolved by the FO membrane, less susceptibility to fouling, low energy input and high water recovery (McGinnis and Elimelech, 2007; Sumikura et al., 2008; Werner et al., 2013). Interestingly, the use of FO membranes is more economical than that of AEMs (Zhang and He, 2012b). According to a theoretical study, water flux is a factor

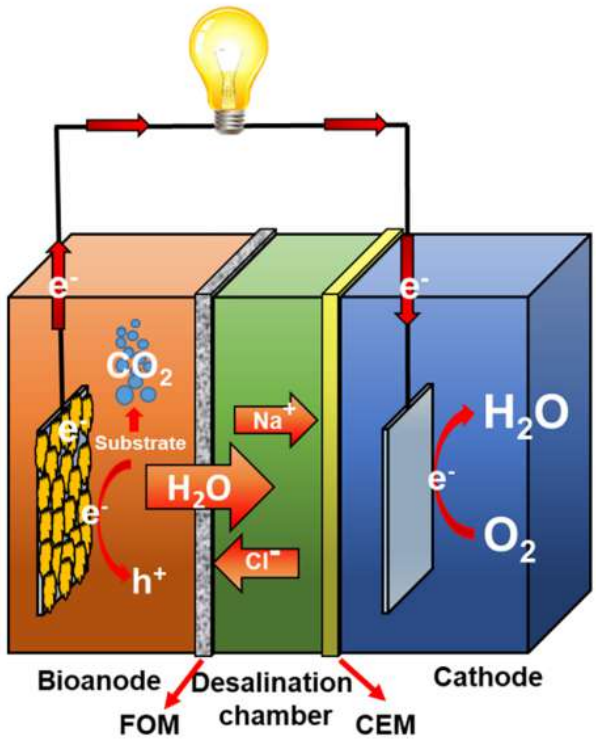


Fig. 5. The illustration of typical OsMDC.

1 limiting the improvement of electricity generation in OsMFCs provided that membrane resistance is minimised (Qin et al., 2015). An OsMFC study has indicated that the cake-enhanced concentration polarisation in a fouled membrane improves ion transportation, thereby generating a higher amount of electricity in OsMFCs than that produced before fouling (Ge and He, 2012). The encouraging effect of the enhanced current generation under the unfavourable condition of biofouling is also verified in OsMDCs used to desalinate real oil field-produced water, which acts as a draw solution (Ismail and Ibrahim, 2015).

OsMDCs can be applied in the form of integrated wastewater treatment, water reuse and desalination, which provides environmental, energy production and economic benefits. Unlike standalone FO systems, high-quality water from wastewater can be recovered, and salinity can be reduced through dilution even though power is generated using OsMDCs. The FO membrane is much cheaper than the AEM, so a lower capital cost for scaling-up OsMDCs than that for scaling-up CMDCs is expected. The dilution effect via water flux is incapable of removing salt; nevertheless, it can effectively reduce the salinity of saline water. The reduction in salinity can potentially curtail energy consumption in RO if OsMDCs are used as RO pretreatment.

2.5. Microbial reverse-electrodialysis cells (MRECs)

MRECs are used to counter the problems faced by CMDCs, including a low desalination rate, a poor electron transfer efficiency, a limited volume saline solution and a low amount of power generation. The complete design of MRECs includes multiple desalination chambers consisting of IEM pairs forming a RED stacked cell, placed between the anode and cathode of MREC cells (Fig. 6a). The installation of pairwise AEMs and CEMs in RED cells provides repeated cells for high- and low-concentration saline solutions, creating a salinity gradient-driven potential. As a result of the salinity gradient formed in RED stacked cells, respective anions and cations are transported across AEMs and CEMs from high-concentration solutions to low-concentration solutions, which flow in opposite directions. Anodic oxidation and cathodic reduction occur simultaneously to generate fuel or electricity (Kim and Logan, 2011b). These mechanical advantages cannot be acquired in the individual system of RED stacked cells and MECs because RED stacked cells are unable to generate power; however, MECs require a supply of external voltage (e.g. water reduction to produce H_2). In a theoretical calculation, one pair of AEMs and CEMs can generate approximately 0.1–0.2 V of potential based on different ratios of high and low concentrations of a salt solution (Kim and Logan, 2011b). An optimum number of cell pairs should be investigated to balance between operational cost and cell performance and further increase the potential.

With an increase in voltage in response to the additional IEM pairs, MRECs can produce a nonspontaneous hydrogen evolution reaction (HER) without an external power input to a system (Kim and Logan, 2011a). A five-membrane pair of AEMs and CEMs in a RED stack forming a series of salinity gradients is sufficient to overcome the electrode overpotential for HER (Fig. 6b), in addition to the oxidation of organic matter in an anode driven by EAB. A study has reported that a small quantity of RED cell pairs, together with an optimised flow rate of a saline solution, use approximately 1% of the produced energy for pumping the solution (Kim and Logan, 2011a). This finding demonstrates that H_2 can be generated from abundant and free supplies of river water, seawater and organic wastewater.

Replacing the saline solution with ammonium bicarbonate in RED stacked cells and cathode chambers in MREC has significantly improved the power generation and COD removal compared with that in MFC modes (Cusick et al., 2013). The utilisation of ammonium bicarbonate as another source of salinity gradient in RED stacked cells encounters the limited area in coastal areas and estuaries. Furthermore, an energy-intensive pre-treatment is required to minimise membrane fouling if seawater and brackish are used (McGinnis and Elimelech, 2007). When ammonium bicarbonate is heated at ~60 °C, ammonia and CO_2 are evaporated and condensed to form an HC solution (McGinnis and Elimelech, 2007). Waste heat and typical distillation techniques can be employed to regenerate HC and LC solutions. Ideally, heat from waste can be converted to electricity, thereby generating hydrogen in MRECs.

In comparison with MFCs, the addition of one and two pairs of RED stacked cells has increased the power production into 5.6- and 6.3-fold, which are equivalent to 1.7 and 1.9 W m^{-2}, respectively. At maximum power generation, about 30–50 times of the enhancement of COD removal is achieved for one and two RED cell pairs of MRECs compared with that obtained in single-chamber MFCs (McGinnis and Elimelech, 2007). The observed result is due to the increased RED stack potential and minimised charge transfer resistance in the anode chamber. More membrane pairs are needed to self-generate hydrogen. For example, in a five-stack cell pair-modified MREC, ammonium bicarbonate is used as a RED stack solution to drive hydrogen evolution. In this study, the stack arrangement of MRECs is modified by adding a low-concentration cell next to the anode chamber to minimise ammonia crossover. As a result, 40% of ammonia–nitrogen, a maximum hydrogen yield of 3.5 mol H_2 mol^{-1} acetate and a Coulombic efficiency of 83% are achieved (Luo et al., 2013).

Although additional RED cell pairs can increase the cell voltage, this increase may be hindered by the internal resistance within a RED stack. The internal resistance may be attributed to the use of nonconductive spacers, such as a thick gasket that potentially blocks the transportation of ions across the membrane, leading to the development of the ohmic resistance of a system. This problem is known as a spacer shadow effect, which is alleviated with newly patterned AEMs and CEMs (Liu et al., 2014). The patterned AEMs and CEMs are prepared by casting the membranes on a protruded polydimethylsiloxane (PDMS) mold, which is fabricated on a perforated SMDDC (PTFE) plate beforehand (Liu et al., 2014). Approximately 22 Ω of RED stack resistance is reduced, resulting in nearly 1.4-fold enhancement of power density compared with that in

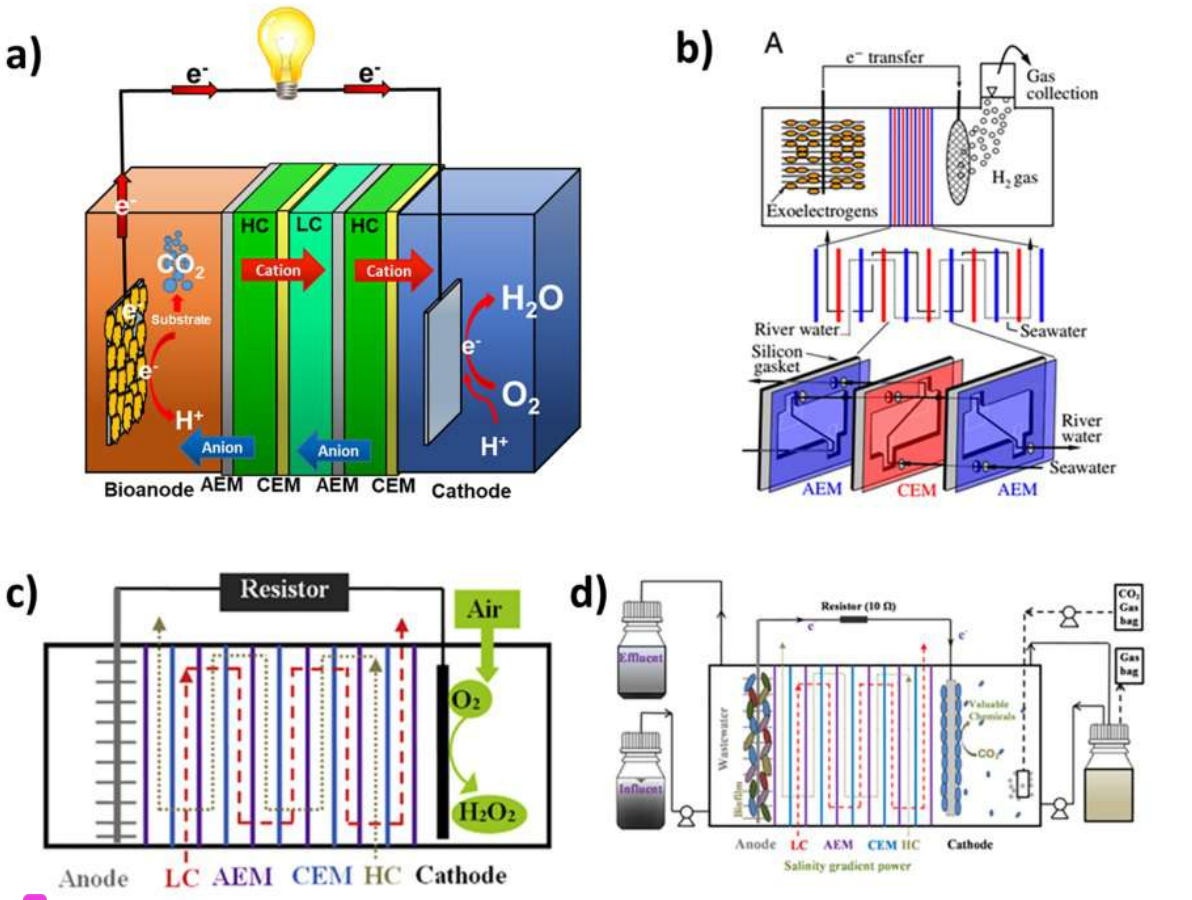


Fig. 6. a) The illustration of typical MRECs design, b) MREC with enhanced power production, reprinted with permission from Kim and Logan (2011b), c) MREC for hydrogen peroxide production, reprinted with permission from Li et al. (2017a), d) MREC for reduction of CO_2 into value-added chemical, reprinted with permission from Li et al. (2018b).

a commercially available membrane and a nonconductive spacer (3.44 ± 0.02 W m^{-2}) (Liu et al., 2014). The use of a patterned membrane consequently increases the overall MREC performance, including the COD removal rate, energy efficiency and Coulombic efficiency (Liu et al., 2014).

With the power of salinity gradient, MRECs are employed to synthesise H_2O_2 from ORR (Fig. 6c) (Li et al., 2017a). Without an external power supply, a high H_2O_2 concentration at 778 ± 11 mg L^{-1} is obtained under the optimised effect of the airflow rate and the catholyte concentration that govern the cathode potential (Li et al., 2018b). This result indicates that O_2 is electrochemically reduced H_2O_2 in a wide pH range (2−10). Importantly, the amount of energy consumed to produce 1 kg of H_2O_2 is as low as 0.45 ± 0.03 kWh.

Overcoming environmental and energy issues is an attractive strategy through the conversion of harmful CO_2 emission to value-added chemicals and biofuels. A biocathode that pre-acclimatises with *Sporomusa ovata* is installed to catalyse the conversion of CO_2 to acetate and methanol by using a modified MREC system with salinity gradient energy (Fig. 6d) (Li et al., 2018b).

MRECs have shown a significant progress in various applications concerning energy and environmental issues. MRECs rely on the production of electrical potential as a result of salinity gradient across the stack of alternating pairs of AEMs and CEMs. These pairs provide alternating HC and LC chambers. Obviously, MRECs need a large number of membrane pairs to self-power a system, resulting in high capital and installation costs. A large-scale MREC necessitates the use of a cost-effective IEM that is inexpensive and exhibits a low internal resistance to induce high-energy recovery and efficiency. In addition to cheap raw materials, new and cost-effective membranes should be developed to provide a large active area for ion transport and minimise an intermembrane distance, as demonstrated by patterned IEMs (Balster et al., 2010; Vermaas et al., 2014). The installation of patterned IEMs can eliminate the use of spacers, which normally increase the electrode spacing in RED cells.

2.6. Submersible MDCs (SubMDCs)

The versatility of the existing MDC technology to address environmental issues is a novel achievement to the extent that nitrate removal in groundwater is also feasible. A proof-of-concept study has introduced a new and simpler cell design called submerged microbial desalination-denitrification cells (SMDDCs) to accomplish the in situ removal of nitrate from groundwater (Zhang and Angelidaki, 2013). The cells are only equipped with anode and cathode chambers, where AEMs and CEMs are installed at each end of chambers in contact with a wastewater/saline water source. SMDDCs are featured without a desalination chamber, making the design simpler than that of CMDCs (Fig. 7a). In a lab-scale setup, MEDCCs are submerged in synthetic groundwater, while anode and cathode chambers are filled with wastewater. In a real application, SMDDCs likely play multiple roles in denitrification, desalination and electricity generation by submerging the cells into the existing reservoir or subsurface of groundwater sources (Zhang and Angelidaki, 2013).

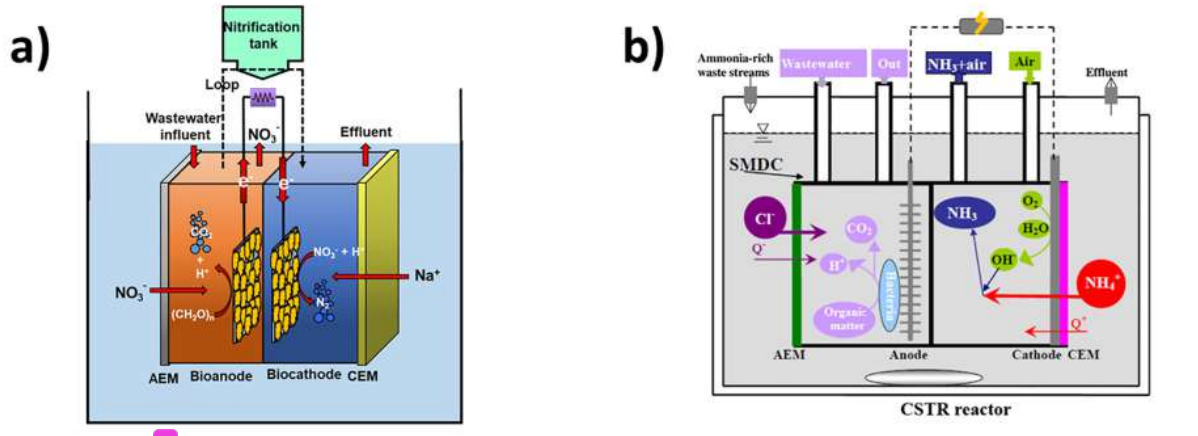


Fig. 7. a) The illustration of pioneered SMDDC design reproduced with permission from Zhang and Angelidaki (2013), b) SMDARC for concurrent ammonia recovery and electricity generation in CSTR, reprinted with permission from Zhang and Angelidaki (2015b).

The principal of this new concept involves three critical steps. 1) Driven by bioanodic electron generation, NO_3^- and Na^+ are diffused from the desalination chamber into anode and cathode chambers, respectively. 2) NO_3^- is then migrated with the anode effluent into the cathode chamber. 3) Next, NO_3^- is subsequently reduced to nitrogen gas catalysed by an autotrophic denitrification reaction on a biocathode to complete the circuit, producing current for the first cycle. Based on the concept introduced by Zhang and Angelidaki (2013), SMDDCs offer simplicity, given that no additional desalination chamber, external power supply, draw solution and water pressure is needed, and this finding is contrary to conventional processes, such as heterotrophic denitrification, reverse osmosis and electrodialysis. The groundwater under a treatment with SMDDCs experienced no cross-contamination of bacterial discharge from an anode or a cathode, as the cell system only allows anions and cations to penetrate through IEMs.

In a pioneering study, 90.5% of nitrate from groundwater is removed at HRT of 12 h, and a current density of 3.4 A m^{-2} is produced (Zhang and Angelidaki, 2013). The ionic strength of groundwater, nitrate concentration, external resistance and HRT are the limiting factors governing the performance of SMDDCs. Although the nitrate removal rate and current generation are positively affected by external nitrification, the total nitrogen removal is not influenced (Zhang and Angelidaki, 2013).

The replacement of a biocathode to an abiotic cathode has turned SubMDCs into a submerged microbial desalination-ammonia recovery cell (SMDARC) by maintaining the AEM and CEM in contact with water (Fig. 7b) (Zhang and Angelidaki, 2015b). Cations such as Na^+ and NH_4^+ can penetrate across CEMs to a cathode chamber (Kuntke et al., 2011, 2012), so SMDARCs can be used to address the high concentration of ammonia in anaerobic reactors because it affords to impede anaerobic digestion (Zhang and Angelidaki, 2015a,b). In an experiment performed in a continuously stirred tank reactor (CSTR; batch mode), the ammonia concentration decreases by 88% from its initial concentration of 6 g·N L^{-1} within 30 days, and this value corresponds to a recovery rate of 80 g·N m^{-2} d^{-1}. With advantageous outcomes such as in situ ammonia recovery, electricity generation, potential wastewater treatment and biogas enhancement, SMDARCs can become a cost-effective green technology for nutrient recovery from wastewater and energy production in the future.

A proof-of-concept-study has presented that SubMDCs may be utilised to remove nitrate from groundwater in situ and recover ammonia from anaerobic digestion with concurrent energy recovery. SubMDCs can be operated without the needs of power input, water pressure, draw solution, and electron donor. Given that anode and cathode chambers are enclosed with AEMs and CEMs, bacterial discharge unlikely occurs. The reactor design is simple, compact and workable for in situ applications, thus facilitating the up-scaling effort. Laboratory testing indicates that the nitrate and ionic strength of groundwater deplete at the end of the process, slowing down the activity and performance of SMDDCs (Zhang and Angelidaki, 2013). Therefore, the ideal operation with SMDDCs is in a continuous flow of media, which take an advantage of an abundant source of real groundwater and wastewater. The size of a reactor should be optimised between its volume and the volume of the surrounding solution to be treated. A small ratio may escalate the internal resistance because of the poor mass transfer between the membrane and the bulk solution around SubMDCs. A large ratio is advantageous for salt removal, but it increases construction costs (Jacobson et al., 2011b).

2.7. Hollow fibre membrane MDCs (HFM-MDC)

Several challenges have remained unsettled, thereby slowing down the progress of MDC technology into field applications. They are related to the incomplete removal of organic compounds and other species (e.g. phosphorus, nitrogen, microorganisms and suspended solids) from anaerobic wastewater effluent. Studies have indicated that the COD removal of anode wastewater effluent is lower than 85% (Davis et al., 2013; Luo et al., 2012c; Qu et al., 2012; Stoll et al., 2015; Zhang and He, 2015).

Membrane fouling is a dominating issue during the long-term operation of seawater desalination (Ping et al., 2013; Zhang and He, 2015), brackish water (Ping et al., 2015) and petrochemical wastewater by using traditional MDCs (Forrestal et al., 2015; Stoll et al., 2015). At least three different types of water need to be supplied into CMDCs, which are wastewater, saline water and catholytes into the anode, desalination/RED stack and cathode chamber, respectively. This operational setup convolutes the MDC system, causing the volume of the diluate to decrease markedly to <40% (Chen et al., 2012b; Kim and Logan, 2011c; Luo et al., 2012c). In enhancing the power generation in CMDCs (Zhang and He, 2015), certain studies have used noble metals (Qu et al., 2012) and nanocarbon materials as catalysts (Zuo et al., 2016b), utilising an air cathode to prevent energy-intensive aeration (Zuo et al., 2016b) and chemicals such as ferricyanide (Cao et al., 2009), which are expensive, too fragile, easy fouling and toxic if they are implemented in large-scale applications.

A study has attempted a new MDC concept by integrating a hollow fibre membrane (HFM) into a MDC, which is designated as HFM-MDC, to overcome these challenges (Zuo et al., 2018). The HFM-MDC is functioned by flowing wastewater into the anode, channelling to the HFM biocathode in the subsequent step and finally into the central

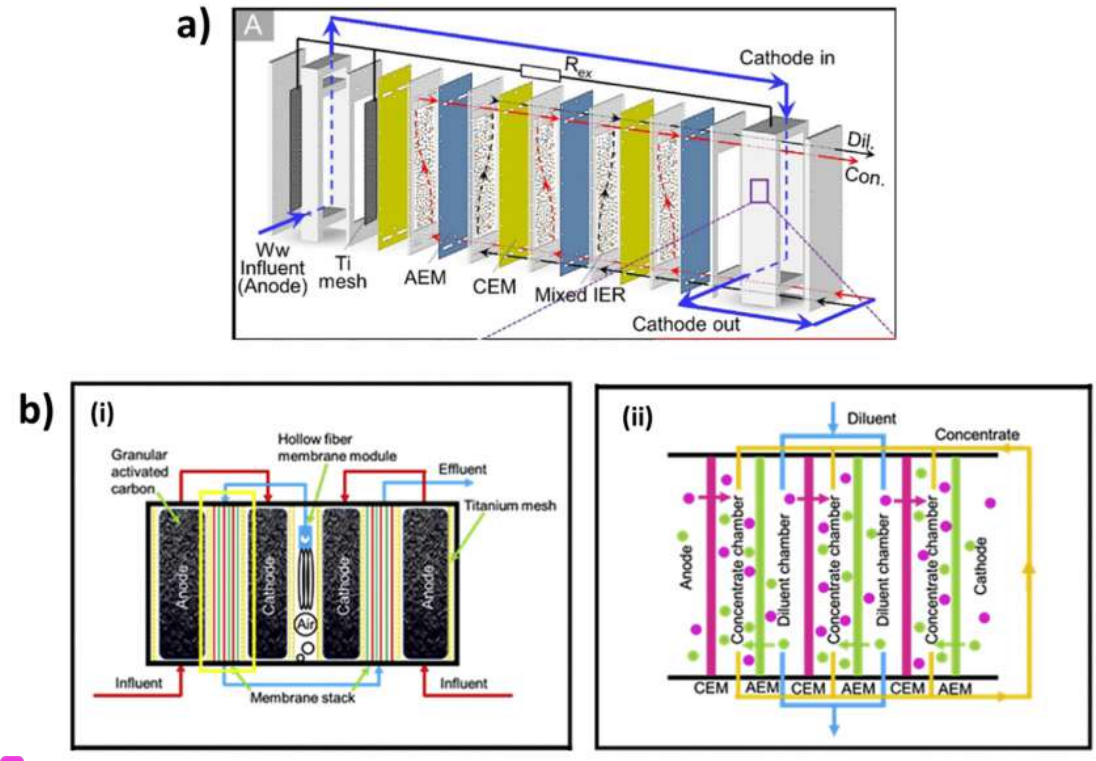


Fig. 8. a) HFM-MDC with ion-exchange resin, reprinted with permission from Zuo et al. (2018), b) DA-MDC with the integration of HFM, reprinted with permission from Liu et al. (2019c).

membrane stack (Fig. 8a) (Zuo et al., 2018). The cell configuration and operating system of the HFM-MDC are considered novel as the quality of the effluent can be improved because of the designated wastewater flow route and charge transport in the central membrane stack. Membrane fouling can also be alleviated because of the low organic content and aeration flushing in a cathode chamber. Furthermore, a different electrolyte type is unnecessary for a different chamber because wastewater is the only water source from which energy can be recovered to drive desalination. The system produces a cheaper and stable electrode than Pt-based cathodes by combining an HFM with a biocathode, and this system is collectively practical for the large-scale development of an HFM-MDC system.

Regardless of the MDC system used, treating real wastewater as a substrate can be challenging, commonly resulting in slower COD removal rates and lower maximum power density relative to simulated wastewater (e.g. acetate). A report has shown that the desalination performance decreases when real wastewater and seawater are treated with CMDCs (Jacobson et al., 2011b). However, these challenges are remarkably minimised using a dual-anode assembled MDC (DA-MDCs), where two-repeating-cell units are fabricated with two anodes and one cathode, which are arranged in parallel and separated by two IEM stacks (Fig. 8b) (Liu et al., 2019c). In the present study, the HFM module is placed within the cathode to filter out microbes and suspended solids. As a result, >95% of ammonia–nitrogen, total nitrogen, chemical oxygen demand and total phosphorous are removed using the modified real wastewater (Liu et al., 2019c).

Investigations on HRT are critical for MDCs, particularly in a continuous mode to achieve an optimised performance (Chen et al., 2017; Liu et al., 2019c; Zuo et al., 2018). A short HRT may result in insufficient duration for ionic transportation into the diluent chamber, thus lowering the desalination efficiency despite the incomplete treatment of organic oxidation, which does not meet the requirement for wastewater discharge. On the contrary, a long HRT hampers the capability of wastewater treatment. Liu et al. (2019b) found that a DA-MDC accomplishes 10 L day^{-1} treatment capacity and exceeds the performance of other reported biocathode-based MDCs for real wastewater treatment under the best HRT (6 h). Interestingly, the COD, total phosphorous and total nitrogen in the final effluent meet the minimum standard of wastewater effluent in China at all HRT (Liu et al., 2019b).

In addition to the excellent performance of HFM-MDC, significant parameters have not been investigated, leading to an equivocal path towards its technological practicality. The parameters that need to be optimised include the dimension of chambers, quantity of membrane pairs, concentration of organic matter, and rate of influent flow. These parameters should be studied in detail to maximise the performance by using various types of wastewater. Another critical issue is the highly concentrated brine disposed from the membrane stack, which is detrimental to the environment if it is directly discharged. A practical way to solve this issue is by directing the brine to MDC, electrodialysis, reverse osmosis or distillation for further concentrations. The system may involve a complicated maintenance procedure, considering the complex cell design of HFM-MDC, which adopts a plate-by-plate stacking. In addition, tuning the influent flow rate in one chamber may affect the HRT in different chambers because of the serial water flowing throughout the cell, thereby influencing the removal efficiency of nutrients, organic and salinity. Nevertheless, the HFM-MDC has realised over 95% removal efficiencies of COD, ammonia nitrogen, total nitrogen and total phosphorous in a single stacked cell. Hence, a techno-economic evaluation between the high-quality effluent and the capital investment for the technology is imperative to determine the feasibility of the system in real applications.

3. Assembled, coupled and scaled-up MDCs

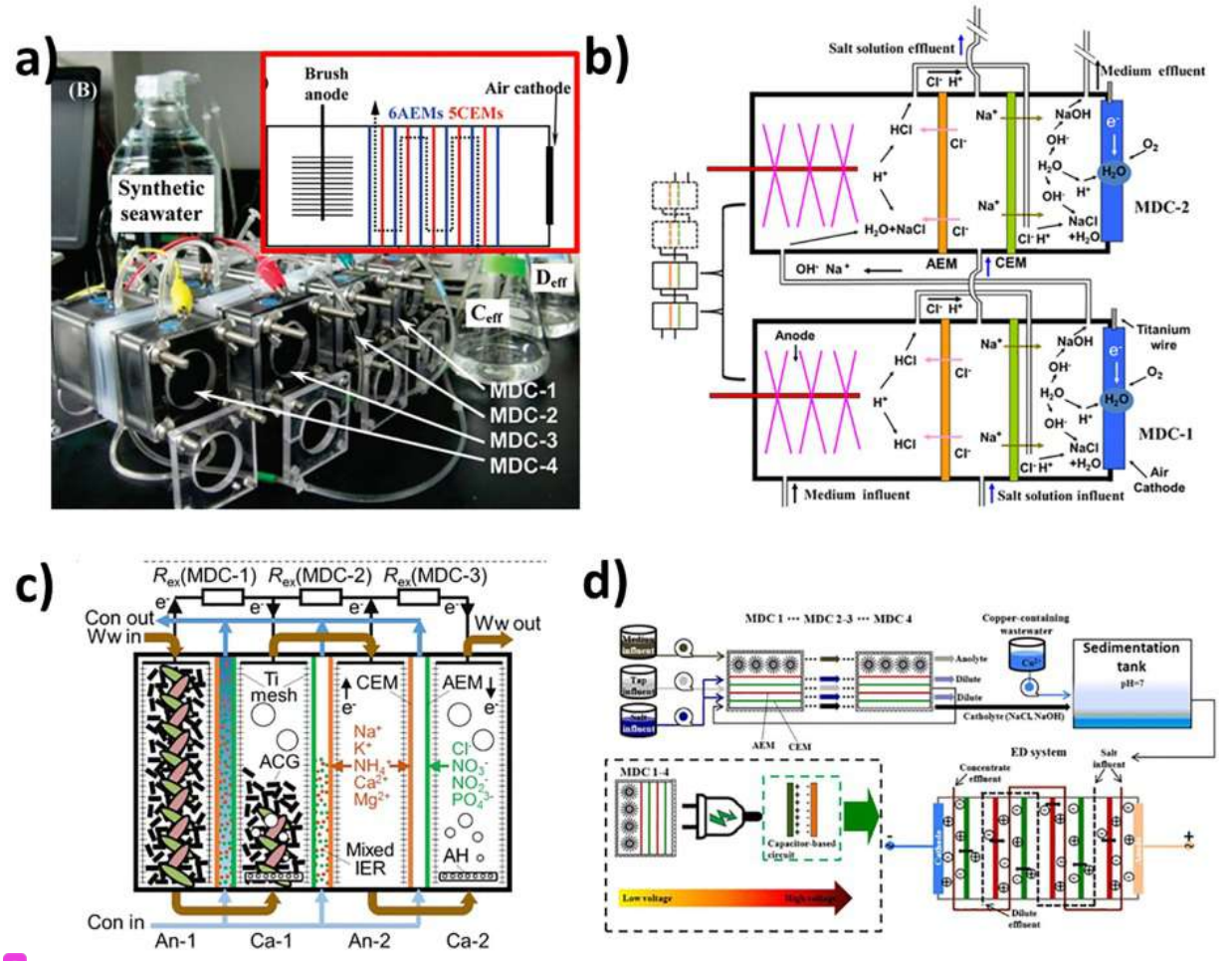
Wastewater treatment with a high COD removal, a high desalination rate and a highly efficient energy production has been proven

challenging in a CMDC. The issues have been reported as a result of a pH imbalance between anolytes and catholytes, severe membrane fouling, high internal resistance, high hydraulic retention time (HRT), incomplete wastewater and desalination treatment, along with a complex operation, which is attributed to the use of different solutions in different chambers. Concerning these issues, researchers attempted new approaches as alternatives to standalone MDC systems, including the assembly of identical MDC units and the coupling of MDCs with other existing technologies, such as FO, RO, ED and CDI. Additionally, few research attempts on MDCs with a large scale (greater than a litre) have been conducted as a platform to identify factors affecting the overall performance during the scaling-up. The progress of these recent research advancements, including the assembled, coupled, scaled-up MDC systems, is hereby reviewed.

3.1. Assembled MDCs

A partial and complete salt removal has been achieved by an assembly of MRECs connected in series by using an identical solution for desalination and cathode chambers, thereby avoiding additional costs for a different type of electrolytes for each chamber (Kim and Logan, 2011c). The quality of desalination effluent is improved through a series of assemblies of MRECs, where the diluate and the concentrate are hydraulically connected in series into the stacks of subsequent MRECs (Fig. 9a) (Kim and Logan, 2011c). The deployment of four stacked MDCs, which bear 20 pairs of desalination chambers, has resulted in a 44% reduction of 35 g L^{-1} NaCl with a comparable power density output as obtained in the standalone MFC without desalination chambers. The desalination is further enhanced to 98% when two additional stacked MRECs (3.8 L of anolytes in total) are added, producing 0.3 L of freshwater.

The configuration of the assembled MREC with one or more desalination chambers between an anode and a cathode can lead to a pH imbalance that may inhibit microbial growth in the anode (especially pH < 6) and potential losses due to an increase in pH (0.095 V per unit of pH) in the cathode, thereby reducing the whole MDC performance (Luo et al., 2010; Qu et al., 2012; Rozendal et al., 2008; Zhao et al., 2006). In addressing this issue, four CMDCs are hydraulically assembled in series where the effluent from an anode is pumped into the cathode chamber and mixed with a catholyte before it is channelled to the anode of the next CMDC cell, consequently helping in balancing the electrolyte solution between anodes and cathodes (Fig. 9b) (Qu et al., 2013). The saline solution from the desalination chamber also flows into each desalination chamber in sequence. A study has found that NaCl removal is HRT dependent, as indicated by an increase in percentage, i.e. 76 to 97% from 1 to 2 days of HRT; conversely, the COD



2 **Fig. 9.** Various assembled MDC systems, a) Series assembly of MREC, reprinted with permission from Kim and Logan (2011c), b) Series assembly of CMDC, reprinted with permission from Qu et al. (2013), c) Multi-stage MDC (MS-MDC), reprinted with permission from Zuo et al. (2016a), d) MREC connected in series and coupled with ED, reprinted with permission from Dong et al. (2017).

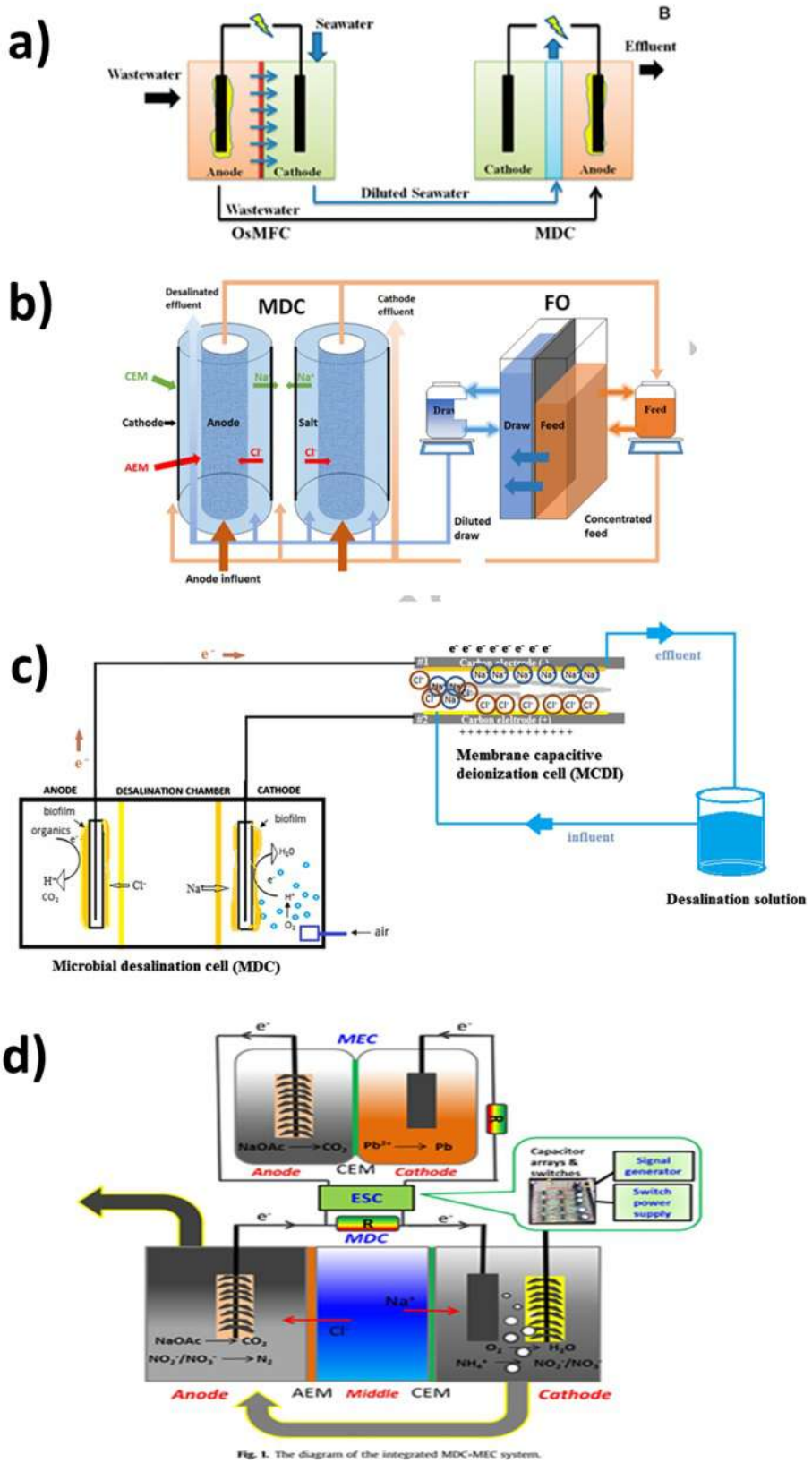


Fig. 1. The diagram of the integrated MDC-MEC system.

removal for both HRTs is not significantly different (~60%) (Qu et al., 2013).

In another research, the elimination of organic matter and nitrogen from real wastewater and the desalination process are simultaneously improved in a novel multistage MDC connected hydraulically in parallel (Zuo et al., 2016a). Investigations on three issues have been reported for CMDCs; i) existing cell designs reported in the literature are mostly ineffective in simultaneously removing organic waste and desalination, given that wastewater and saline solution are filled in a different chamber separated by membranes (Luo et al., 2012c; Zuo et al., 2013); (ii) a passive nitrification and denitrification reaction in MDC anode cells, causing incomplete nitrogen removal in the produced anode effluent (Chen et al., 2012c; Mehanna et al., 2010a; Stoll et al., 2015; Zhang and He, 2015); (iii) the presence of actual wastewater in a thin desalination chamber (millimeter scale), leading to membrane fouling, which is not the case for a pure saline solution as employed in a lab-scale experiment (Lindstrand et al., 2000). Therefore, a CMDC is designed to be operated in multistage to realise an improved concurrent treatment and desalination of wastewater (Zuo et al., 2016a). The multistage MDC (MS-MDC) is installed with two consecutive anode and cathode chambers stacked in parallel, separated by CEMs and AEMs facing the anode and the cathode, respectively, creating a thin desalination chamber. The real wastewater flows in a sequential order into 1st anode ➔ 1st cathode ➔ 2nd anode ➔ 2nd cathode; conversely, a concentrated saline solution is channelled into the desalination chamber (Fig. 9c). In such a flowing manner, a result of 11.4 mA, 52.4%, 92.5% and 87.0% is achieved for the current generation, desalination efficiency, effluent COD and total nitrogen removal, respectively, because of the presence of charge migration and effective nitrification/denitrification reaction (Zuo et al., 2016a).

A diluted high-strength industrial wastewater is treated using a similar design of MS-MDC and partially desalinated with a sequential flow: 1st anode ➔ 1st cathode ➔ 2nd anode ➔ 2nd cathode (Zuo et al., 2017). The initial wastewater influent contains 8723 ± 456 mg L^{-1} of COD and 24,612 ± 772 μS cm^{-1} of conductivity. The treatment has resulted in a remarkably low COD below 500 mg L^{-1} in the effluent, which complies with the minimum requirement for discharged wastewater, whereas 31.6% of the initial salt concentration was removed at 1.0 V (Zuo et al., 2017).

Hydraulically connecting a five-chamber MREC with the four other units of the MREC of the same design has accomplished the concurrent desalination process and the treatment of the synthetic copper-containing wastewater; this process is catalysed by the produced alkalinity (Fig. 9d) (Dong et al., 2017). A study has confirmed that 5000 mg L^{-1} of copper concentration is removed almost completely with a removal rate of 5.07 kg m^{-3} d^{-1}, leading to the effluent that is in accordance with the emission standard for electroplating in China. The harvested power from the MREC comprising four identical units is used to operate the conventional (ED) system as a post-desalination treatment for water recovery purposes by employing a capacitor as a charge collector (Dong et al., 2017). The MREC-powered ED produces the highest achievable desalination efficiency of 30.4%, which indicates a low performance and should be further improved.

Studies have presented that a promising COD removal, desalination and power generation can be achieved by hydraulically linking multiple units of identical systems in series or parallel connection. An assembled MDC system, which is connected in parallel, confirms that a simultaneous elimination of organic matter/nitrogen and desalination can be realised from the similar stream of domestic wastewater (Zuo et al., 2016a). This process provides an exemplary of the simplified operation, considering the utilisation of a single type of wastewater sources. The assembled system also offers a selection of operation modes for targeting an enhanced wastewater treatment or energy recovery. However, without a proper control at each compartment of the cell, the maintenance process may be laborious and challenging in dealing with a plate stacking design. The configuration of multiple MDCs connected hydraulically in series is simpler; thus, monitoring and maintaining an individual cell are more manageable.

3.2. Coupled MDCs

Individually, OsMFCs have been proven to recover water from wastewater (Zhang et al., 2011), and CMDCs have been tested for removing salt from a saline solution (Cao et al., 2009; Jacobson et al., 2011b; Qu et al., 2012). In OsMFCs, saline water, which plays a role as a draw solution and a catholyte, are subjected to low conductivity because of water migration from an anode chamber to a cathode chamber, a condition that can prevent the recirculation of a draw solution. OsMFC is incapable of eliminating salt from the draw solution, which can be channelled into the desalination chamber of CMDCs to remove salt driven by electricity generation (Fig. 10a). The anode effluent from OsMFCs can be transferred into a UMDC anode to further remove the organic content in wastewater through anaerobic digestion. Through coupling, the efficiency of desalination significantly increases because of dilution in OsMFCs and salt elimination in UMDCs despite the improved removal of organic matter, which is achieved sequentially in both systems (Zhang and He, 2013). The hybrid system succeeds a 95.9% reduction of the conductivity in the hybrid system and the energy production of 0.160 kWh m^{-3} with the treated saline water. Two main drawbacks have been identified by analysing the presented coupling system. Although COD removal and desalination efficiency are significantly improved in the hybrid system, the HRT of the saline solution takes 3.5 days (Zhang and He, 2013). Additional energy is also needed for aeration in the OsMFC, which is not practical in a large-scale system. In our perspective, the balance between the energy utilisation and treatment capability of both systems must be considered to produce excellent coupled MDC systems in real applications.

The coupling between the UMDC and FO is another attractive approach driven by its ability to treat wastewater and energy-efficient water. However, it is limited by an incomplete wastewater treatment or desalination in both systems, which are the major hindrance for further development, including the synergism between two technologies. A hybridisation effort is realised between UMDCs with an existing lab-scale FO cell to mitigate this problem (Yuan et al., 2015). Synthetic wastewater is inserted into the UMDC anode, and the produced effluent flows to the FO cell to further improve water recovery; conversely, the draw solution from the FO is channelled to UMDC for a desalination step (Fig. 10b). In comparison with the standalone MDC or FO system, the observation has emphasised that the COD removal and desalination are significantly improved in the coupled UMDC-FO system. Besides, the current generation is more stable, and the desalination rate is quicker in the coupled system than that produced in the standalone MDC. Moreover, the conductivity rate and COD removal rate perform proportionately at the shortened HRT by manipulating the HRT.

Another issue faced by CMDCs is the reduction of the conductivity of saltwater after a long desalination process, causing an increase in internal ohmic resistance and slowing down the desalination rate (Mehanna et al., 2010b). Although efforts have been devoted to addressing the problem, the documented inventions, such as the MREC and the addition of IER in a desalination chamber, have demonstrated a tedious and costly strategy. Adapting capacitive deionisation (CDI) onto IEM to produce membrane-CDI (MCDI) can be an alternative solution to the conventional technology of CDI. An investigation has been carried out to evaluate the desalination performance of two MDCs, which are

Fig. 10. Various coupled MDC systems, a) coupled OsMFC and MDC system, reprinted with permission from Zhang and He (2013), b) coupled UMDC and FO system, reprinted with permission from Yuan et al. (2015), c) coupled MDC and MCDI system, reprinted with permission from Wen et al. (2014), d) coupled MDC and MEC system, reprinted with permission from Li et al. (2017b).

connected in series or parallel to be used as the power supply of MCDI (Fig. 10c) (Wen et al., 2014). Another study has revealed that a parallel connection system produces the highest electrosorption capacity of 264.8 μmol/g, which equivalent to a 60% improvement as compared with that achieved using a potentiostat under a similar operational voltage. The result obtained corresponds to a desalination rate of 3.7 mg h^{-1} in one cycle (Wen et al., 2014).

In another approach, the coupling of a CMDC with an MEC has mediated a simultaneous function of nitrogen removal from domestic wastewater in anodes and cathodes, salt removal in a desalination chamber and the reduction of metals originating from industrial wastewater in the cathode of connected MEC systems (Fig. 10d) (Li et al., 2017b). These novel multipurpose reactions are achievable, given that nitrogen removal can be quickened by the presence of high-conductivity seawater. A biological reaction is manageable by flowing the cathode effluent containing an autotrophic denitrifier and its converted product of nitrate/nitrite into the anode chamber (Li et al., 2017b). The mixture of autotrophic and heterotrophic denitrification in the anode chamber leads to an enhanced efficiency of nitrogen removal (Li et al., 2017b). The nonspontaneous metal reduction carried out in an MEC is powered by MDCs through charge storage in capacitors. Notably, the problem associated with a pH imbalance in anodes and cathodes, as generally found in a CMDC reaction, is alleviated in the developed coupled system (Li et al., 2017b). Additionally, the electrical energy harvested by the capacitor has powered the MEC to reduce 99.5% of lead (II) from industrial wastewater in the cathode of MEC. The energy balance from the current hybrid system is positive, exhibiting an encouraging use of MDCs for multipurpose applications (Li et al., 2017b).

Coupling MDCs with other BESs or existing water reclamation technologies also enhances organic matter removal and water desalination compared with that of standalone MDCs or other BESs. This approach forms a mutual necessity between two distinct systems, complimenting each other's shortcoming. It can power the other connected system to simultaneously extend desalination or carry other cathodic reactions in the connected cell because of the ability of MDC producing electricity. In contrast to the multiple connections of identical MDC units, the coupling strategies require a greater control, considering the multitude of reactions that occur simultaneously in both systems. In a large-scale application, the magnitude of challenges and complexity of operation and maintenance is immense. The real challenge is to ensure that the targeted performance (e.g. COD removal and desalination efficiency) is greater than the standalone MDC after scaling-up.

3.3. Scaled-up MDC

The most critical aspect of the research and development of MDCs is the reactor scale-up, and studies have been performed to solve the fundamental and technical issues dealing with a laboratory scale. A large-scale MFC has failed to reproduce the high power density as obtained in a small-scale MFC (Clauwaert et al., 2008; Logan, 2010). Therefore, research on a large-scale MDC must explore the challenges and other limiting factors affecting the performance of MDCs during scaling-up.

In the early attempt of scaling-up, a litre scale of UMDCs is introduced to evaluate the performance and energy production in a slightly larger laboratory scale than previous ones (Fig. 11a) (Jacobson et al., 2011a,b). A study has indicated that HTR influences the removal rate of TDS even though the elimination of TDS in the salt solution is higher than that in artificial seawater (Jacobson et al., 2011b). A study has revealed that >70% of TDS is removed, which is predominantly affected by the current generation, despite other minor factors, such as water osmosis and an unidentified process. The UMDC is ideally operated under high-power generation instead of high current production because of the quantity of energy output. By contrast, high current production is more favourable for treating seawater desalination as a result of low salinity in the effluent (Jacobson et al., 2011b). Under the high-power output condition of UMDC set up as a pre-desalination tool for an RO system, the use of the salt solution and artificial seawater may account for 58% and 16.5% electrical production, which may contribute to operating a downstream RO purification system.

A large-scale stacked MDC (>10 L), which is loaded with a mixed IER, has been developed and conducted in a batch mode to desalinate 0.5 g L^{-1} NaCl, a concentration comparable with domestic wastewater (Fig. 11b) (Dong et al., 2017). After 12 h of operation, 0.5 g L^{-1} NaCl is reduced to 0.02 g L^{-1}, a quality of effluent almost equivalent to the allowable maximum salt concentration in the effluent produced out of RO filtration. The result corresponds to the desalination efficiency of 96%, which is benefited by the loading of IER and circulation flow rate of 80 mL min^{-1}.

The largest scale of the MDC system is 105 L working volume, which is conducted in a tubular form, i.e. the UMDC type (Fig. 11c) (Zhang and He, 2015). The use of the UMDC-105 L scale in this study has become the research platform to investigate the responsible factors affecting the performance of MDCs during a scale-up step. A thorough analysis of each module has indicated that the performance is severely non-uniform throughout the experiment. After an external voltage of 1.1 V is applied, the current generation is markedly enhanced by nearly three-fold compared with the electrical current recovered without an external voltage (670 mA). A current increment is also achieved by establishing multiple feeding points, providing an improved substrate distribution. As a result of the electrical current enhancement, the salt removal rate is also considerably increased from 3.7 kg m^{-3} day^{-1} to 9.2 kg m^{-3} day^{-1}. Another substantial observation from this study is the drop in the current generation because of a large COD loading, leading to the reduction of salt removal due to the growth of heterotrophic microorganisms on the cathode surface. Besides, approximately 40–60% of energy can be saved if catholyte recirculation can be reduced.

The development of large-scale MDC has been used as a steppingstone to investigate factors affecting MDCs during the scaling-up process. Investigations on large-scale UMDC have identified several parameters critical for the scaling up, and they include feeding strategy, organisation between wastewater treatment and desalination, circuit connection of multiple units and systematic energy estimation (Zhang and He, 2015). The number and configuration of desalination cells is also a key factor affecting the performance of large-scale MDC, which has been proved in previous study (Chen et al., 2016). However, large-scale MRECs loaded with IER (>10 L) have unsolved issues, including the incomplete removal of organic matter and bacteria, membrane fouling and ion exchange resin (IER), the uncertainty of the MDC performance with real wastewater and seawater, brine disposal from RED stacks and optimisation of stack dimension and quantity (Zuo et al., 2014). In the cell architecture for large-scale applications, the plate-by plate stacking design offers multiple reactions with a different mode of operations compared with that of two tubular cell designs, which are applied to UMDCs. However, the cost-effectivity of both cellular designs should be estimated to determine their practicality for real applications. In reality, the cost estimation of the development of a large-scale reactor may be inappropriate because of a considerable difference in material expenses and production costs between a high-grade lab-made goods and an industrial-produced system. Notwithstanding, software tools, such as RESYSpro and IPSEpro, can be used because they are proficient in carrying out economic, technical and ecological analyses (Karagiannis and Soldatos, 2008).

4. Comparative analysis on the performance of MDCs

A cross-comparison of the performance of MDCs has been difficult because of several constraints: 1) different cell configurations used, 2) various approaches for data collection and data reporting, 3) the employment of a broad range of methods for measurement and analysis and 4) lack of normalisation. These issues may negatively affect the development of MDC technology and diminish the impact on desalination market sector: operators, utilities and management. In view of the

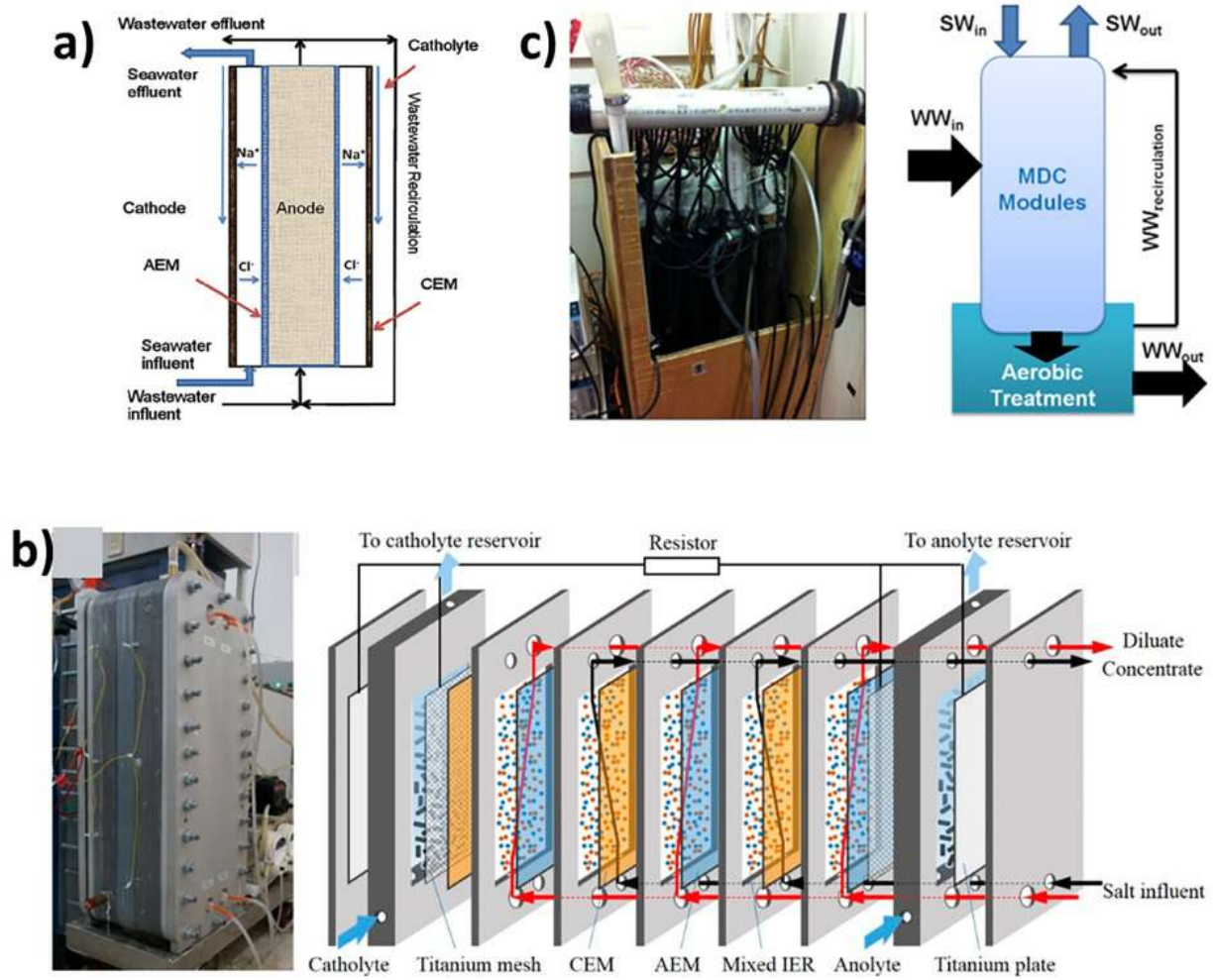


Fig. 11. Scaled-up MDCs design, a) 1 L UMDC, reprinted with permission from Jacobson et al. (2011b), b) 10 L of MREC, reprinted with permission from Zuo et al. (2014), c) 105 L UMDC of tubular form, reprinted with permission from Zhang and He (2015).

importance of evaluating the continuous advancement of MDCs, the present review provided a comparative analysis between the standalone MDC and the assembled MDCs and coupled MDC system and between the lab-scale system and the large-scale MDC system. The important parameters of the COD removal rate, nominal desalination rate (NDR), desalination rate and maximum energy output are included for comparison purposes (Table 2). The NDR parameter has been frequently overlooked in any MDC research even though this parameter is common in membrane technology, which can help implement MDCs at a real scale after development. In brief, NDR refers to the normalised amount of fresh water per square meter of membranes during every desalination cycle. Some parameters are well reported, so the present review carefully extracts the experimental data and normalised them into a comparable unit for each parameter.

Studies have demonstrated that an assembled MDC system can be an effective approach to enhance the COD removal, desalination rate and energy production. For instance, a series assembly of MREC has resulted in 3-, 3.2- and 1.4-fold enhancement against the standalone MREC in terms of NDR, desalination rate and maximum power generation, respectively (Kim and Logan, 2011c) (Table 2). Although the COD removal rate has not been well reported in the standalone MREC, about 91% of the COD removal efficiency is attainable after 12 h in an assembled system. The NDR value of the assembled system is also considerably higher than that of the single MREC, suggesting that the water transport is greater in the assembled system. This condition reduces the water quality and recovery in the diluate, which is obtained from electroosmosis and osmosis (Kim and Logan, 2011c). Three stages of four MREC modules are sufficient to achieve 97.6% desalination to extend this process. Interestingly, the power generation is comparable with those obtained in MFCs during desalination in MRECs.

Conversely, the assembled hydraulically connected CMDC (four units) does not show an enhancement effect compared with that on a single CMDC (Qu et al., 2013). The COD removal rate, desalination rate and power generation decrease as the effluent from the anode and desalination chamber flow downstream. Despite the reduction of COD removal rate in downstream CMDC as compared to the first CMDC, the total COD removal efficiency did not improve significantly (60 ± 2%). Conversely, the reduction of desalination rates in the downstream CMDCs is attributed to the diminished salt content in the desalination chamber, causing an increase in internal resistance; thus, potential losses are large (Qu et al., 2013). The low power generation in multiple CMDCs (four units) is likely due to a reduced substrate concentration in the downstream MCDCs, which can affect the viability of EAB, hence, the whole performance. The discrepancy between MRECs and CMDCs with

Table 2
Comparative performance of selected standalone, assembled, coupled, and scaled-up MDCs system.

System	COD removal ($kg\ COD \cdot m^{-3} \cdot d^{-1}$)	NDR[a]; Desal.[b] ($L \cdot m^{-2} \cdot h^{-1}$); ($g \cdot L^{-1} \cdot d^{-1}$)	Max. energy density ($W\ m^{-3}$)[@]	COD removal ($kg\ COD \cdot m^{-3} \cdot d^{-1}$)	NDR; Desal. ($L \cdot m^{-2} \cdot h^{-1}$); ($g \cdot L^{-1} \cdot d^{-1}$)	Max. energy density ($W\ m^{-3}$)[@]	Ref
A single unit of MDC				Assembled MDCs			
Stacked MREC	N/A	0.002; 9.88*** (Vol. of diluate ↓, vol of conc.↑)	18.66	N/A (91% COD reduction)	0.006; 31.25*** (Vol of diluate ↓, vol of conc. ↑)	26.66	(Kim and Logan, 2011c)
Stacked CMDC	~1.6	N/A; 5.8*	21.5	~1.0	N/A; 0.8*	17.8	(Qu et al., 2013)
System	A single unit of MDC			Coupled MDC			
OsMFC + UMDC[#]	0.7	N/A; 4.05***	27.81	6.2	N/A; 9.66***	53.33	(Zhang and He, 2013)
UMDC + FO	0.4	N/A; 11.2***	0.056	0.42	1.68; 25.5***	0.064	(Yuan et al., 2015)
System	0.85 L-scale (0.85 L)			Larger scale (2.75 L)			
UMDC	N/A	N/A; 7.50** (Jacobson et al., 2011a)	30.8 (Jacobson et al., 2011a)	6.78	0.01; 11.61*** Closed circuit 0.006; 9.9 Open circuit	28.9	(Jacobson et al., 2011b)
UMDC	Refer to UMDC-2.75 L (Jacobson et al., 2011a)			Larger scale (105 L) 1.44	N/A; 9.2***	36.6 (at 1.1 V)	(Zhang and He, 2015)

[a]NDR: Norminal desalination rate, [b]Desal.: desalination rate, [#]: total volume (1.95 L), *Initial conc. of NaCl: *20 g L^{-1}; **30 g L^{-1}; ***35 g L^{-1}, N/A: data not available, [@]: volumetric power density.

regard to the performance in the assembled system is attributed to the effect of salinity-gradient power in MRECs despite the bioelectrochemical oxidation in the anode chamber. The comparison of both systems reveals that the desalination and energy production of MRECs are better than those of CMDCs, but the cost of the former is higher than that of the latter because of the installation of membrane pairs. Therefore, the use of a cost-effective membrane in MRECs is critical.

The comparative analysis of two coupled MDC systems, i.e. FO-MFC + UMDC (Zhang and He, 2013) and UMDC + FO (Yuan et al., 2015), against a single MDC of a similar coupled system is not easy mainly because of a different system orientation and the lack of normalisation. The performance of both coupled systems is higher in terms of the COD removal rate and desalination rate than that of the standalone UMDC system (Table 2). A cross-comparison between two different coupled systems (OsMFC + UMDC and UMDC + FO) concerning the desalination rate infers that the UMDC + FO coupled system is more robust. Yuan et al. (2015) reported that the coupled system of UMDC + FO produces a more stable current and a faster desalination process than the standalone UMDC does. Furthermore, a clean water recovery and a lesser wastewater volume are feasible with UMDC + FO. Contrastingly, the COD removal rate and power density of both coupled systems differs, i.e. it is higher in OsMFC + UMDC than in UMDC + FO. This observation can be explained, given that OsMFCs and UMDCs have anodic microbial oxidation in both individual systems. It is noted that the NDR was not consistently reported, making evaluation for both systems unfeasible.

To date, UMDC has been progressively researched for an up-scaling study from a millilitre or several litres of up to >100 L scale (Jacobson et al., 2011a,b; Zhang and He, 2015). These studies have been selected on the basis of the reported works that are the products of similar main scholars and the research concepts and operational systems that are maintained.

UMDC has been scaled up through a stepwise increment. For comparison purposes, a detailed review on the UMDC design at each stage is provided. The introductory design of the UMDC presents a tubular reactor design, which comprises two tubular compartments separated by an IEM. The inner tubular compartment is created by a rolled AEM, which contains with graphite granules and graphite rods as an anode and a current collector, respectively. The dimension of the tubular membrane is 6.15 cm diameter and 40 cm length to contain 500 mL of anolytes. The outer tubular compartment is a slightly larger roll of a CEM with 7 cm diameter and 40 cm length to accommodate 350 mL of saline solution. The outer surface of CEM is coated with Pt/C (0.2 mg Pt cm^{-2}) ink in a slurry form, which is then covered with two layers of a carbon cloth that functions as a cathode. The connection between a cathode and an external electric circuit is completed with Pt wire (Jacobson et al., 2011a). In a large-scale UMDC, graphite granules in the anode chamber are replaced with carbon brushes as the anode (Jacobson et al., 2011b). The diameter of tubular AEM and CEM is maintained, but the length of both tubular membranes is extended to 70 cm, resulting in a total volume of 2.75 L (1.90 L of anolyte excluding carbon brushes and 0.85 L of saline solution). The platinum loading coated on a carbon cloth is increased to 0.4 mg Pt cm^{-2}. To date, the largest UMDC is scaled-up to an operating volume of 105 L (Zhang and He, 2015). Under a similar reactor setup, the dimension of tubular AEMs and CEMs shrinks to 5 and 5.6 cm, respectively, and the length of both tubular membranes is further elongated to 86 cm. The anode and desalination chamber are filled with 2 L of anolytes and 0.5 L of saline solution, including liquid in the connecting parts. A 1 m length carbon brush anode is inserted into the AEM tube, whereas the cathode is the carbon cloth coated with activated carbon powder (5 mg cm^{-2}) without platinum. The cathode is wrapped with a titanium wire around the tubular CEM.

Although maintaining the performance of MDCs during scaling-up is challenging, a stepwise volumetric increase in the total cell volume from 850 mL to 2.75 L has manifested the possibility (Jacobson et al., 2011a, b). Although the COD removal between UMDC-0.85 L and UMDC-2.75 L is incomparable because of the unavailability of data in a small scale, the desalination rate is better in the large-scale and comparable power output between two scales (Table 2). The enhanced TDS removal rate is associated with the advantage of having a larger ratio between the salt solution and wastewater volume (Jacobson et al., 2011a). Although the accurate explanation has been widely explored, a study linking the enhancement to the low salt accumulation in the anode in response to a higher flux of the anolyte. A high flux ensures an adequate organic supply for microbial electron generation and facilitates ion exchange because of the large membrane surface (Jacobson et al., 2011b). The NDR under a closed-circuit condition is lower than that under an open circuit condition, suggesting that the current generation can drive the TDS removal in the UMDC. At a large scale containing 30 UMDC modules with a total working volume of 105 L, the COD removal rate noticeably decreases compared with that in UMDC-2.75 L. Although multiple feeding points have resulted in a high current generation because of the improved mass transfer, the COD removal does not increase accordingly because of the complex condition of UMDC-bearing

multiple units. The desalination rate in a highly complex system of UMDC-105 L is also unexpectedly lower than that in the UMDC-2.75 L even after the external power of 1.1 V is supplied. This problem may be stemmed from a highly nonuniform performance in each module (Zhang and He, 2015). Relative to UMDC-2.75 L, the power density is significantly higher in UMDC-105 L contributed by the potential input at 1.1 V. Therefore, future work should focus on the optimisation between energy requirement and MDC performance.

A fair judgement on which of the selected system works the best is not easy, considering the different configurations, dimensions and orientations of cells or reactors. However, based on the normalisation of data carried out in the present review, the highest performance for COD removal rate, desalination rate, and power density was realised by the UMDC-2.75 L, assembled MREC, and coupled OsMFC + UMDC system, respectively. In view of up-scaling strategy, it is important to ensure that the maximum performance attainable in a lab-scale-size reactor can be maintained or performed better. In another issue, we recommend a standardised COD removal rate (kg COD m^{-3} d^{-1}), a nominal desalination rate (L m^{-2} h^{-1}), a desalination rate (g L^{-1} d^{-1}) and a power density (W m^{-3}) in response to the lack of normalisation in presenting important data, which are more practical for the comparison between systems towards industrialisation. Although the present analysis is conducted in a small sample of reported studies, the comparative evaluation between MDC systems may provide substantial points in decision-making for future studies.

5. Prevailing challenges and potential mitigation strategies for future practicality

The development of MDCs has been concentrated on a laboratory-scale design. A large-scale research should be performed to evaluate the strength and compatibility of materials used to construct a large reactor. The factors responsible for the failure or unsatisfactory output from a scaled-up reactor should be recognised. In dealing with real wastewater and seawater during field testing, the developed MDC system is exposed to an unpredictable outcome, requiring ways for the pre-treatment of wastewater and saline solutions. Several ways to alleviate issues related to low conductivity, unstable electrode, membrane fouling, negative net energy balance and capital cost are another concern. Besides, a great attention should be given to investigate the durability of the system over a long operation period while treating the diverse nature of wastewater at varied temperatures because it influences the maintenance cost on membrane biofouling. An excellent performance of a lab-scale cell/reactor not necessarily reproducible in a large scale system mainly because of the poor substrate, ionic and charge transfer. Before MDCs are used in real applications, the system must be capable of generating a high power density and fuel production to provide a sustainable energy source. Although few research efforts have devoted to scaling up, a promising result has yet to be demonstrated. Hence, this section presents the prevailing challenges that emerge from a lab-scale MDC system and potential mitigation strategies based on large-scale specifications.

5.1. Reactor configuration, challenges and solutions

Reactors designed for a millimeter-scale system are partially adaptable for the construction of a litre-scale reactor mainly due to the incongruent mass and charge transfer between the two scales. The factors that have a close association with mass and charge transfer include the reactor design, electrode configuration, electrical connection between electrodes and selection of membrane and granular resin.

The largest MDC system is a tubular design made of an AEM tube inside of a CEM tube, forming a double cylinder (Zhang and He, 2015). As an assimilation from MFCs (Zhuang et al., 2012), a large-scale of an MDC tubular design can be materialized by extending the length of the tube, where additional tubular modules are connected in series, forming an MDC stack, which can be operated under a continuous flow mode. Studies have suggested that the tubular design can preserve its optimal cross-sectional dimension during scaling up (Kim et al., 2010, 2011; Scott et al., 2007). A tubular design also results in a minimal dead space, which encourages a near plug-flow regime, promoting a steady-state flow condition (Kim et al., 2011). The tubular design also provides a large surface area, which is beneficial to the efficient salt removal from the middle chamber.

Despite the practicability of tubular designs for scaling-up, the fabrication of new lab-scale MDC designs has been focused on plate-by-plate stacking forming a cell. The circumstances may be due to the large expense on costly large membranes needed to build a tubular MDC and regular membrane replacement. The favourable feature having a flat plate design for MDCs is that the stacking with multiple modules is feasible and simpler, requiring a smaller membrane size and a lesser membrane supporter than that a tubular design. In addition to wastewater treatment and desalination, various reactions can occur in the cathode chamber at once other than OER, such as fuel generation, pollutant degradation or chemical production with the additional chamber.

Desalination efficiency is influenced not only by the potential difference generated in the anode and cathode but also by the ratio of reactor dimension (Ebrahimi et al. (2018). A coefficient value (α) as a result of the ratio between the volume of the anolyte and the desalinated water is referred as a measure to evaluate the desalination efficiency and the salinity reduction. For clarity, the lower the α value, the higher the desalination efficiency; thus, it is more economical in terms of anolyte consumption. For that reason, a quadripartite MDC is developed, emphasising the ability to minimise the anolyte consumption over saline water. Four sets of desalination and cathode chambers are combined to form a single anode chamber (Fig. 2d). Although the cell installation is complex, this approach can reduce the construction cost because of the minimised size of an anode chamber.

In terms of electrical connection, several BES reactors can be electrically connected in either parallel or series (Aelterman et al., 2006). However, a series connection can induce a short-circuit effect with adjacent reactors because of liquid connection (Jacobson et al., 2011a,b). A parallel connection with multiple BES reactor units has proven to achieve a long-term and stable power generation without any electrical failure (Kim et al., 2020). In another example, the connection of 30 UMDC modules is achieved through a parallel connection of different types: a combined circuit and an individual cell. In the combined connection, all anodes (cathodes) are combined as a single circuit cell, whereas each module is connected to the resistor between an anode and a cathode in an individual cell, producing 30 circuit cells. However, both electrical connections do not cause significant changes in the COD removal and desalination rates (Zhang and He, 2015). Additionally, the generated current via an individual connection is significantly higher than that via a combined connection, which is postulated due to intercharge amongst the anode of multiple modules (Zhang and He, 2015).

Studies have demonstrated that the use of brush anodes in a millilitre-scale reactor design can generate high power density, which is attributed to a large electrode surface area (Hays et al., 2011; Hutchinson et al., 2011; Logan et al., 2007). However, design factors, such as diameter, brush length, configuration, orientation and quantity of brushes, should be optimised to minimise the internal resistance and improve power density. Moreover, the architectural design applied to construct a reactor should provide the system with an enhanced mass transfer from the electrode surface to electrolytes and vice versa. Studies have reported that membranes, which also act as separators, also deform after a long period of operation in either small or scaled up MFC (Dekker et al., 2009; Zhang et al., 2010). A similar event can occur in the MDC system. Membrane deformation can reduce the volume of the anode chamber and cause gas trapping, which degrades performance. Therefore, the fabrication of a supporting structure is necessary to maintain the separator integrity while increasing the maximum surface area attainable for the electrodes.

The selection of membranes and the number of membrane pairs affect the reactor construction cost. However, an offset is observed between high power density and treatment capacity even with the use of an expensive membrane. Fundamentally, an optimised reactor design that generates high power density is the basis for yielding a high treatment efficiency, given that a higher current generation leads to a higher COD removal. In particular, MDC designs, such as MRECs, have a considerably high internal resistance because of the installation of RED stack cells between anodes and cathodes, separating the electrode further away. Hence, the number of membrane pairs and their distance in the RED stack cells should be optimised to balance between internal ohmic resistance and desalination efficiency and push the MREC into a litre-scale application. High internal resistance may also source from the use of a nonconductive granular material (Zuo et al., 2016a). Hence, the use of a conductive granular material (e.g. resin) is recommended to improve wastewater treatment in the anode chamber. The most widely used conductive granular in BESs is granular activated carbon (GAC) because it is an inexpensive material. GAC has a highly porous adsorbent and conductive properties because of heat treatment during its production. Stuffing a reactor with GAC improves the electron transfer from suspended EAB to circuit during substrate oxidation. Aelterman et al. (2008) suggested that the utilisation of 3-D structures, such as GAC, increases the anode surface area, which helps reduce internal resistance, consequently improving electricity generation. MFC studies have shown that the use of GAC reduces the internal resistance from 84 Ω to 17 Ω (He et al., 2006), and increases the columbic efficiency from 33.7% to 45% (Wang et al., 2010).

Connecting multiple identical units and coupling CMDCs with existing conventional technologies, such as RO, FO and ED, are considered the most practical strategies to expedite MDCs into practicality. In hydraulically connected MDC modules, wastewater flows in series and assists in alleviating the pH imbalance issue and improving the desalination rate as compared with that in standalone CMDC (Qu et al., 2013). In another advancement, a significant COD removal from a high-strength industrial wastewater is accomplished using MDCs stacked in parallel comprising a repeating set of an anode and a cathode, i.e. anode 1 ➔ cathode 1 ➔ anode 2 ➔ cathode 2 (Zuo et al., 2017). The high COD removal is ascribed to the alternate reaction under anaerobic/aerobic conditions.

In summary, if the main goal of a large MDC system is for wastewater treatment, desalination and power output, then a tubular design is suitable for this function, but it is compromised because of the utilisation of a large membrane. Conversely, if the main aim is to provide another reductive reaction in the cathode beside ORR (e.g. hydrogen and CO_2 reduction), then the plate design reactor is more appropriate. A systematic study between multiple reactors of tubular and plate designs should be carried out to determine the lowest construction cost and the highest MDC performance. A comparative study should consider various factors, including reactor orientation, electrode configuration, electrical connection between electrodes, membrane selection and ratio between anodes and desalination chambers, to make the study more practical.

5.2. Overcoming issues associated with a low conductivity and an unstable electrode

One large obstacle in BESs is the low power generation because of potential losses during electron transportation between electrodes (Rabaey and Verstraete, 2005). This issue is commonly associated with the selection of electrode materials (Li et al., 2018a; Zhao et al., 2015). Low-cost carbon-based materials, such as carbon paper, carbon felt and graphite rods, have been widely employed in studies on MDCs mainly because of high stability and low cost (Pant et al., 2010b). However, these materials exhibit certain inherent drawbacks, especially the relatively low surface active area for biofilm growth and low substrate mobility, thereby a low power generation in MDCs (Wang et al., 2013).

Many studies have geared towards the development of 3D porous electrodes for anode materials to significantly increase the power output in MDCs (Flexer et al., 2013; Hou et al., 2014a,b; Yuan et al., 2018). A 3D porous electrode can provide a large surface area assisted by the existence of abundant macrochannels and pores for quick mass transport pathways, allowing low charge transfer resistance, facile substrate transportation and high charge storage capacity for enhancing electrochemical reactions. Studies on nanocatalyst-loaded 3D macroporous materials, including carbon nanotube-coated sponges, have also been actively explored to improve the performance of MFCs (Xie et al., 2012), Pt-loaded graphene for 3D electrode development (Zhao et al., 2015), nanoparticle TiO_2-coated loofah sponge carbon-based electrodes (Tang et al., 2015) and graphene foam (Yang et al., 2015). However, studies have yet to investigate the influence of these modified materials as electrodes in the MDC system.

In addition to bioanodes, membranes and cell chambers, a cathode is an essential component of the MDC system. Although platinum is the best catalyst for accepting electrons (e.g. oxygen reduction or hydrogen generation), its high cost prevents it from being used on a large scale. In the search for materials that exhibit comparable efficient catalytic properties as Pt at a low cost, nickel alloys or stainless steel is the best alternative. In hydrogen production, the gas diffusion type cathode fabricated through a single nickel chemical deposition process is capable of generating hydrogen (Hrapovic et al., 2010). Other researchers also demonstrated that tungsten carbide (Harnisch et al., 2009), molybdenum disulfide (Tokash and Logan, 2011), and Ni alloys (Manuel et al., 2010) are potential candidates for HER catalyst.

Oxygen as a terminal electron acceptor in the cathode of MDCs has been widely applied in several studies because of the insignificant toxic effect of atmospheric oxygen and the safe end product of water (Alvarez-Gallego et al., 2012). However, the use of an air cathode is problematic because of a passive redox kinetic at ambient pressure and high energy consumption for operating mechanical aeration that is used to maintain sufficient dissolved oxygen levels in catholytes. The limitation that is typically observed in the MFC air cathode may equally affect MDC systems. Logan (2010) proposed several strategies to improve the MDC performance. The proposed methods include 1) exposing one side of the air cathode to the atmosphere to capture the optimum dissolved oxygen in the cathode and 2) the employment of activated carbon with a high surface area to enhance the ORR eliminating the need for a precious co-catalyst (Biffinger et al., 2007; Freguia et al., 2008).

Nanotechnology application has created a new branch of research. For instance, electrodeposited palladium nanoparticles and nickel-based nanomaterials as hydrogen catalysts have been successfully demonstrated. Stainless steel, nickel foam and MoS_2 have been frequently used in MEC systems, which can also be adapted for MDC systems. These materials are used because of their accessibility, inexpensive and comparable catalytic performance compared with those of platinum. This review suggests that stainless steel may be a good material in terms of structural strength, durability and facile route to large-scale production. However, more research should be performed to improve the conductivity of stainless steel.

Alternatively, a biocathode can be used to replace a noble metal as a cathode catalyst because of its simple preparation, low operating cost, no need for a metal catalyst or a chemical electron mediator, excellent stability and versatile for the production of a valuable compound. However, similar to the developed cathode catalysts, the efficiency of a biocathode on a large scale has never been tested. Therefore, more tolerant and efficient biofilms should be studied for large-scale applications. Depending on the form of a terminal electron acceptor used in the cathode chamber, a biocathode is categorised into two types, i.e. aerobic and anaerobic cathodes. For an aerobic biocathode, oxygen is often used as an oxidant because of a high redox potential and accessible, thus low cost. Although aerobic biocathodes have gained full attention over the last few years (Jafary et al., 2015, 2019), the main problem with

this approach is the level of dissolved oxygen needed, as the lack of dissolved oxygen impedes the performance of the system. In response to the constraints mentioned above, current research progress has been directed to the use of an anaerobic biocathode. However, initiating an anaerobic condition for biofilm growth is more complex and time consuming (Butler et al., 2010; Morita et al., 2011). Specific electrotrophs have been used to represent the microorganisms responsible for directly or indirectly receiving electrons from a cathode. A biocathode can use electron acceptors, such as manganese, sulfate, fumarate, nitrate, arsenate, iron and carbon dioxide (Clauwaert et al., 2007; Cournet et al., 2010). Interestingly, the use of carbon dioxide as an electron acceptor for algal-based biocathode can be a promising strategy to reduce the carbon toxicity effect in the atmosphere.

The typical development of new electrodes is initiated with a small dimension to avoid a huge material cost in a large scale. As the dimension of a reactor increases, the size of electrodes should increase accordingly. However, a high current generation of lab-scale electrodes does not increase proportionately with the electrode size. Hankin et al. (2017) hypothesised that the effects of electrode configuration and geometry in an electrochemical reactor influence the performance of electrodes with 0.1 scale range and larger. The authors also suggested that the current dissemination in large electrodes may be non-uniform because of potential losses in a current dissemination layer. Despite the use of a highly conductive substrate, a 3D porous structure, a nanocatalyst or a biocathode, more electrical contact points should be provided on an electrode surface to ensure an adequate charge transport throughout a substrate over a long distance. In relation to electrode stability, a biocathode may be a promising approach for an economic system in the future. However, real data from a large-scale MDC system are yet to be demonstrated.

5.3. Issues related to membrane fouling and solutions

An ion exchange membrane is one of the main components in the MDC system as it creates different chambers to accommodate different reactions and functions and allow the transport of specific ions. The MDC activity is closely influenced by membrane conditions (desalination and wastewater treatment processes). The primary issue with membranes in BESs is fouling. Fouling is more drastic when actual wastewater and seawater are used because of more complex compositions than simulated wastewater and saline solution. Two fouling types are identified: biofouling and scaling. Membrane biofouling takes place when a membrane is in contact with organic matter and microorganisms present in real wastewater (Vrijenhoek et al., 2001) and transparent exopolymer particles from seawater. Both substances can initiate the formation of biofilms (Yiantsios and Karabelas, 1998). Meanwhile, the deposition of inorganic materials can form scaling.

An investigation related to long-term effects (8 months) on membrane fouling in a MDC has found that scaling in CEMs is not noticeable. Furthermore, biofouling formed at an AEM increases the MDC resistance, thus reducing both current generation and desalination efficiency (Luo et al., 2012b). However, the finding is impractical for real applications because the desalination chamber is filled with the simulated seawater of NaCl and $NaHCO_3$. Conversely, another study has shown that the resistance of CEMs because inorganic scaling increases to a greater degree than AEM does, suggesting that CEMs require more maintenance during the MDC operation (Ping et al., 2013). Another study has shown that the existence of cations such as Ca^{2+} and Mg^{2+} can substantially reduce the desalination efficiency and power generation, whereas anions such as Br^- and SO_4^{2-} have no significant effect on system performance (Luo et al., 2012a). Contrary to membrane scaling, biofouling on AEMs does not show any radical effect on the MDC activity (Luo et al., 2012a). In another observation, a reduction of Coulombic efficiency, salt removal rate and power density are attributed to biofouling on CEMs affecting aerobic biocathode MDCs after 5500 h of operation (Zhang et al., 2016). A study has suggested that an effective way to restore the MDC performance is to replace membranes (Zhang et al., 2016), which inevitably require additional cost for membrane replacement.

Unlike biofouling on IEMs, biofouling on an FO membrane has been reported to improve the generation of electricity by 50% after the addition of a small amount of HCL for buffering purposes, but it does not affect the water flux (Ge and He, 2012). In another set of experiment, the fouled FO membrane diminishes the water flux function but improves the current generation (Ge and He, 2012). However, an attempt to do membrane backwash by using NaCl (0.2–0.5 M) does not significantly overcome biofouling membranes (Ge and He, 2012). The current enhancement is attributed to fouling-assisted enhanced ion diffusion and ion exchange/neutralisation mechanism, which is discovered in a MFC system (Zhu et al., 2016). It is also applicable to explain a similar event in MDC. Hence, FO membranes can be a potential fouling-tolerant separator for BES technologies, including MDCs.

Fouling is a significant weakness that hinders the use of membranes for industrial scale. However, if a fouling phenomenon is deeply investigated and understood with appropriate prevention and control, the formation of fouling can be minimised or removed completely. For this purpose, the structure and fouling composition should be carefully observed, and fouling on the MDC performance should be investigated. Various approaches have been suggested to control or prevent fouling. The current trend of control and prevention has been devoted to the invention of membrane materials integrated with antifouling properties (Mulyati et al., 2012, 2013; Vaselbehagh et al., 2014). However, antifouling-integrated membrane only target specific fouling types, which are impractical to prevent numerous types of fouling from a complex solution. Hence, more research should focus the efficacy of existing modified membranes and modified IEMs, which are resistant to fouling having various properties. The physicochemical characteristics of different fouling types and their mixtures are also another key factors in understanding the underlying mechanism of fouling.

A conventional technique of pressure-driven processes as a pretreatment technique is potential because it avoids the contact of fouling agents on IEMs (Nataraj et al., 2007; Shi et al., 2014; Yang and Yang, 2004), but this technique incurs additional cost and reduces profit margin. Alternatively, methods such as overlimiting current (OC) (Bukhovets et al., 2011; Nikonenko et al., 2014) and pulse electric field (PEF) (Malek et al., 2013; Nikonenko et al., 2010) may be able to solve the fouling problem soon because it does not require additional investment and suitable to prevent and control various fouling types. Practically, the electrical input can be harvested from the generated power output from MDCs. Furthermore, OC and PEF can reduce the concentration of polarisation, which is another major problem, leading to an increase in the processing cost. However, OC is immature in terms of the mechanical feasibility against fouling, and PEF is more established and offers an in situ cleaning for membrane fouling having different natures, in addition to minimising concentration polarisation and simple installation. Although a PEF has been successfully carried out on a laboratory scale, the abovementioned advantages are also promising for industrial scale.

The in situ electrolytic cleaning of membrane modules by using electrically conductive feed spacers is an encouraging solution for field applications. Studies on feed spacers of membrane modules are necessary to optimise the distance that separates adjacent membrane layers. Many investigations have been carried out through computer modelling at a lab scale (Bucs et al., 2014, 2015; Fimbres-Weihs and Wiley, 2010; Siddiqui et al., 2017). Additionally, state-of-the-art feed spacers with new features are an attractive option for reducing fouling in modules by enhancing the shear forces of the membrane surface. The main challenge is to advance the fabrication of a conductive membrane through feed spacer optimisation so that membrane cleaning by using the PEF technique can be realised for an industrial scale with minimal power utilisation.

5.4. Challenges and solutions for actual wastewater and seawater treatment

The use of simulated wastewater (e.g. acetate) and seawater (e.g. NaCl) in a laboratory cannot represent the exact performance of MDCs during a field application. Besides, a cross-comparison on the reported performance of MDCs is difficult because the system performance is often affected by a variety of factors that generally have a different experimental setup where accurate information is usually lacking. In contrast to acetate-based substrates designed to simulate real wastewater, actual wastewater with an unknown composition can come from different sources, such as dewatered sludge, sewage sludge, domestic wastewater, industrial wastewater, leachate and engine oil. The realisation of MDCs for field applications is exceptionally challenging because of the complex nature of these wastewater, high concentrations and the existence of recalcitrant compounds. A different wastewater source also leads to the varied MDC performance amongst the reported studies with regard to COD removal, hydraulic retention time and power generation.

Although the different concentrations of NaCl are studied to simulate seawater in most of the lab-scale MDC systems, the inherent composition of real seawater contains more than Na^+ and Cl^-, which include magnesium (Mg^{2+}), sulfate (SO_2^{4-}), potassium (K^+) and calcium (Ca^{2+}). These ions make up about 99% of all sea salts (Castro and Huber, 1992). Competitive ion transportation across AEMs and CEMs during desalination is expected, bringing more fouling and scaling issues to the membrane. For all types of MDCs, the use of real wastewater as a substrate can remarkably reduce the COD removal rate and maximum power density compared with that of the systems by using acetate. Moreover, an OsMFC system operated with actual wastewater and seawater exhibits a decrease in water flux across the FO membrane in contrast to the use of sodium chloride or acetate (Ge et al., 2013; Zhang et al., 2011).

Linking individual cells/reactor modules hydraulically in series or parallel allows each cell receives the effluent from the previous cell, which is used to a nearly complete COD removal and desalination (Dong et al., 2017; Kim and Logan, 2011c; Qu et al., 2013; Zuo et al., 2016a). As complex organic substrates are broken down, a simpler substrate composition becomes available to cater to the growth of microbes in subsequent cell/reactors. The HRT should be adjusted on the basis of the composition and waste strength to accomplish this strategy. The pre-treatment of wastewater, such as pH adjustments, addition of buffers, improvement of conductivity and addition of carbon sources, are also substantial to improve the performance of MDCs or minimise the culture start-up times. However, these alteration methods may elevate operational costs. Amongst the proposed pre-treatment method, pH adjustment is more preferred due to its simplicity and proven practical in an industrial scale.

Adopting the protocol from conventional seawater reverse osmosis (SWRO) technology, the feed water must undergo pre-treatment to minimise the unwanted fouling materials from seawater because a poor feed water quality leads to shortening the lifetime of the RO membrane; thus, it has high maintenance costs. Pre-treatment can modify the biological and physicochemical properties of feed water, leading to an increase in the SWRO performance. Several traditional and modern pre-treatment techniques have been widely used in SWRO desalination, which is also adaptable to MDC. The pre-treatment includes pH adjustment, coagulation/flocculation, medium filtration and scale inhibition. As a result of membrane technological enhancement, the pre-treatment approach for feed water is advanced through microfiltration (MF) and ultrafiltration (UF) (Valavala et al., 2011). Membrane-based pre-treatment is more attractive and has been widely used to improve the performance in SWRO (Abdessemed and Nezzal, 2008; Amy et al., 2017; Prihasto et al., 2009), which is also adaptable for an MDC system.

Wastewater and seawater pre-treatments in conventional water reclamation technology are as crucial as in MDC systems. A direct assimilation of pre-treatment strategy as used in the conventional treatment is deemed realistic for MDCs to expedite use of the MDC technology for field applications. As mentioned above, pH adjustment and microfiltration/ultrafiltration are the most practical for the large MDC operation of wastewater treatment and desalination, respectively.

5.5. Negative net energy balance and recommendation to alleviate the issues

For MDC commercialisation, technological barriers described earlier should be overcome, and the marketability of this technology should be identified in the first place. The construction cost should be considered when the scale of a reactor is increased. Energy requirement commonly accounts for a large percentage of operating costs. Although MDCs can extract energy sources from wastewater, they require energy to operate. Thus, a net energy balance should be assessed precisely by considering the power consumption to run a system. An energy balance analysis is performed on several MDC technologies, including CMDC, MREC, OsMFC and MBR (Yang et al., 2019). Except MRECs, the energy consumption of all these systems is high, exceeding the amount of energy the system can generate. Although an MREC requires a low amount of energy input to operate, the energy retrieved from the system is still considerably low for an industrial scale (Yang et al., 2019).

A continuous effort to improve this technological challenge is believed to enhance the efficiency of the system by targeting a zero net energy balance. The target is achievable by obeying the following recommendation: 1) flowing the saline water into the desalination chamber at a high flow rate, promoting a high salt removal rate and minimising energy input; 2) empowering MDCs with an applied voltage to improve the desalination rate and reduce the total energy consumption (Luo et al., 2013; Mehanna et al., 2010b; Zhu et al., 2014); 3) exploiting the use of stored energy from abundant renewable energy for system operation; 4) optimising the power requirement for recirculation pumps to significantly minimise power input with a low effect on desalination performance; 5) concentrating on the self-energy sufficiency system such as in MREC by optimising the number of the membrane in the RED stack and the flow rate of LC and HC solutions to increase the energy recovery that fit for a scale-up. A concerted effort to reduce the capital cost of MDCs is critical to achieve this significant milestone.

5.6. High capital cost and reduction strategies

The capital cost for the MDC system is considerably higher than the standalone MFC system, MEC and conventional wastewater treatment of activated sludge in the order of MDC > MEC > MFC > activated sludge mainly because of the additional membrane, compartment and electrode material. If the construction cost of MEC by using laboratory materials and designs is estimated to 800 times greater than an anaerobic digester (Rozendal et al., 2008), the construction cost for a MDC reactor must be significantly higher. Hence, the capital cost for scaling-up MDC systems is imperative as it influences the profit margin of the generated valuable products. Notably, the construction cost for the assembly and coupling type of MDCs is much higher than the standalone MDC. Although reducing the cost for a reactor setup, an electrode and a membrane material is rather manageable, the overall cost for MDCs such as construction, operation and maintenance cost is still considerably high for field applications (Zhang and Angelidaki, 2016).

(Zhang and Angelidaki, 2016) pointed out that the estimated capital densities of MDC, MEDCC, MREC and MREEC can reach 7105, 9237, 10,658 and 11,842 $ m^{-3}, respectively. The cost analysis reveals that polycarbonate for reactor construction holds the largest portion of the overall cost for all types of MDCs, followed by BPMs, IEMs, cathodes (platinum catalysed), anodes and power supplies (Zhang and Angelidaki, 2016). An earlier report has estimated that the largest expenditure for a lab-scale single BES cell design is allocated for cathodes, which account for 47%; meanwhile, about 40% is expected to be

primarily spent on the current collector for scaling up (Rozendal et al., 2008). A considerable investment for the current collector at a large scale is considered relevant to cater a high charge transfer throughout a large reactor, allowing a high BES performance.

From the perspective of MDC systems, membrane cost can become the most prominent investment, especially for MREC and MRECC, to a large extent. A large fraction of capital cost is allocated for a Nafion membrane and a platinum-based cathode, which accounts for up to 85%, corresponding to \$471.2 m^{-2} and \$589 m^{-2} for the membrane and the cathode, respectively (Rozendal et al., 2008). Several alternative membranes have been proposed to replace Nafion membranes. For instance, Ultrex membranes have been widely used (Chen et al., 2012a; Clauwaert and Verstraete, 2009; Luo et al., 2014) due to relatively low cost (\$ 130 m^{-2}) (Li et al., 2014). For even cheaper membranes, Zirfon ion-permeable membrane offers \$ 53 m^{-2} (Pant et al., 2010a), and it has been successfully used in BESs. Moruno et al. (2018) reported that the non-patterned AEM of quaternary ammonium poly(2,6-dimethyl 1,4-phenylene oxide) (QAPPO) has realised a 1.6-fold higher desalination rate when it is benchmarked with a commercially available AEM. The improvement is ascribed to its greater ionic conductivity and an adequately thin membrane, leading to a minimised area-specific resistance. The development of this membrane is a step towards cost reduction for an MDC system while maintaining better performance than the commercial AEM.

Platinum (Pt) has been widely used in a research on BESs because of a high catalytic activity where a stable cathode activity must be achieved. However, large-scale Pt applications are not feasible because of the high cost and negative impact on the environment (Kundu et al., 2013). Most studies have focused more on the development of cost-effective alternative materials, considering that the material cost used in the lab is sufficiently high. As previously mentioned, nickel-, stainless steel- or MoS_2-based cathodes have shown a comparable or better catalytic performance than that of a Pt-based cathode at a considerably low cost (i.e. \$ 370 m^{-2} for Ni and \$ 57 m^{-2} for MoS_2-coated stainless steel (Selembo et al., 2009, 2010; Tokash and Logan, 2011). A low cost and stable material should be developed, but it is compromised with a high catalytic activity as a result of a high catalytic surface area, which can be achieved through nanostructuring. In the same concern, a biocathode has been considered as an attractive alternative to the precious metal cathode indebted by its simple preparation, relatively low operating cost, the absence of metal catalysts or charge scavenger, good stability and green strategy. However, the effectivity of a biocathode at a large-scale operation remains unidentified. Therefore, biofilms that are more effective and have higher tolerance should be developed and investigated.

One of the main attractions of BESs is the energy recovery from wastewater treatment, which is more appealing than conventional treatments (Escapa et al., 2012; Gil-Carrera et al., 2013; Heidrich et al., 2013). Cusick et al. (2010) estimated that the energy recovered in a BES can reach \$0.19 kg^{-1} COD at a fixed price of hydrogen (\$6 kg^{-1}). However, the cost estimation is only based on the balance between the energy input into the BES and hydrogen generation. Another benefit of BES is the production of a low amount of biomass relative to aerobic treatment (Logan, 2008; Wang and Ren, 2013). This privilege affords to reduce the operation cost in BESs as the energy requirement for activated sludge normally accounts for 50% of operation cost (Ahn and Logan, 2010), requiring ~0.6 kWh m^{-3}, and half of which is mainly to pump air for an aeration basin (McCarty et al., 2011). Indeed, this

Table 3
Summary of the prevailing challenges and potential mitigation strategies for scaling-up MDC.

Challenges	Detail of challenges	Mitigation strategies
1) Cell/reactor configuration based on large-scale specification	• Lack of knowledge and understanding on how to push from a prototype scale to a pilot scale	• Carry out critical analysis to recognise factors responsible for the failure or unsatisfactory output from the scaled-up reactor • Activate more pilot-scale research • Use actual wastewater and seawater
	• Poor mass transfer and low power density • Membrane deformation • High internal resistance	• Use anode with a large surface area (e.g. carbon brush) • Install a supporting structure • Optimise the distance between an anode and a cathode • Optimise the number of membrane pairs if RED is used
	• Incomplete COD removal and desalination	• Assemble the identical MDC units or couple with existing technology
2) Low conductivity and unstable electrode	• Poor electrocatalytic activity of carbon electrode • Low microbial attachment on the electrode • Passive redox kinetics of air cathode	• Develop 3D porous electrodes for anode materials • Develop nanocatalyst-loaded 3D macroporous • Modify a carbon electrode with aryl diazonium • Expose one side of air cathode to the atmosphere to capture an optimum oxygen level • Electrodeposition of nanoparticles on a hydrogen evolution reaction (HER) catalyst • Use activated carbon with a considerably high surface area
	• Expensive platinum as a cathode	• Substitute with stainless steel, nickel foam and MoS_2 • Use a biocathode
3) Membrane biofouling	• Lack of knowledge on the mechanism of membrane biofouling and scaling • Membrane replacement incur an additional cost	• Carefully observe the structure, composition and effect of fouling on the performance of MDCs • Integrate the membrane with antifouling properties • Employ a pressure-driven technique • Apply overlimiting current (OC) or pulse electric field (PEF) • Integrate a membrane with a feed spacer
4) Low actual wastewater treatment and desalination process	• The complex nature of the various source of wastewater • Exhibited a drop in water flux across the FO membrane • Ineffective conventional pretreatment technique	• Link individual cells/reactor modules hydraulically in series or parallel • Pretreat wastewater such as buffer addition, pH adjustments, conductivity increment, carbon source addition, coagulation/flocculation and medium filtration • Use advance microfiltration (MF), ultrafiltration (UF) or membrane-based pretreatment
5) Negative net energy balance	• Energy consumption exceeded the amount of energy the system can generate	• Introduce a high feeding rate of saline water into the desalination chamber • Empower MDCs with continuously applied voltage • Exploit the stored energy from the abundant renewable energy for system operation • Optimise the power requirement for recirculation pumps • Emphasise on the self-energy sufficiency systems such as MREC
6) High capital cost	• Expensive Nafion membrane • Expensive Platinum as cathode	• Replace Nafion with an Ultrex membrane or a Zirfon ion-permeable membrane • Substitute Pt with nickel-, stainless steel- or MoS_2-based cathode as they have shown a comparable or better catalytic performance than that Pt-based cathode • Increase the surface-active area of the electrode through nanostructuring • Use a biocathode

estimation can reduce the energy requirement cost in BESs, including MDCs.

A recent scale-up progress on MDC systems is one magnitude smaller than that of anaerobic digesters and desalination plants, and the precise estimation of the capital cost is impractical to be conducted at this scale. This phenomenon is due to a significant difference in material costs between high-quality materials for research use and low-cost industrial products. However, future scale-up studies on MDCs should focus and optimise the utilisation of cost-effective materials for reactor designs, electrodes, membranes and electrical connectors. Additionally, a future large-scale MDC studies should investigate the feasibility of a biocathode versus a bioanode over wastewater treatment, desalination and energy production. Above all, MDC technology can only become economically favourable if the capital cost is traded-off by highly profitable products such as fuel or value-added chemical production. A summarised discussion under this topic is provided in Table 3.

6. Integrating MDCs with existing water reclamation technologies and future prospects

Nowadays, RO stands as a dominant option for harnessing freshwater from brackish water and seawater, where approximately 60% of global desalination capacity is dominated by RO plants (Voith, 2010). The rest of the water reclamation technology mainly goes to distillation and electrodialysis technology. However, the downside of these technologies is energy intensive and hence costly. These technologies have been well accepted due to an absolute demand for clean water and progressive research towards technological enhancement and gradual cost reduction of RO technique. In the past, conventional RO was limited to low- and medium-desalination capacity. However, in recent years, RO technology has been applied to large desalination of brackish water. It is mainly attributed to the decreased energy consumption of brackish water and the continuous advancement of membrane technology. Seawater desalination using RO has also been exploited, but it has an elevated cost. Attractively, issues can be potentially alleviated using MDC technology as a pre-desalination system to reduce salinity before it can be further desalinated with RO; thus, the energy consumed by the RO device is decreased.

Certain MDC technologies, such as CMDCs and MRECs, offer desalination with electricity or fuel generation (Chen et al., 2011; Kim and Logan, 2011b; Tufa et al., 2016), which can be potentially harvested to power a system's operation. Connecting multiple MDC units that form an assembly system or coupling with other BES technologies can accelerate the use of MDC technologies into practical applications (Dong et al., 2017; Wen et al., 2014; Yuan et al., 2015; Zhang and He, 2012a, b). A former approach is more practical to avoid complexity in terms of operation and maintenance when MDC is integrated with an existing RO infrastructure. Partial or almost complete desalination has been achieved through an assembly system (Kim and Logan, 2011c; Qu et al., 2013; Zuo et al., 2016a), minimising the energy requirement for an RO operation and extending the RO membrane lifetime.

Combining MDCs with the existing wastewater treatment technology such as anaerobic digester (AD) can pave the way for industrialisation. In brief, AD is a series of processes of breaking down biodegradable materials by microorganisms without the presence of oxygen. The process typically utilised for domestic or industrial purposes to treat wastewater or to generate fuels (biogas). In 2016, 60.8 billion m^3 of biogas was produced in the world that contained the energy potential of approximately 1.31 exajoule (EJ) (Association, 2018). However, achieving COD removal that meets the effluent quality standard is a major hurdle in an AD process, especially the elimination of nutrients, such as nitrogen and phosphorus. A nitrogen-rich waste cannot be removed in AD process, given that no oxidising agents present in AD. Nevertheless, oxidising agents such as oxygen, nitrate, nitrite and sulfate should not be added due to an additional −330 mV redox potential is required to maximise the AD performance (Zupančič and Grilc, 2012). Therefore, an AD system needs a supporting process to extensively remove the effluent COD and nutrients (Deng et al., 2006, 2008), which increase operational and maintaining costs. Hence, the proposal of integrating AD and MDC can extend the COD removal and elimination of nitrogen-rich waste in a DA-MDC (Liu et al., 2019b,c) or SMDDC and SMDARC (Zhang and Angelidaki, 2013, 2015b). Moreover, channelling the sludge from an anaerobic digester into the MDC system may potentially reduce sludge production via microbial oxidation in an anode. Additionally, the start-up culture enrichment in MDCs can be potentially shortened as it is supplied with an active microbe-containing influent from an anaerobic digester.

The idea of integrating an MDC with RO is to exploit an existing membrane-based RO facility if a pilot-scale MDC will be performed in the future. Hence, no additional cost is needed to build a RO system. In a conventional RO technology, the energy required to desalinate water is proportional to the initial concentration of water salinity (RO influent). Coupling a MDC with RO can benefit 63–70% reduced salinity from MDCs, which operate without power input (Jacobson et al., 2011a; Mehanna et al., 2010b). Further processing the pre-desalinated water (>50% desalinated water) into the RO system may prolong the shelf life of the RO membrane, thereby lowering the capital cost for membrane replacement. Despite the mentioned advantages, the maximum volumetric desalination rate should be considered in MDCs to cater a large volume of freshwater that needs to be produced at a fast rate in the RO process.

In this section, a proposed integrated system involves the initial anaerobic digestion of domestic or industrial wastewater to yield biogas. The treated sludge-containing effluent from the anaerobic reactor is channelled to the assembled MDCs to fuel the EAB in the system. Simultaneously, the desalination of seawater takes place in the MDC as a pretreatment prior to a complete desalination in the RO device. The selected MDC systems, namely, CMDC, OsMDC, MREC and MEDCC, are proposed to be integrated with the existing infrastructure of AD and RO technologies, which are designed as AD-CMDC-RO, AD-OsMDC-RO, AD-MREC-RO and AD-MEDCC-RO, respectively. Each MDC comprises four identical hydraulically connected units. No single pilot study on MDC has been reported, the present review is trying to determine the applicability of these integrated systems based on their weighted score on the installation cost, operating cost, energy requirement, maintenance cost, COD removal efficiency, desalination efficiency, energy production efficiency and environmental impact (Fig. 12). The evaluation is also supported by the pros and cons of each MDC design, in addition to the reported performance of selected MDCs.

6.1. Prospect 1 (P1): AD-CMDC-RO

In Section 4, the CMDC in assembly orientation not only alleviates the pH imbalance issue, which typically occurs in standalone CMDC, it also increases the total NaCl removal from 76 ± 1% to 97 ± 1% when the HRT is extended from 1 day to 2 days. This achievement deserves the highest score for desalination efficiency (Table 4). Although the Coulombic efficiency is significantly reduced (49 ± 4% to 35 ± 1%) as a result of an increased HRT, the total COD removal is not significantly changed (60 ± 2%, 2 days; 59 ± 2%, 1 day). If a similar performance can be maintained at a larger scale, the integration of CMDCs with AD and RO can be a promising prospect in the future.

The realisation of CMDC must consider the installation, operating and maintenance cost. The cost includes the construction of four identical cells, the installation of four pairs of AEMs and CEMs, a set of hydraulic tubes connecting individual cells and tubes channelling effluent in the integrated AD-CMDC-RO system, with the operation of three pumping devices.

The ORR through an air cathode mechanism is applied on the basis of a documented CMDC design, thereby eliminating the need for an additional energy for aeration. However, the downside of air cathode application is its sluggish reaction (Al-Mamun et al., 2018), and ORR requires

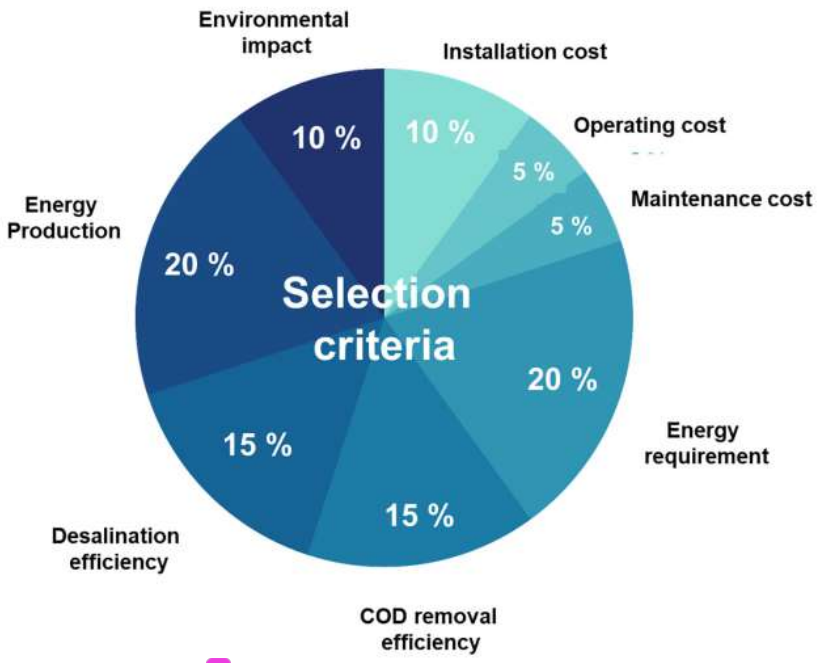


Fig. 12. Selection criteria for the potential prospect of MDCs.

higher potential than HER at pH 7. Therefore, the recovery of power output from the air cathode-installed CMDC to self-power the system may not be effective at a large scale. Even if excess energy such as renewable energy is accessible to generate hydrogen through MEDC (A-type CMDC), directly powering the pumps, which are used for CMDC and RO devices, is more preferable.

A life cycle assessment study on MDCs has demonstrated that the polytetrafluoroethylene (PTFE) binder used for the manufacturing of cell chambers and IEMs can contribute to significant environmental impacts (Zhang et al., 2018).

6.2. Prospect 2 (P2): AD-OsMDC-RO

The prospect of AD-OsMDC-RO offers a lower installation cost than the AD-CMDC-RO. The AEM is replaced by the FO membrane in OsMDC, which is much cheaper than the CMDC; as such, the highest score for installation cost is obtained. The operating cost, maintenance cost and energy requirement for OsMDC may be equal in comparison with CMDC. However, a slightly lower maintenance cost may be imposed to OsMDC because of the fouling-tolerant properties of the FO membrane (Ge and He, 2012). However, the extent is not yet determined.

In terms of COD removal efficiency, OsMDC can achieve 92% in 21 days with a single cell (Ismail and Ibrahim, 2015). A complete COD removal is deemed feasible in an assembled system of four OsMDC units. Thus, AD-OsMDC-RO is credited with the highest point. OsMDC has been proven for the treatment of high-salinity waters because of a stronger water flux for a dilution effect (Zhang and He, 2012b). Although the process is beneficial to the extraction of water from an anode chamber and the decrease in conductivity, OsMDCs do not perform better in terms of salt removal than CMDCs (Zhang and He, 2012b). In this case, OsMDC only gained two points.

One main advantage of the FO membrane is the ability to increase the current generation even under a fouling condition (Ismail and Ibrahim (2015). The phenomenon is driven by the cake-enhanced concentration polarisation in a fouled membrane, which improves ion transport, thus producing a higher electricity than that of prior fouling (Ge and He, 2012). However, a single OsMDC only produces 48.52 mW m^{-2} (Ismail and Ibrahim, 2015), which is much smaller than a single MREC unit can generate, that is, 800 mW m^{-2} (Kim and Logan, 2011c). Therefore, AD-OsMDC-RO receives three points for energy production.

According to the life cycle assessment of MDC (Zhang et al., 2018), the effect of OsMDC to the environment is supposed to be lesser than CMDC systems because of the absence of AEMs; therefore, AD-OsMDC-RO attains the highest score.

Table 4
The selection criteria and weighted score for integrated MDCs with AD and RO.

Selection criteria	Weightage (%)	Score			
		P1 (CMDC)	P2 (OsMDC)	P3 (MREC)	P4 (MEDCC)
Installation cost	10	5	9	1	3
Operating cost	5	6	6	1	4
Maintenance cost	5	5	6	1	3
Energy requirement	20	6	6	9	2
COD removal efficiency	15	6	9	9	3
Desalination efficiency	15	9	2	7	7
Energy production	20	5	3	9	3
Environmental impact	10	6	9	4	4
Total	100	6.1	5.8	6.6	3.5

6.3. Prospect 3 (P3): AD-MREC-RO

Amongst MDCs, MRECs require the highest number of membrane pairs (AEM|CEM) to create multiple narrow chambers for reverse electrodialysis. Significantly high installation, operation and maintenance cost are anticipated, so they deserve a lowest score. In MRECs, a high energy requirement for pumping is needed for wastewater influent, high and low feed concentrations (LC and HC). Ideally, HC can also be the feed solution for cathodes because of high conductivity; therefore, a single pump can be used for dual purposes.

One main advantage of MREC is the ability to self-drive the system because of the recovered energy of substrate oxidation by the EAB and salinity-gradient power (seawater: freshwater, 50:1), which can contribute to 0.2–0.3 and 0.5–0.6 V, respectively (Kim and Logan, 2011a). For pilot testing, the proposed integration of the AD-MREC-RO system is ideal to be located at the vicinity of freshwater as a low concentration (LC) solution and seawater as the source of a high concentration (HC) solution.

An early study on MREC has demonstrated that 4.3 W m^{-2} is generated, and 98% COD is removed when it is equipped with RED cells containing five membrane pairs with a salinity ratio of 50 at 1.55 mL min^{-1} (Kim and Logan, 2011b). Surprisingly, the energy for pumping only accounts for <2% of the generated power (Kim and Logan, 2011b). Therefore, the highest score is credited to energy requirement, COD removal and energy production efficiency if a similar performance can be preserved at a large scale.

The use of MREC for hydrogen production is attractive and economically benefits systems. The price of hydrogen as fuel is already competitive with fossil-based fuels in niche applications ($ 3.7 kg^{-1}; Glenk and Reichelstein, 2019). Hydrogen is one of the highest energy densities per mass than that of fossil-based fuel, which can be harvested using a fuel cell system. For example, MREC with a hydrogen production of 27 mL H_2 production at 1.2 m^3 H_2 m^{-3} d^{-1} is recovered with sufficiently five pairs of membrane pairs (AEMs and CEMs) by relying on electron generation from substrate oxidation and salinity-gradient potential (Luo et al., 2013). However, without the integration of microbial culture (electrodialysis mode), a RED cell requires 25 membrane pairs to produce a power density of 3 W m^{-2} at a salinity ratio of 50 (Tufa et al., 2016). These results show that the application of MREC can surpass conventional electrodialysis in producing energy at minimal membrane pairs; thus, the cost is reduced.

Although the installation of additional membranes, which are higher than CMDC, OsMDC and MEDCC can contribute to an increase in cost, the continuous improvement in membrane advancement may gradually allow a trade-off between high capital cost and high energy recovery efficiency in the future. A low-cost and high-quality membrane is expected to be invented through technological advancement in the near future. Valuable fuel or energy production without energy input is more economically favourable.

Like CMDC, MREC may implicate a more significant environmental impact because of the number of membrane pairs used. However, when MREC is integrated into the existing RO plant, it can reduce the energy consumption of RO operation and be operated by recycling the wasted brine from the RO plant process as a concentrate and wastewater effluent as a diluate in the RED stack cell. This alternative may alleviate the environmental problem faced by the RO desalination industry.

6.4. Prospect 4 (P4): AD-MEDCC-RO

With the additional chamber and membrane for acid production, an increase in installation, operation and maintenance cost is foreseen, which may be comparable with MREC. A lab-scale investigation has demonstrated that an additional number of a membrane with optimised orientation is still needed to improve desalination and chemical production (Chen et al., 2012b; Liu et al., 2015; Luo et al., 2017).

An external energy is required to enhance ion removal in saline solution, substrate oxidation and ORR (Chen et al., 2012a,b; Liu et al., 2015; Luo et al., 2017). In an MEDCC system, the problem of large pH changes in anolytes can be alleviated by the presence of an acidic chamber, which permits the transport of H^+ and OH^- across a bipolar membrane and a CEM. However, the pH of catholytes becomes highly basic and creates an imbalance pH between an anode and a cathode. Theoretically, pH increases as the overpotential increases, i.e. 0.059 V/pH. Alternatively, the produced acidic solution in an acid production chamber can be directly channelled into catholytes to decrease pH, but the pumping requirement is compromised.

Even if the produced chemical can be enhanced and marketable, the revenue should be correlated with the amount of energy needed to produce the chemical, typically by targeting a maximum positive energy balance. The reported performance of chemical production from MEDCCs is still in the millimolar scale (Chen et al., 2012b; Liu et al., 2015; Luo et al., 2017); thus, further research is required. Considering the requirement of a large power input (i.e. 1.0–1.3 V (Chen et al., 2012b)) into four units of a MEDCC, its integration with AD-RO may not be sustainable because of the escalated operating cost and energy requirement to run the system. Moreover, the produced chemical needs additional step for purification, given that the actual wastewater and seawater contain complex ionic compounds, which may diffuse or cross over the membrane.

For CMDCs and MRECs, an additional chamber and a membrane can further affect the environment via chemical pollution. MEDCCs with additional membrane pairs (e.g. consecutive order of BPM-AEM-CEM) can have a commensurate environmental impact similar to that of MREC.

6.5. Outlook of the proposed integrated system

Table 4 shows that AD-MREC-RO is merited with the highest score. The proposed integrated system consists of four identical units of MREC, which is hydraulically connected. Each unit is installed with six AEMs and five CEMs in the RED stacked cell. The membranes are arranged consecutively to create five chambers for seawater (HC: high concentration) and five chambers for river water (LC: low concentration) (Fig. 13). The reactors are electrically and hydraulically connected in series. The large current production, which typically occurs in a large scale, can be prevented by connecting the reactor in series; hence, they can reduce the circulating current (Escapa et al., 2016). Seawater is used as a catholyte, which is pumped into a cathode chamber and flow through each HC cell in the RED stack cell. Simultaneously, river water is channelled into LC cells in the opposite direction against the seawater flow. During operation, seawater and river water flow continuously (Fig. 13). The ideal location for the selected prospect (AD-MREC-RO) is near the abundant source of seawater and river/brackish water (Fig. 14).

Fig. 14 illustrates the proposed integrated system of AD-MREC-RO involving a conventional AD and RO plants. The AD receives domestic wastewater from a residential area (**line a**, **object 1**). Under the self-powered mode, the MREC system must first be acclimatised with potential EAB in each anode chamber by receiving the effluent from AD (**line b**, **object 2**) and flow through the anode of subsequent reactors under active anaerobic fermentation before it ends up into seawater (**line c**). Four MREC cells are expected to provide effluent quality that meets the standard requirement for emission (Kim and Logan (2011c). The cathode chamber is pumped with seawater and allows to flow through the HC chamber of the first and subsequent RED stacked cell in the next reactors (**line d**, **object 2**) before it flows back into seawater (**line e**). River water is pumped into and flows through the LC chamber in the first and subsequent RED stacked cells in the next reactor (**line f**, **object 2**). Both water sources flow simultaneously and continuously in the opposite direction to create salinity-gradient power. Finally, the diluate effluent (LC), which contains less salt from the last reactor, is channelled into the RO plant for further desalination (**line g**, **object 3**).

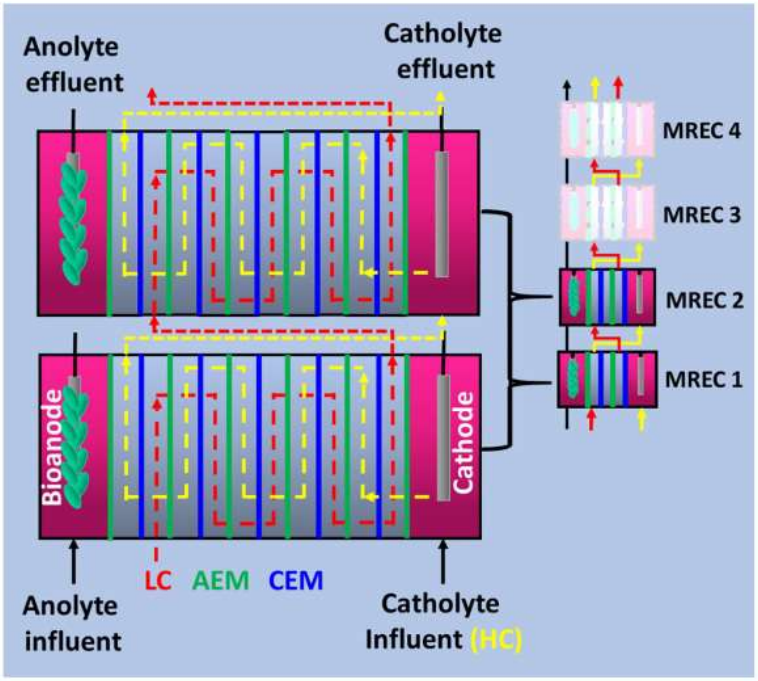


Fig. 13. The proposed MREC stacked system.

The desalinated water from the RO facility is distributed to a residential area (**line h**, **object 8**). One big advantage having RO connected with an MREC system is the possibility of pairing a desalination waste brine as HC (**line i**) and wastewater effluent as LC into the RED feed stream (**line j**) (Mei and Tang, 2018). Such a loop system may alleviate the issue on the absence of nearby river water and environmental issue related to waste brine disposal management from a desalination plant.

The biogas production of methane from AD can be stored in a gas holder (**line k**, **object 4**) during low demand and converted to electrical energy by using the existing combined heat and power unit (**line l**, **object 5**). The stored energy can be controlled and distributed by electrical microgrids (EMs) (**line m**, **object 6**) to power up the water pump and other electrical appliances in AD-MREC-RO plant facilities (**line n**) or directed to the residential area via a power grid (**lines o & p**, **object 7 & 8**). EMs can assist the integration of renewable energy sources and turn any

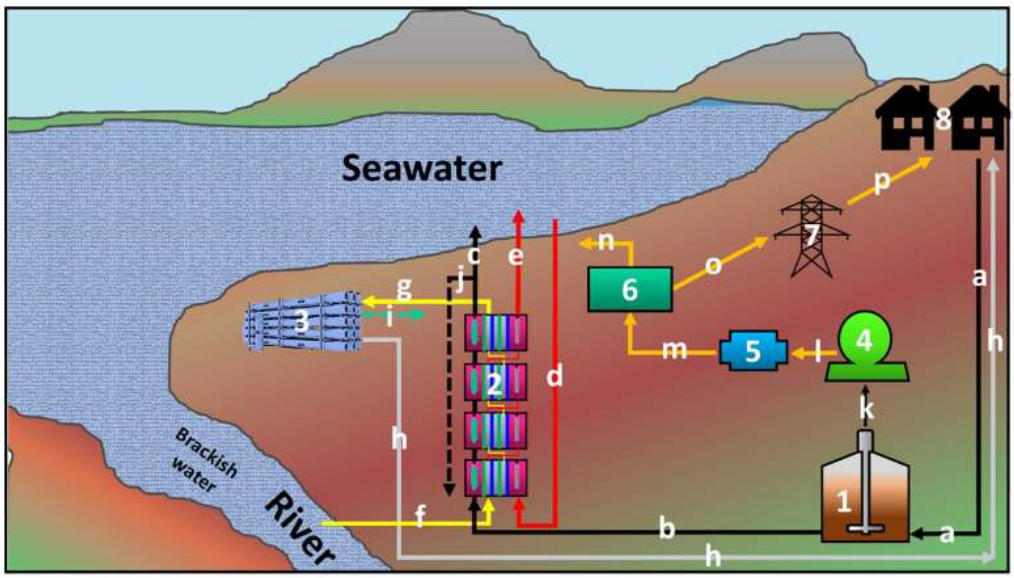


Fig. 14. Schematic illustration of the proposed integrated AD-MREC-RO system. Each numbered object represent; 1) anearobic digester, 2) stacked MREC, 3) RO plant, 4) gas holder, 5) combine heat and power unit, 6) electrical microgrid, 7) power grid, 8) residential area.

conventional power system into more controllable, cost-effective and flexible systems (Zhou et al., 2014).

If hydrogen should be produced through salinity gradient power in MREC, the system can be rewarded with a positive energy balance mainly because of the ability of a self-powering system, a high energy density per mass of hydrogen and the marketability of hydrogen as fuel. However, to reuse the hydrogen to self-power the MREC system is uneconomical because the supplementary device of a proton exchange membrane fuel cell (PEMFC) requires a significant additional capital cost. The analogy is equally applicable if a renewable energy-producing device such as a solar panel or a wind turbine is to be installed nearby to power the proposed integrated plant.

7. Concluding remarks and recommendations for future studies

In the present review, the evolution of MDC designs aims to address the issues related to technical challenges and unsatisfactory performance that emerges from conventional standalone MDC systems. The emergence of advanced MDC designs also aims to accommodate multipurpose reactions involving value-added chemical production, biosensing and environmental remediation, in addition to wastewater treatment, desalination and energy production.

However, the multiple functions of a single MDC unit is inadequate to completely remove COD and salinity in its effluent stream. As such, the recent research trend is geared towards the formation of assemblies with identical MDC units or the coupling with other BESs. Studies have indicated that an almost complete COD removal (>85%) and desalination (>95%) have been achieved (Kim and Logan, 2011a; Qu et al., 2013; Yuan et al., 2015; Zhang and He, 2013). However, they have not been tested with actual wastewater and seawater.

Efforts devoted to scaling up MDC systems have achieved a stepwise-increment strategy from a millilitre up to a hundred-litre operating volume, highlighting that each stage has its own complexity and hurdles. The largest scale ever attempted comprises 30 units of UMDCs with a total working volume of 105 L and exhibits a highly non-uniform performance from each unit. Imbalance and poor performance are observed when MDCs are operated with synthetic wastewater and saline water. These conditions may exacerbate if actual wastewater and seawater are used. Hence, further in-depth large-scale mitigation measures utilising real wastewater and seawater are critically needed.

Before MDC technologies can be brought out of the laboratory, the prevailing challenges related to scaling up should be addressed thoroughly. While encountering the physicochemical and performance issues that arise from a large-scale MDC system, the cost estimation of building up a large cell or a reactor should not be neglected. A large capital investment is inevitable during the research and development of a high-performance MDC system. The maximum performance can be achieved by using a post-effective strategy by providing alternative material choices, construction costs and energy requirements.

Amongst MDC variabilities, we believe that the MREC, an integrated system between MREC and an existing wastewater treatment and desalination method, is likely a candidate for large-scale and pilot testing. A detailed and supportive approach to simulate the dynamic behaviour of MREC at a large scale through mathematical modelling should be conducted. In addition, system costs should be estimated in terms of economic, technical and ecological impacts by using software tools such as RESYSpro and IPSEpro, which are proficient in carrying out economic, technical and ecological analysis (Karagiannis and Soldatos, 2008).

The current status of large-scale MDC systems has indicated that the number of up-scaling studies is inadequate compared with that on MFC and MEC systems. Consequently, insufficient information is provided, delaying its practicality to satisfy the increasing needs of humans. Guided by the established and continuous progress of the up-scaling effort of other BESs (i.e. MFC and MEC systems), the present review lays out several recommendations for future up-scaling studies. With respect to technological challenges and techno-economic considerations, the following recommendations should be considered:

- The flat plate reactor should progress alongside tubular reactors design during the scaling-up effort to ensure adequate knowledge on the challenges and potential mitigation strategies of each design in the literature.
- If an anode is made of brush, the diameter, brush length, number of brushes, electrode orientation and spacing should be optimised to reduce internal resistance, thus increasing power generation.
- A conductive granular material should be used to reduce internal resistance, which is not promoted by nonconductive granular.
- Factors (e.g. the structural and supporting materials of a reactor) affecting the mass and charge transfer from electrolytes to electrodes and vice versa should be identified.
- The fabrication of conductive membranes should be enhanced through feed spacer optimisation to achieve industrial-scale membrane cleaning by using a PEF technique with minimal power utilisation.
- Future up-scaling studies should be operated with actual wastewater and seawater to expedite the practicality of the MDC technology.
- Wastewater and seawater influent having different natures should be preliminarily characterised to identify the compatibility of influents with operating MDC systems.
- MDC operations should be extended (>1 year) to assess the durability and performance of systems and other important measures, such as operation and maintenance costs, which are parts of the capital cost of a large-scale operation.
- The development of a large-scale MDC reactor should be coupled with an RO device of the same scale to assess the feasibility of using them with existing plants.
- Above all, if a high-efficiency wastewater treatment, a high desalination rate and a high energy production are the goals, the capital cost of operating MDC systems must be comparable with current waste-to-energy and water reclamation technologies. This aim can be achieved through critical techno-economic considerations.

List of abbreviations

ACC	activated carbon cloth
AD	anaerobic digester
AEM	anion exchange membrane
AMNRC	advanced microbial nutrient recovery cell
AFMBR	anaerobic fluidized bed membrane bioreactor
AnEMBR	anaerobic electrochemical membrane bioreactor
BES	bioelectrochemical system
BPEC	biophotoelectrochemical
BPM	bipolar membrane
CDI	capacitive deionisation
CEM	cation exchange membrane
CMDC	conventional microbial desalination cell
COD	chemical oxygen demand
DA-MDC	dual anode microbial desalination cell
DC-MDC	dual cathode microbial desalination cell
EAB	electroactive bacteria
FO	forward osmosis
HC	high concentration
HER	hydrogen evolution reaction
HFM	hollow fibre membrane
HFM-MDC	hollow fibre membrane-microbial desalination cell
HRT	hydraulic retention time
IEM	ion exchange membrane
IER	ion exchange resin
LC	low concentration
LCA	life cycle assessment

MBR	membrane bioreactor
MDCs	microbial desalination cells
MCDC	microbial capacitive deionisation cell
MEC	microbial electrosynthesis cell
MEDC	microbial electrodialysis cell
MEDCC	microbial electrodialysis and chemical production cell
MF	microfiltration
MRECC	microbial reverse electrodialysis and chemical production cell
MS-MDC	multi stage-microbial desalination cell
OC	over limiting current
ORR	Oxygen reduction reaction
PBA	photobioanode
PEF	pulse electric field
PEMFC	proton exchange membrane fuel cell
PhMDC	photosynthetic microbial desalination cell
PMDC	photomicrobial desalination cell
PTFE	polytetrafluoroethylene
QMDC	quadripartite MDC
RED	reverse electrodialysis
RMDC	recirculation microbial desalination cell
RO	reverse osmosis
SMDC	stacked microbial desalination cell
SubMDC	submersible MDC
SMDARC	submerged microbial desalination-ammonia recovery cell
SMDDC	submerged microbial desalination-denitrification cell
SWRO	seawater reverse osmosis
TDS	total dissolve solid
TTMDC	two tubular chamber MDC
UF	ultrafiltration
UMDC	up-flow microbial desalination cell

CRediT authorship contribution statement

Mohd Nur Ikhmal Salehmin: Conceptualization, Writing - Original Draft. **Lim Swee Su:** Writing - Review & Editing. **Ibdal Satar:** Writing - Review & Editing. **Wan Ramli Wan Daud:** Supervision, Project administration, Writing - Review & Editing,

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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