Advances in Science, Technology & Innovation IEREK Interdisciplinary Series for Sustainable Development

Inamuddin · Anish Khan Editors

# Sustainable Materials and Systems for Water Desalination





# Advances in Science, Technology & Innovation

## IEREK Interdisciplinary Series for Sustainable Development

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Inamuddin · Anish Khan Editors

# Sustainable Materials and Systems for Water Desalination



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# **Natural Zeolites for Seawater Desalination**

#### Joshua Gorimboo, Charles Rashamao, and Clayton Bhondayio

#### Abstract

Freshwater shortage is one of the major challenges confronting the world today mainly due to climate change, population growth and urbanisation. Seawater desalination appears to be an attractive means for freshwater production. Utilisation of natural zeolites in seawater desalination is currently one of the understudied pathways probably due to other competing technologies, variable properties which makes designing and controlling the zeolite-based process difficult. In some cases, zeolites do not produce water suitable for human consumption. However, zeolites abundancy, low cost of production, high water fluxes, high ion rejections, high thermal and chemical stability as well as compatibility with soil when disposed by landfilling renders them an attractive and sustainable option for intergration into future seawater desalination applications. Research into the use natural zeolites in desalination is therefore ongoing. In this chapter, we review the recent research that focuses on seawater desalination using natural zeolites. The chapter starts with a brief overview of natural zeolites properties and relative abundancy. We then talk about current desalination technologies that exist commercially in industry before delving into the research on seawater desalination using natural zeolites. Generally, not much work has been done on the use of zeolites to desalinate seawater when compared to its use in wastewater treatment. The available literature suggests that researchers have applied natural zeolites in adsorptive desalination, membrane and ion exchange technology taking advantage of zeolites' molecular sieve and cation exchange properties. On adsorptive desalination zeolites have demonstrated potential to replace or work in

combination with silica gel as adsorbents. In the membrane technology the thermally driven pervaporation is preferred over reverse osmosis for both natural and synthetic zeolites. When natural zeolites sectioned from host rocks are used directly without any modification they tend to deliver low water fluxes. On the other hand, synthetic zeolites are costly to produce, although they offer opportunities to tailor make the required membrane properties. Research into the use of natural zeolites for seawater desalination to produce water for agricultural processes by exploiting the cationic exchange properties of this material seems to be gaining traction especially in Japan. The research so far indicates high rejection of Na<sup>+</sup> from seawater and when the cationic exchanger is coupled to an anion exchange material, water suitable for agricultural purposes can be produced. This use of natural zeolites is attractively sustainable in that it can remove only the unwanted salts leaving those that are beneficial to plants in the main product. This ensures that the exchanger can treat more volumes before getting saturated. The use of seawater for agricultural purposes alleviates the dependence on freshwater sources making it available for other needful purposes like drinking, sanitation and industrial manufacturing uses.

#### Keywords

Adsorption • Ion exchange • Zéolites • Pervaporation • Reverse osmosis • Desalination

Nomenclatu	re, Acronyms and Abbreviations
AD	Adsorptive desalination
IZA-SC	Structure Commission of the International
	Zeolite Association
LD	Liquid discharge
MED	Multi-effect distillation
MED-AD	Multi-effect distillation and adsorption cycle
MKP	Monopotassium phosphate

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MSF	Multi-stage flash
MVC	Mechanical vapor compression
RO	Reverse osmosis
RR	Recovery ratio
SDG	Sustainable development goal
TDS	Total dissolved salts
TDS	Total dissolved solids
WHO	World Health Organization
ZLD	Zero discharge
ZSM-5	Zeolite Socony Mobil #5

#### 1 Introduction

The ever-increasing water demand as a result of rapid growth in global population, urbanisation and the diminishing supply of water due to climate change are exacerbating water scarcity in some regions in the world (Djuma et al., 2016; Richter et al., 2013). More water is being drawn out of the conventional water sources than is being replaced by precipitation, runoff and recharge (Richter et al., 2013). It is thus expected that by 2025, 60% of the world population will face severe water scarcity (Schewe et al., 2014). This is an infraction of the Sustainable Development Goal (SDG) 6, which is aimed at ensuring the availability of clean water to current and future generations (United Nations, 2015). Richter et al. (2013) defines water scarcity as a depletion in usable water from available freshwater sources. This means that a portion of the drawn water is consumed and is not returned to the water source. Additionally, if the water is returned with properties different from its original state, i.e. heavily polluted or it is returned to a different source from where it was originally drawn, this contributes to water scarcity because this depletes the volume of available water on the original source. Thus, one way of mitigating water scarcity is sustainable use of water otherwise known as demand side management. This approach includes strategies such as water conservation, recycling, rain harvesting and improved efficiencies at point of use. While these may help to narrow the gap between demand and supply side other means to obtain water must be explored.

Unconventional water resources, such as seawater can be desalinated, to fill the gap between demand and supply. The earth contains 97.5% seawater with average salinity of 35,000 ppm while freshwater sources make up for the remainder. Of the freshwater 80% is locked in glaciers while remaining 20% is in rivers, lakes, and aquifers (Ibrahim et al., 2017). There is a compelling case to tap into the seawater resource. Different technologies exist for seawater desalination and the major ones are thermal distillation which demands a lot of thermal energy and reverse osmosis which demands high electrical energy input to operate high

pressure pumps (Youssef et al., 2014). Another challenge with reverse osmosis is the use of many strong chemicals in water pre-treatment and membrane cleaning processes (Saliby et al., 2009). High energy demand and chemical consumption results in environmentally unsustainable operations due to greenhouse gas (carbon dioxide, nitrous oxides and coal dust) emissions from power plants and chemicals that end up in the environment where they affect biodiversity. Youssef et al. (2014) highlighted that adsorption desalination is a much less energy demanding option and uses virtually no chemicals. However, the technology is still under commercial development (Ng et al., 2014). This is the same status with technologies like zeolitic pervaporation, cationic exchange and reverse osmosis using zeolitic membranes (Aghakhani et al., 2013; Sazali et al., 2019; Wajima, 2019). A review of studies related to natural zeolites use in seawater desalination creates a quick reference source for academia, public or private sector industry stakeholders in their decision making towards this subject.

#### 2 Natural Zeolite Chemistry

Natural zeolites are commonly used in environmental applications and in different industrial applications because of their high abundance, availability and low costs (Stocker et al., 2017). Most naturally occurring zeolites originate from volcanic activity. During and after a volcanic eruption, magma which is molten rock breaks through the earth's crust and flows out in form of lava accompanied by gases, dust and thick ash (Moshoeshoe et al., 2017). Detailed zeolite formation is explained somewhere (Bish & Ming, 1955; Glauco & Ermanno, 2018). Natural zeolite chemical composition is governed by the geological settings and conditions during mineral formation, and they represent a heterogeneous mixture of zeolite minerals together with varying amounts of gangue minerals (e.g. quartz, feldspars and phyllosilicates) (Gorimbo et al., 2014; Stocker et al., 2017).

Natural zeolites are hydrated aluminosilicate minerals made from interlinked tetrahedra of alumina (AlO<sub>4</sub>) and silica (SiO<sub>4</sub>), with pores occupied by water, alkali, and alkaline earth-metal cations (Moshoeshoe et al., 2017). The Si/Al distribution within the zeolite tetrahedral framework determines the nature of the extra framework cations. The greater the substitution of Si with Al, the larger the charge deficiency which must be compensated by cations entering the structure. The +3 charge on the Al makes the framework negatively charged and the extra framework cations inorganic or organic cations balances the charge to keep the overall framework neutral (Auerbach, 1983). Zeolites generally act as good cation exchangers as a result of their negatively charged surface. The zeolite chemical

composition can be best described as having three components: (i) extra framework cations, (ii) framework (aluminosilicates) and (iii) sorbed phase (which is mainly water). Table 1 shows some of the documented zeolites with the three components. The sorbed phase is governed by the extra framework cations, thus divalent channel cations will have more channel H<sub>2</sub>O than mono-valent cations. Divalent cations, like Ca<sup>2+</sup> and Mg<sup>2+</sup>, generally have larger hydration spheres than monovalent cations, like Na<sup>+</sup> and K<sup>+</sup> (Atkins, 2018). There is also more room in the channels for H<sub>2</sub>O because one Ca<sup>2+</sup> occupies less space than two Na<sup>+</sup> ions (Peter et al., 2018).

A database of zeolite structures provides structural information on all of the Zeolite Framework types that have been approved by the Structure Commission of the International Zeolite Association (IZA-SC) (Baerlocher et al., 2007). The zeolites are then grouped according to their structural morphology and as per Nickel-Strunz classification as: (Auerbach, 1983; First et al., 2011; Merkle & Slaughter, 1967). The chain of fibrous zeolites

- The singly connected 4-ring chain
- The doubly connected 4-ring chain
- The 6-ring, single or double
- The hexagonal sheet with handles
- The heulandite unit.

These complex structural units mostly are simply connected to form the actual frameworks, but in some cases vertices, edges or also faces are shared with nearby units. Pictorial views of representative structure are given in Fig. 1.

The general formula of most natural zeolite is best expressed as crystallographic unit cell as  $Mx/n[(AIO_2) x (SiO_2)y].wH_2O$  where *M* is the cation of valence *n*, *w* is the number of water molecules and the ratio y/x usually has values of 1–5 depending upon the structure (Glauco & Ermanno, 2018). The sum (x + y) is the total number of tetrahedra in the unit cell. The portion in square brackets

Structure type code	Schematic chemical formula	Chemical formulae of natural zeolites	Average Si/ (Si + Al + Be)	Range of Si/ (Si + Al + Be)	Dominant extra framework cations	Subordinate extra framework cations
HEU	Heulandite	(Na, K,	0.81	0.73–085	Na, Ca, K	Mg, Sr, Ba
	Clinoptilolite	$Ca_{0.5}_{7}[Al_{7}Sl_{29}O_{72}]$ 0.22H <sub>2</sub> O				
CHA	Chabazite	(Ca <sub>0.5</sub> , Na, K) <sub>4</sub> [Al <sub>4</sub> Si <sub>8</sub> O <sub>24</sub> ] 0.12H <sub>2</sub> O	0.67	0.58–0.81	Ca, Na, K	Sr, Na, Ba, K
	Willhendersonite	$\begin{array}{c} Ca_2(Ca_{0.5},K)_2[Al_6Si_6O_{24}]\\ 0.10H_2O \end{array}$	0.50		Са	K
ANA	Analcime	Na <sub>16</sub> [Al <sub>16</sub> Si <sub>32</sub> O <sub>96</sub> ]	0.67	0.60-0.74	Na	Ca, K, Mg
	Wairakite	$\begin{array}{c} 0.16H_{2}O\\ Ca_{8}[Al_{16}Si_{32}O_{96}]0.16H_{2}O\\ \end{array}$	0.67	0.65–0.70	Ca	Na, Cs
NAT	Natrolite	$Na_{16}[Al_{16}Si_{24}O_{80}]$	0.60	0.58-0.61	Na	Ca
	Scolecite	0.10H <sub>2</sub> O Na16Ca16[Al48Si72O240]	0.60	0.58-0.62	Na, Ca Ca	- Na
	Gonnardite- Tetranatrolite	$\begin{array}{c} 0.64H_2O\\ Ca_8[A1_{16}Si_{24}O_{80}]0.24H_2O\\ Na_{12}Ca_{2.5}[A1_{17}Si_{23}O_{80}]\\ 0.20H_{10}O\end{array}$	0.57	0.52–0.63	Na	Ca
LAU	Laumontito	$0.20H_2O$	0.67	0.65.0.60	Co	No K
LAU	Laumonute	$Ca_4[A1_8S1_{16}O_{48}]0.1011_2O$	0.07	0.03-0.09	Ca	
FER	Ferrierite	$(Mg_{0.5}NaK)_{6}[Al_{6}Si_{30}O_{72}]$ 0.20H <sub>2</sub> O	0.83	0.78–0.88	Mg, Na, K	Ca, Sr, Ba
LEV	Levyne	(Ca <sub>0.5</sub> , Na,) <sub>6</sub> [Al <sub>6</sub> Si <sub>12</sub> O <sub>36</sub> ] 0.18H <sub>2</sub> O	0.66	0.62–0.70	Ca, Na	K
LOV	Lovdarite	Na <sub>13</sub> K <sub>4</sub> [Be <sub>8</sub> AlSi <sub>27</sub> O <sub>72</sub> ] 0.20H <sub>2</sub> O	0.75	-	Na	K
MAZ	Mazzite	$\frac{Mg_{2.5}K_{2}Ca_{1.5}[Al_{10}Si_{26}O_{72}]}{0.30H_{2}O}$	0.72	-	Mg	K, Ca, Na, Ba

Table 1 Schematic chemical formulae of some of the natural zeolites structural group as per Nickel-Strunz classification (Bish & Ming, 1955)

"Nickel-Strunz Classification—Primary Groups 10th ed". mindat.org. Retrieved 22 March 2020



**Fig. 1** Four representations of the periodic zeolite framework **a** heulandite (HEU) series viewed down the crystallographic [001] direction, representing the chains of  $T_{10}O_{20}$  tetrahedra (T = combined Si and Al), **b** chabazite framework (CHA) representing chains of 6-membered rings (tabular zeolites) viewed normal to [001] direction,

**c** analcime framework (ANA) representing chains of single connected 4-membered rings (structural projection along [111] direction), **d** natrolite framework (NAT) representing zeolites with  $T_5O_{10}$  units (T = combined Si and Al), the fibrous zeolites viewed along [001] direction (Baerlocher et al., 2007)

represents the tetrahedral framework and is characterised by an overall negative charge which increases as the Si/Al ratio decreases. The part outside the square brackets consists of exchangeable extra framework cations, which neutralise the framework negative charge, and, finally, water molecules which often coordinate the extra framework cations (Auerbach, 1983; Bish & Ming, 1955).

#### 2.1 The Major Difference Between Natural and Synthesis Zeolites

Use of synthetic zeolites started around 1950 (Sherman, 1999) with an advantage of tailoring structural properties such as internal pore volumes, molecular-size pores, regularity of crystal structures, and the diverse framework chemical compositions. Dozens more artificial, synthetic zeolites are in use. The main differences between natural and synthetic zeolites include the time scale in which they are produced, the amount of material used to produce them and the quality of zeolites. Natural zeolites have many more chemical elements in its structure, and they are found in many geological environments hence they have much greater structural and crystal chemical variability than synthetic zeolites. Synthetic zeolites can be created in few hours or few days in the laboratory whereas natural zeolites can be generated in years or a million years (Auerbach, 1983). Clearly a synthetic compound has a chemical composition made of the ingredients used in the synthesis, hence well-known composition to the producer. The production of a synthetic zeolite is also aimed at providing the material with certain wanted properties, i.e. it tends to be tailored to specific industrial needs. Whereas a natural zeolite is made

of the most common mineral-forming elements and its composition may vary not only from a mineral species to another but also within the same mineral species.

#### 2.2 The Relative Abundance of Naturals Zeolites

Annually global production of zeolites was consistently around 3.2 million tons between 2002 and 2012. This production was spread across 26 countries with the main producer, China contributing approximately 65% of the world total production (Marantos et al., 2012). The second main producer of zeolites produces only 450,000 tons per year while the rest of the countries each produced between 700 and 160,000 tons per year. The combined zeolite production from Canada, Russia, the Philippines, Greece and Italy is just 4500 tons. Very little production come from Germany, Argentina, Serbia and Slovenia with these countries' combined production per year falling below 2000 tons. South Africa is the only country in Africa that mines zeolites at two sites one in Northern KwaZulu Natal (KZN) and the other one in Western Cape (WC), Riversdale both producing a total of 10,000 tons per year. The South African commodity is composed of mainly clinoptilolite, sepiolite, albite, quartz, muscovite and mordenite with the KZN product averaging 50% zeolitic minerals while the WC contain between 20 and 50% zeolitic minerals (Diale et al., 2011). Though currently most of the global zeolite production is used in applications construction and agricultural industries (Mumpton, 1999), the scope for their sustainable use in seawater desalination is the focus in this review. The bar chart, Fig. 2 depicts the statistics for 26 zeolite producing countries across the globe.

**Fig. 2** World zeolite production statistics (drawn from data in Marantos et al., 2012). Others include Ukraine, Hungary, New Zealand, Cuba, Bulgaria, South Africa, Australia, Spain, Canada, Greece, Italy, the Philippines, Russia, Mexico, Argentina, German, Serbia and Slovenia with each contributing less than 1% of the global total production



#### 2.3 Desalination

Desalination is the process of removing salts and other dissolved chemicals from the seawater from water to produce water that meets the desired quality/salinity (Darre & Toor, 2018; Sepehr et al., 2017). For potable water, the requirements are defined by the World Health Organizations (WHO) with the salinity limit set at 500 ppm (Esfahani et al., 2016). Figure 3 is a typical schematic of a desalination system. Currently four technologies are available for water desalination and these include thermal processes, membrane processes, chemical desalination and adsorption technologies (see Fig. 4). Of the four, thermal-based and membrane-based processes dominate the seawater desalination market and accounts for the bulk of the 15,906 desalination plants that are currently operational (Jones et al., 2019). Membrane-based processes include ultrafiltration, electrodialysis, and reverse osmosis (Darre & Toor, 2018). Among the membrane processes, reverse osmosis is the mature technology and accounts for approximately 60% of desalination plants installed and is considered to be the standard desalination technique (Charcosset, 2009). Thermal-based processes include multi-stage flash (MSF), multi-effect distillation (MED), vapour compression distillation, humidification-dehumidification (El-Ghonemy, 2018; Jones et al., 2019). Globally, thermal-based processes especially MSF and MED accounts for 26% and 8% of desalinated water respectively. Other technologies as categorised by Jones et al. (2019) contribute less than 1%.

#### 2.3.1 Reverse Osmosis (RO)

This process is based on applying excess pressure (hydraulic pressure  $\gg$  osmotic pressure) to force water to flow across a semi-permeable membrane from higher to lower solute concentration. High pressure pumps are used to apply

the excess pressure. According to Shahzad et al. (2017) operating plants typically consist of five major components as shown in Fig. 5 which include: (i) the seawater intake system, (ii) feed pre-treatment facility, (iii) high pressure pumps, (iv) RO membranes, and (v) brine disposal and post-treatment facility (Harandi et al., 2017). These components contribute to the overall performance of a desalination plant which in most cases is measured as a recovery ratio (RR). The recovery ratio also known as the purification process efficiency and it is a measure of the fraction of feed water that is converted into high quality water. Several factors obviously influence RR, these include, feedwater quality and availability, product water quality, economics, pre-treatment, post-treatment requirements and desalination technology capability. However, it is sufficient to state that improvements in energy recovery systems as well as membrane technology have improved RR from around 25% in the 1980s to 35% in the 1990s with current estimates ranging between 45 and 50% (Ghaffour et al., 2013; Xu et al., 2013). Membranes that are used for desalination are made from synthetic organic polymers, RO membranes are either cellulose acetate or polysulfone coated with aromatic polyamides (Johnson & Busch, 2010) and they usually come in four basic modules viz. plate-and-frame, tubular, spiral wound, and hollow fibre. A thorough discussion on these modules can be found elsewhere (Johnson & Busch, 2010).

#### 2.3.2 Thermal-Based Processes

Thermal-based processes rely on supplying thermal energy to seawater to convert the water into vapour then condensing the vapour to get potable water (Harandi et al., 2017). Thermal technologies are mostly used for water with high salinity levels and in regions where the cost of energy is low or where waste heat is readily available (Al-Sahali &

J. Gorimbo et al.



Ettouney, 2007; Ng et al., 2013). Figure 6a, b shows the schematic of the MSF and MED while description of the other thermal process can be found herein (Al-Sahali & Ettouney, 2007; El-Dessouky & Ettouney, 2002). Table 2 compares the thermal-based technologies.

#### 2.3.3 Zeolite-Based Systems

A cursory look at literature indicates that zeolites have numerous uses (Eroglu et al., 2017; Gorimbo, 2011; Gorimbo et al., 2014; Kallo, 2001). However, in relation to seawater desalination, the available data suggest that the



technologies that can be applied vary depending on whether one is considering natural or synthetic zeolites. Thus, water desalination research has been performed using zeolite membranes in both Reverse Osmosis (RO) and pervaporation systems. Examples of studies conducted using both natural and synthetic zeolites are shown in Table 3. Generally, the research on the membrane technology seems to be dominated by synthetic zeolites when compared to natural zeolites as shown in Table 3 with research on using pervaporation and conventional reverse osmosis is available. On the other hand, natural zeolites membranes research seems to have picked up recently (articles around 2012) with most of the work based on pervaporation. However, for desalination purposes natural zeolites seem to have found favour in ion exchange technology with huge potential in producing water for agricultural purposes from seawater (Wajima, 2019) while synthetic zeolites have typically been used for wastewater treatment, e.g. (Metes et al., 2004; Watanabe et al., 2004). The choice of a zeolite for a function stems from their inherent differences. The following observations are made and documented: (Auerbach, 1983; Glauco & Ermanno, 2018; Moshoeshoe et al., 2017).

- Synthetics are costly to produce since they are produced from energy consuming chemicals while naturals are processed from natural ore bodies,
- Synthetic zeolites have a silica to alumina ratio of 1–1 and natural clinoptilolite (clino) zeolites have a 5–1 ratio,
- Natural zeolites are more resistant to mildly acid environment, than synthetic zeolites.

The end use of the desalinated water can also dictate the choice of zeolite.

Technology	Advantage and disadvantages	Type of feed water
Multi-stage flash (MSF)	Advantages         (i) Reliable proven technology         (ii) Can operate with waste thermal energy         (iii) Can handle large capacities         Disadvantages         (i) Requires highest amount of energy of all technologies         (ii) Larger footprint required	Seawater
Multi-effect distillation (MED)	Advantages(i) Requires less energy than MSF(ii) Can operate using waste thermal energy(iii) Can handle large capacities(iv) Very high quality product water (<10 mg/L TDS)	Seawater
Mechanical vapour compression (MVC)	Advantages         (i) Improves process thermal efficiency         (ii) Meet needs in remote areas,         (ii) Transportable         (iii) Accommodates wide range of salt concentrations         Disadvantages         (i) Compressor needs higher levels of maintenance         (ii) Limited to smaller sized plant	Seawater and brackish water
Humidification-dehumidification	Advantages (i) Can utilise sustainable energy sources (ii) Can operate at low temperature (ii) Low technology level requirements Disadvantages (i) Small capacity production plants	Seawater or brackish

Table 2 Summary of thermal bases technologies (El-Dessouky & Ettouney, 2002; Younos & Tulou, 2009)

 Table 3
 Research and technologies for seawater desalination using zeolites

Technology		Available research				
		Natural zeolites	Synthetic zeolites			
Membrane	Reverse osmosis	No information available to date (according to authors knowledge)	Duke et al. (2009), Li et al. (2004), Lin and Murad (2001), Zhu et al. (2014)			
Pervaporation		An et al. (2014), Swanson et al. (2012)	Cho et al. (2011), Drobek et al. (2012), Duke et al. (2009)			
Ion exchange		Shimzu et al. (2007), Wajima (2013), Wibowo et al. (2017)	Wajima (2018)			

#### Synthetic Zeolites-Reverse Osmosis

The research on using synthetic zeolites as RO membranes for desalination processes can be traced back to the modelling work that was done by Lin and Murad (2001) wherein a perfect crystal of all-Si, ZK-4 membrane was shown to reject 100% Na<sup>+</sup>. This work became the precursor for experimental validation that was later performed by Li et al. (2004). In this work, Li op. cit. synthesised aluminasupported MFI-type zeolite membranes through in situ crystallisation and tested the membrane through an RO set-up. Results using a 0.1 M NaCl solution demonstrated 76% Na<sup>+</sup> rejection rates at a water flux of about 0.112 kgm<sup>-2</sup> h<sup>-1</sup> under an applied pressure of 2.07 MPa. The synthetic membrane was also tested for by multi-ion water and it revealed rejection rates ranging from 58 to 88% with water flux of 0.058 kgm<sup>-2</sup> h<sup>-1</sup>, and applied pressure of 2.4 MPa (Li op. cit.). More research followed, e.g. Zhu et al. (2014) synthesised MFI-type silicalite membrane and tested the membrane with RO using any artificial seawater (0.3 wt % TDS). The work indicated rejection rates above 93% for major ions while an 83% rejection rate was observed for Na<sup>+</sup>, the experiments were run for 145 h under an applied pressure of 700 kPa. Zeolites-based membranes are more energy efficient with higher permeability and selectivity than organic polymeric materials making nanocomposites RO membrane that incorporate organic polymers and zeolites

better performers in desalination than organic polymer only based RO units (Shi et al., 2019; Subramani et al., 2011). These membranes incorporating zeolites also find application in RO, pervaporation, nanofiltration and membrane distillation desalination systems (Cho et al., 2011).

#### Synthetic Zeolites-Pervaporation

Work has also been done with synthetic zeolites while applying the pervaporation membrane technology. Duke et al. (2009) synthesised silicalite and ZSM-5 zeolite membranes and performed seawater (3.8%w/w) desalination tests using pervaporation, and reverse osmosis (RO). Results from the pervaporation mode of operation indicated rejection rates above 99%, optimal temperature for pervaporation was found to be 80 °C and flux was found to be a function of Si/Al ratio; the authors observed a 30% increase in flux for Si/Al ratio 100. Cho et al. (2011) also performed pervaporation desalination tests with NaA zeolite membrane. The authors claimed that to their knowledge their work was the first application of this type of membrane for pervaporation in seawater desalination. In this work instead of synthetic seawater, the authors obtained seawater from Boryeong Beach on the West Sea coast of the Korean peninsula. Their results indicated more than 99.9% salt rejections for all the ions except for boron, water flux was measured to be 1.9 kg/m<sup>2</sup> h at 69 °C. Further work on desalination using pervaporation was done (Drobek et al., 2012). They synthesised ZSM-5 membranes through secondary growth on tubular ceramic supports and desalination was done on synthetic seawater NaCl in concentrations corresponding to brackish (0.3-1 wt%), sea (3.5 wt%) and brine (7.5-15 wt %) water. Results from this work show rejection rates ranging between 75 and 90% depending on concentration of NaCl. Fluxes as high as 11.5 kg/m<sup>2</sup> h for seawater at temperature of 75 °C. As of 2018 no known pervaporation desalination plant had been operationalized though pervaporation as a technology is commercially applied in other processes like alcohol separation (Xie et al., 2018). Collaborators from two Chinese universities, Ningbo University and Jiaxing University are busy with research on developing composite pervaporation membranes that contain nanotube material and polyamide (Shi et al., 2019). Benefits of such a material include higher water permeability and selectivity. Similar research direction has also been reported in some Malaysian universities (Sazali et al., 2019).

#### Synthetic Zeolites-Ion Exchange

The high ion exchange capacity and absorptivity of zeolites have long been known (Watanabe et al., 2005) consequently they have been used for wastewater treatment. Wajima (2018) tested the exchange ability of both natural and synthetic zeolites for seawater desalination. The followings synthetic zeolites were tested: zeolite-4A and zeolite-13X and a natural zeolite (Nitto). Wajima tested the ability of the synthetic zeolites to remove  $Na^+$  from seawater treated with calcined hydrotalcite (to remove anions). Their work indicated that  $Na^+$  removal using natural zeolite is lower than that using synthetic zeolites and the decrease in salinity was a function of  $Na^+$  content in the solution.

#### **Natural Zeolites-Pervaporation**

Membranes made of natural zeolites have molecular sieve properties (largest channel of the clinoptilolite framework is  $0.44 \times 0.72$  nm (Kazemimoghadam & Mohammadi, 2007)), and they typically have been used in the thermal driven pervaporation (Table 2). Two types of membranes from natural zeolites have been identified viz. (1) membranes, directly sectioned from mineral deposits (Swenson et al., 2012) and clinoptilolite-based phosphate composite membranes (An et al., 2014). The work on sectioning a membrane from a natural deposit of a zeolite was done using high density deposit zeolites obtained from Mt. Kobau and Manery Creek area of British Columbia, Canada. Run of mine samples were machined into thin sections that were used in the pervaporative desalination of water samples with varving salinity levels, including synthetic seawater. Results indicated complete rejection of magnesium, sodium and calcium ions. Water flux decreased from 2.5 to 0.39 kg/m<sup>2</sup> h when feed concentration was increased from 100 and 5500 mg/L Na<sup>+</sup>. Tests were done at 95 °C. Similarly, the same authors (Swenson et al., 2012) further tested their concept's efficacy to reduce contamination in wastewater obtained from in situ extraction of oil sands. Their results indicated again a reduction in the ionic content and near complete removal all toluene from synthetic process water by pervaporation. It was noted that membrane flux was a function of toluene concentration.

While using membranes machined from hard rock as molecular sieves offers a great opportunity to utilise natural zeolites with minimum modification, it however may have limited industrial uses for the following reasons:

- (i) membranes require high density zeolites and therefore its deposit dependent (An et al., 2014),
- (ii) mining and transportation methods must be precise to minimise fracture propagation and,
- (iii) the low fluxes may render operations uneconomical.

Instead, An et al. (2014) suggested manufacturing low-cost clinoptilolite-based phosphate composite membranes that are scalable and property tuneable. Phosphate mixture was prepared by mixing Monopotassium Phosphate (MKP) and Magnesium oxide (MgO) on 1:2 ratio. This mixture was further mixed with Clinoptilolite powder on 1:1 ratio and dry pressed and steamed to produce the membrane. Pervaporative water desalination was evaluated on the membrane at two levels of salt concentration, i.e. 50 and 1400 ppm, interestingly, these membranes show improved sodium removal rates (over 95%) at higher water fluxes (15 kg/m<sup>2</sup> h) when desalinating 1400 ppm saline water. These membranes offer great potential.

#### Natural Zeolites-Ion Exchange

Seawater desalination that takes advantage of cation exchange capacity of natural zeolites has also been demonstrated mostly by Tomoe et al. (2007), Wajima (2013, 2019) and Wibowo et al. (2017). The bulk of the work has been centred on the removal of Na<sup>+</sup> from seawater for purposes of crop cultivation and reducing salinity in salted soils. It has been established that agriculture utilises about 70% of available freshwater, consumes more than more than 50% of drawn water (Shiklomanov, 2000) and is dominantly responsible for water depletion and scarcity (Richter et al., 2013). Seawater is a readily available resource that can be tapped into for agricultural water supply. It is known that seawater contains essential elements that are important for plant growth, however the excessive amounts of NaCl makes it unsuitable to be used directly. Wajima (2018) argues that the conventional and established methods of seawater desalination are all designed to produce high quality water suitable for domestic and industrial use; agriculture does not require the same quality of water. Wajima's work spanning from the mid-2000 to date indicate a possibility of using natural zeolite desalinated seawater for agriculture. For instance, in their early work (Tomoe et al., 2007), they used a combination of Hydrotalcite as inorganic matter to remove anions such as  $Cl^{-}$  followed by cation removal by a mordenite-type natural zeolite; both chloride and sodium ions were reduced. Seawater used in this study was collected from the surface of layer of Imari Bay, Saga Prefecture, Japan. Ion exchange experiments were performed by adding 20 g of natural zeolite to 100 mL of anion depleted seawater and stirred for an hour. Figure 7a shows the results obtained.

Wajima (2013) further expanded on this work by testing several kinds of Japanese natural zeolites, i.e. Iizaka-zeolite (IZ), Tenkawachi-zeolite (TZ), Koriyama-zeolite (KZ), Futatsui-zeolite (FZ) and Niki-zeolite (NZ) following the procedure presented by Shimzu et al. (2007) with the following exceptions: (1) each treatment was stirred for two hours before filtration (2) the treatments were repeated 10 times. Table 4 shows the cation exchange capacity of each zeolite use in this work.

Generally, Na<sup>+</sup> concentration in seawater could be reduced by natural zeolite treatment. The trends (Fig. 7b) suggest that the higher the CEC number the higher the reduction in sodium ions except for sample FZ. Thus, IZ and KZ indicate higher Na<sup>+</sup> reduction than those with TZ and NZ. Note also that sample IZ and KZ have higher Ca<sup>2+</sup> ions than TZ, NZ and FZ, thus it could be deducted that these reductions correlated closely with an increase in Ca<sup>2+</sup> ions. Wajima (2018) expanded on the work they performed (Shimzu et al., 2007) into a single step process where both

**Fig. 7** Variation of Na<sup>+</sup> concentration as of number of treatments with zeolite **a** Tomoe et al. (2007) **b** Wajima (2013)

500 500 concentration (mmol/L) concentration (mmol/L) Ъ (a) 400 40 300 300 0 200 200 0 5°C ∆ 25°C 100 100 □ 60°C Na<sup>+</sup> Na<sup>+</sup> -D-F7 0 0 2 3 4 5 6 7 8 1 0 2 4 6 8 10 Number of zeolite treatments Number of zeolite treatments

**Table 4**Cation exchangecapacity (CEC) for samples tested(Wajima, 2013)

		Exchang				
Sample	Type of crystal	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	CCE/mmol g <sup>-1</sup>
IZ	Mor.	0.56	0.21	0.03	0.44	1.67
TZ	Mor.	0.7	0.11	0.06	0.24	1.27
KZ	Mor.	0.58	0.28	0.02	0.4	1.51
NZ	Cli.	0.88	0.2	0.02	0.23	1.27
FZ	Cli.	0.84	0.71	0.02	0.7	1.67

Mor.: Mordenite-type zeolite, Cli.: Clinoptilolite type zeolite

the Sodium ion and chloride ion are removed by a mixture of natural zeolites and Calcined Hydrotalcite (CHT). This work suggest that it is possible to produce water for agricultural cultivation from high salinity water using the mixture of natural zeolite and CHT. Finally, after having established this possibility Wajima (2019) used ion-exchanged natural zeolite to test the possibility of growing Radish sprout using the solution produced. Two sets of desalination experiments were carried out. In the first set desalination of seawater was conducted using five different ion-exchanged natural zeolites, Na<sup>+</sup>-, K<sup>+</sup>-, NH<sup>4+</sup>-, Mg<sup>2+</sup>- and Ca<sup>2+</sup>-zeolite following the procedure described by Wajima (2013). In the second set of experiments a column with Ca-exchanged natural zeolite was prepared by running a solution of  $1 \text{ M Ca(NO_3)}_2$ through a column packed with zeolite particles with the diameter of 1.0 mm. Seawater was then circulated through this column for 2 h and repeated 3 more times on fresh columns. Results from these experiments show that seawater can be desalinated with ion-exchanged natural zeolites however the Radish sprout could only grow in the solution after 4 times zeolite treatment of seawater.

Wibowo et al. (2017) also carried out some seawater desalination experiments using natural zeolites obtained from Sukabumi, Bandung, Indonesia. Their work involved activating natural zeolites by heating in a furnace under atmospheric condition. Desalination was performed by adding different masses of activated zeolites to 100 ml of seawater with average salinity of 35.0 ppm. Their results also confirmed that natural zeolites could be used for seawater desalination.

#### 3 The Future Perspectives of Seawater Desalination Using Natural Zeolites

#### 3.1 Membranes

Research on seawater desalination systems that takes advantages on the molecular sieve and cation exchange capacity of zeolites has been done. The research on molecular sieve properties has been focused primarily on the thermally drive pervaporation membrane process. Two major types of membranes have been identified viz. (i) membranes sectioned from high density zeolite deposits and (ii) clinoptilolite-phosphate composite. While these present a major step towards understating and eventual utilisation of natural zeolites in seawater desalination, the use of membranes sectioned from deposits of high density zeolites may find limited use industrial wise because of low water fluxes and the dependents on deposit which does not offer control and tuneablity of critical parameters/sieving 11

characteristics of type membrane; however, the work on composite membranes may offer great opportunity of producing zeolite membranes that are scaleable, tuneable and cost fraction of that require to produce synthetic membranes. There is opportunity for further research to improve both cation rejection and water flux. The work reported in literature (An et al., 2014) used a clinoptilolite membrane, other zeolites or a mixture thereof may yet produce different performance characteristics. Cost-benefit analysis need to be carried out with results from actual seawater/brackish water run on pilot scale.

#### 3.2 Ion Exchange

The cation exchange capacity of natural zeolites has also been tested for seawater desalination and it offers the greatest opportunity for sustainable desalination of water. Research reported to date used actual seawater and is specifically targeted at removing mainly sodium chloride from seawater with a specific target of producing water for agricultural purposes. Experimental results by Wajima (2019) prove that it is possible to then grow plants using zeolite ion-exchanged seawater. The natural zeolites can be reused repeatedly by exchange with other, e.g. K<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, Mg<sup>2+</sup> (Wajima et al., 2006).

If the use of natural zeolites to desalinate water for agriculture is successful, it will go a long to reduce water scarcity by reducing amount freshwater used for agricultural purposes. While there is an obvious opportunity further work is required to establish how sustainable the process given that experimental data show that to get appreciable reduction in NaCl, 5–10 cycles of mixing and filtration with a new dose of fresh zeolite at every cycle must be done. Each mixing cycle lasting 2 h. This needs to be optimised investigated further because each mixing cycle consumes energy albeit at levels lower than current desalination methods.

#### 4 Conclusion

Research to utilise abundant, low-cost natural zeolites for desalination is ongoing. The bulk of the work that uses natural zeolites is based on ion exchange technology. This research work using ion exchange technology specifically targets water suitable for agricultural purposes. However, there is also little research activity focused on using zeolites as adsorbents in adsorptive desalination. Natural zeolite-organic polymer composite membranes suitable to desalinate through reverse osmosis or pervaporation to meet potable and industrial water quality are also being studied. Acknowledgements The authors are grateful for the financial support provided by the University of South Africa (UNISA), National Research Foundation (NRF) of South Africa and the Institute for the Development of Energy for African Sustainability (IDEAS) research unit at UNISA.

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### **Desalination Using Electrodialysis**

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#### Abstract

Dwindling water quality has become a global matter of concern due to the overgrowing expansion of industrial/ agricultural activities and climate change that threatens to cause significant alterations to the hydrological cycle. There is an urgent need to treat water from unconventional sources due to the increase in the pollution levels and a decrease in groundwater availability. Membrane desalination technology has gained tremendous importance in the recent past owing to its exceptional capability of water treatment. The electrodialysis (ED) is one of the promising membrane desalination technologies, thanks to its wide range of applications, the flexibility of operation, and robustness. The present chapter covers the essential aspects of the desalination process carried out using ED technology. The chapter highlights the history of ED development, the principle of operation, different parts of ED cell, and preparation of dimensionally stable anode. It also highlights the principle of operation and the concept of reverse electrodialysis (RED) while considering challenges and opportunities in the field.

#### Keywords

Membrane desalination • Electrodialysis • Dimensionally stable anode • Reverse electrodialysis

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#### 1 Introduction

The desalination process is the membrane-driven technology which is used for the removal of dissolved minerals from seawater, brackish water, and wastewater. Considering the availability of water on the earth's surface, 71% of the total water resources are in the form of ice and oceans (Chen et al., 2010; Woo et al., 2019). However, only 3% of the water has considered fresh and appropriate for drinking purpose (Mohammadi & Kaviani, 2003; Sadrzadeh & Mohammadi, 2008). Due to high total dissolved solids (TDS) and salt content, the water from oceans and seas is not drinkable and cannot be utilized directly for other applications as well. In order to make water more suitable for various applications, desalination is necessary that reduces the salts and other impurities. It can be done in multiple ways by adopting various processes while improving the quality of water. The various processes which are used around the globe are reverse osmosis (RO), electrodialysis (ED), multi-stage flash desalination (MSF), and multi-effect evaporation/distillation (MED); these processes are considered as leading methods which are being used by millions of people for freshwater production (Al-Amshawee et al., 2020; Carolin et al., 2017). However, some processes, such as membrane capacitive deionization (MCDI) and capacitive deionization (CDI) are also tested but only at the laboratory scale (Carolin et al., 2017). The benefits, drawbacks, and energy consumption of some of the desalination technologies are illustrated in Table 1.

The ED is a promising method that has been explored for many years. It is an electrochemical method that works on the principle of ion separation through the membranes which are electrically charged. The ions move from one solution to another under the impact of an electric current as of the driving force (Al-Amshawee et al., 2020). Typically, an ED cell consists of ion-exchange membranes arranged one after another between cathode and anode to form separate cells. The ED process has been widely utilized for the retrieval of useful chemicals, mainly from effluents, salt production,

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Sr. No.	Technology for desalination	Consumption of energy	Benefits	Limitations	References
1	Membrane operations using pressure	(kWh m <sup>-3</sup> ) 2-6	<ul> <li>(i) Less</li> <li>utilization of</li> <li>energy</li> <li>(ii) Ease in</li> <li>scale-up</li> <li>(iii) No need for</li> <li>chemical</li> <li>additives</li> <li>(iv) No need for</li> <li>energy recovery;</li> <li>devices</li> <li>(v) Less space</li> <li>required</li> <li>(vi) Proven and</li> <li>well-studied</li> <li>technology</li> </ul>	<ul> <li>(i) Less durability and fouling of membranes</li> <li>(ii) A lesser rate of water recovery</li> <li>(iii) Excess scaling issues</li> </ul>	Ahmed and Ahmaruzzaman (2016), Buonomenna and Bae (2015), Subramani and Jacangelo (2015)
2	Direct osmosis or forward osmosis	~ 21	<ul><li>(i) Production of a highly salted solution</li><li>(ii) Option to select appropriate draw solute</li></ul>	(i) Higher energy requirements	Zhao et al. (2019)
3	Electrodialysis (ED)	1–12	<ul> <li>(i) Rate of brine</li> <li>removal is high</li> <li>(ii) Production of</li> <li>a highly salted</li> <li>solution</li> <li>(iii) Lesser</li> <li>scaling formation</li> </ul>	<ul><li>(i) Capital cost requirement is very high</li><li>(ii) Less water recovery</li></ul>	Nguyen et al. (2013), Zhao et al. (2019)
4	Mechanical vapor compression (MVC)	20–25	<ul><li>(i) Proven</li><li>technology</li><li>(ii) Easy to</li><li>control</li><li>parameters</li></ul>	<ul><li>(i) Scaling issues</li><li>(ii) Consumes</li><li>more energy</li></ul>	Zhao et al. (2019)
5	Multi-effect evaporation/distillation (MED)	7–25	(i) Proven technology	<ul><li>(i) Excess energy requirement</li><li>(ii) Scaling issue</li></ul>	Zhao et al. (2019)
6	Multi-stage flash desalination (MSF)	14–25	<ul><li>(i) Low Scaling formation</li><li>(ii) High rate of evaporation</li></ul>	(i) Lower water recovery rates	Al-Amshawee et al. (2020), Zhao et al. (2019)
7	Desalination using adsorption	1.38	<ul> <li>(i) No limitation</li> <li>on feed water</li> <li>(ii) Stationary</li> <li>operation</li> <li>(iii) High</li> <li>rejection rate</li> </ul>	(i) Used only up to demonstration level	Ng et al. (2013), Subramani and Jacangelo (2015)
8	Membrane distillations	22–67	(i) Can withhold salinity up to $200 \text{ g L}^{-1}$ (ii) Less fouling	<ul><li>(i) Requires high energy</li><li>(ii) Low water recovery</li></ul>	Zhao et al. (2019)

 Table 1
 Summary of wastewater treatment processes using desalination

industrial wastewater, and drinking water. The operating conditions, challenges, and advantages of some of the applications of ED have been shown in Table 2.

#### History

In 1980, the ED was first introduced by Maigrot and Sabates (Campione et al., 2018; Shaposhnik & Kesore, 1997).

Table 2	Strength and	challenges	involved in	n electrodialvsi	s technologies	for desalination

Sr. No.	Electrodialysis technologies	Strengths of ED process	Challenges	References
1	Power free electrodialysis (PFED) for freshwater generation	<ul> <li>(i) Technically feasible</li> <li>(ii) Economically attractive</li> <li>(iii) No requirement of batteries</li> <li>(iv) The voltage produced by RED is steady</li> </ul>	<ul> <li>(i) Concentration polarization</li> <li>(ii) RED internal resistance is slightly</li> <li>lower than ED resistance</li> <li>(iii) At high concentration, less</li> <li>perm-selectivity of membrane</li> </ul>	Luo et al. (2017)
2	For the brine groundwater desalination, the mono and divalent membrane used for selective ED	<ul> <li>(i) The increasing current density with selective membranes turns into excess elimination of salt at a higher flow rate</li> <li>(ii) Considering the massive scope of working operations, a selective membrane demonstrated stable ions</li> <li>(iii) The inorganic components were removed at higher applied voltage</li> <li>(iv) The concentration polarization of the membrane interface was controlled by pulsed electric field</li> </ul>	<ul> <li>(i) The increasing flow rate decreases salt removal efficiency</li> <li>(ii) Stack resistance and membrane deposition</li> <li>(iii) The efficacy of desalting using a selective monovalent membrane was found to be the same when compared with the efficiency of a standard grade membrane during the pilot and bench-scale ED</li> </ul>	Xu et al. (2018)
3	Electrodialysis reversal (EDR) for industrial RO salt-water treatment	<ul><li>(i) Capable of generating high concentrated salt solution</li><li>(ii) Organic fouling was not noticed even after six days of continuous working</li></ul>	<ul><li>(i) Variation in points due depletion of ions</li><li>(ii) The resistance in the EDR process enhanced during 8 h of operation</li><li>(iii) Precipitation of calcium carbonate</li></ul>	Zhao et al. (2018)
4	Ion-exchange membranes for electrodialysis desalination	<ul> <li>(i) High recovery</li> <li>(ii) Little to no feed pre-treatment required</li> <li>(iii) Membrane-based desalination technology does not need any thermal energy</li> <li>(iv) It can be used for desalting a wide range of salinity concentration level from brackish to seawater</li> </ul>	<ul><li>(i) Only removes ions, organics and colloids are not removed</li><li>(ii) Selection of membranes and stacks highly dependent on feed water chemistry</li></ul>	Online and Dharmalingam (2015)
5	Electrodialysis with a nanofiltration membrane (EDNF)	<ul> <li>(i) Reduction in resistant for the migration of ions</li> <li>(ii) EDNF enhances the limiting current density</li> <li>(iii) Improved ion flux</li> <li>(iv) It offers higher acid recovery and demineralization rate</li> </ul>	<ul><li>(i) Soluble elements cannot be separated from the water</li><li>(ii) Less chemical resistance and limited lifetime of membranes</li></ul>	Ge et al. (2015), Van der Bruggen et al. (2008)
6	Photovoltaic electrodialysis desalination (PVED)	<ul> <li>(i) The brackish water with 1000–</li> <li>5000 mg/L TDS content along with seawater can be efficiently and effectively processed using PVED</li> <li>(ii) At higher temperatures and lower flow rates in the feed stream, the effectivity of PVED is higher</li> </ul>	(i) Higher setup cost	Development of Photovoltaic Electrodialysis Desalination System (2016)
7	Bench-scale batch ED operation for improvement in bio-refinery streams	<ul><li>(i) Low cost</li><li>(ii) The ionic migration rate increases with an increase in salt content</li></ul>	(i) 0.3–6.3% losses of organic moieties on the ED membrane	Luiz et al. (2017)
8	RED pilot-scale for municipal and seawater treatment	<ul><li>(i) Renewable and sustainable energy source</li><li>(ii) High efficiency</li></ul>	<ul><li>(i) Spacers clogging</li><li>(ii) Inorganic precipitation due to fouling</li></ul>	Nam et al. (2018)

The ED was utilized as an early concept for the demineralization of sugar syrup using a carbon electrode. Permanganate as the membrane and dynamo was used for the supply of current. Though the term electrodialysis was never used by the group, it was officially found in a patent that was published in 1900 (Grebenyuk & Grebenyuk, 2002). In this patent. Schollmever described the use of ED for the purification of sugar syrup but with soluble iron or zinc anodes. Regardless of this, it was also theoretically claimed that ED was never surfaced until 1911 when Teorell experimentally confirmed the barring principle presented by Donnan after a few years of its first report (Donnan, 1924; Grebenyuk & Grebenyuk, 2002). In view of this principle, it was likely to prepare membranes that allow the selective transfer of cations and anions using fixed negative and positive charges, respectively. The development in the theory of electrochemical principles leading the behavior of ion-exchange membranes (IEMs) came with the possible expansion of newer membrane sheets and to the conceptualization of multiple compartment electrodialysis (Shaposhnik & Kesore, 1997). The actual perception of ED with multiple compartments with alternate anion and cation exchange membrane (CEM) was only realized in 1950. Two scientists, namely W. A. McRay and W. Juda, had developed the first ion-exchange (synthetic) membrane using resins. The membranes were first used in 1954 by Ionics (USA) for Aramco (Saudi Arabia), which is the first desalination plant based on ED technology (Grebenyuk & Grebenyuk, 2002). However, many other ED units were subsequently developed. Figure 1 illustrates the history and development of ED and related processes with respect to technological developments.

In 1974, there was a breakthrough expansion of a concept called electrodialysis reversal (EDR) (Campione et al., 2018; Grebenyuk & Grebenyuk, 2002). Using the new concept of EDR, the ED process can be used by a periodic change in the current, preventing membrane fouling. This process has become a revolution for large-scale industrial applications. Thereafter, many applications based on ED and alternatives were established and made available to the research community, offering further support for developing electromembrane technologies.

The introduction of ED at commercial levels was achieved in the early 1960s, and that was even ten years before the implementation of RO (Chen et al., 2010). The successful progress of the ED system provided a cost-effective technology for the removal of salt from seawater. Further ED was expanded for the treatment of food processing, pharmaceutical manufacturing, and water/ wastewater treatment. ED was a well-established technology with a multitude of schemes functioning worldwide; however, its exposure to seawater desalination was very less compared to thermal or RO methods. In the 1990s, there was not a single plant for desalination working on the principle of electrodialysis with a size of 500 m<sup>3</sup>/day or higher (Turek, 2003). In the World Congress on Desalination and Water Reuse, only one report was surfaced in 2002 determining ED. In 1974, several plants in Japan, China, and India used electrodialysis for desalting seawater but were relatively small, with a capacity of around 120 m<sup>3</sup>/day (Seto et al., 1978). In 1981, A 200 m<sup>3</sup>/day capacity ED setup was installed and operated at Xisha Islands, China, for seawater desalination. After several years of execution, the ED technology was modified to work beyond the design indices. Lately, several plants were opened in India between 1988 and 1993, with overall size each of 5  $m^3$ /day (Turek, 2003). However, it was always stated that ED could be compared with thermal methods or RO (in the range salinity of feed water up to 8-10 g/L) due to the cost associated with ED desalination which is relative to the amount of salt that has to be carried through the membrane (Turek, 2003).

#### **ED** Applications

The ED has a broader application for the removal of salt from brackish water to drinkable water. In Japan, the production of table salt is carried out using the ED process. Initially, ED was mainly used for the desalination of cheese



Fig. 1 Timeline for the development of desalination technologies (Copyright 2019 Elsevier. All rights reserved, reprinted with permission) (Woo et al., 2019)

whey in the food industry. Annually, the total volume of 3 million tons of cheese whey was treated using more than  $25,000 \text{ m}^2$  of installed membrane area (Grandison, 1996). There are various plants that exist with an estimated size of 50,000 kg/day demineralization of liquid whey. Around 10% of the solid part consists of salts (mainly KCL), which forbids its direct use in the formulation of infants due to the requirement of a high-level demineralization (purification). It is projected that about 65% of the ED demineralized whey discovered an end-use in the new-born formulation. Another application of whey minerals (reduced) comprises drinks, dry mixes, animal feeds, ice cream, bakery goods, salad dressing, and confectionery coatings. It was also described that there was an improvement in the textural and flavor quality of milk products (fermented) using the deacidification and demineralization process. ED is also used for the extraction of salts from wine and grape musts with improved stability. Moreover, ED is also potentially used in the refining of sugar from beet and cane. However, the commercialization of these applications is limited due to membrane fouling issues caused by the occurrence of a wide range of organic compounds. ED has found a bunch of applications in biotechnology, particularly for process control, i.e., changing the pH of media used for fermentation or enzyme reactors or as one of the components in down-streaming processing. The ED process can also be integrated for reactor designs or into continuous fermentation. The ED can also be used for the isolation of inhibitive metabolites for continuous fermentation. A group of researchers demonstrated an increase in lactic acid production with the use of ED technology (Grandison, 1996).

#### ED for Desalination

The ED finds its major application in the desalination process. It is a method that is driven with the help of electrical energy separating salt ions from water using IEMs. It only constituted 3% of the worldwide desalination capacity to feed water. The frequency of ED is expected to grow due to novel, cost-effective membranes. The application of ED is significantly particular as it is a membrane technology considered to be a progressive environmental technology that allows the progress of fresh recovery and treatment of water. As the reusability of water, an area of great interest has witnessed massive progress in the treatment procedures, the production of new membranes has gained tremendous attention. For more than 60 years, ED has been considered as a well-proven technology for the treatment industrial and municipal wastewater and brackish water along with the removal of heavy metals and production of bases and acids via its ability to eliminate non-ionic and ionic substances using the effect of electric current (Al-Amshawee et al., 2020). These applications are improved by continuous enhancement in the IEM with improved physicochemical

and electrochemical characteristics. The main benefits of ED include ease in operation, extended membrane life, a higher rate of water recovery compared to RO. Moreover, unlike RO, it does not require a lot of pre- and post-treatment while being able to operate at a higher temperature.

#### Principle of ED

In general, ED is the process of the removal of salt resulted from the application of electric potential. Due to the presence of salt in the solution as separated ions, the applied electric current attracts the ions on the basis of the charge. In ED, two electrodes viz. cathode and anode are kept separately using the stack altering the anion and CEMs with the solution in between them. After applying electric field to both cathode and anode, anions get attracted toward the anode, while passing through the anion exchange membranes and are held by CEMs. Similarly, the cations move toward the cathode, passing through the CEMs, and are held by anion exchange membranes (Fig. 2). Thus, the process comes with two different streams: the concentrate (brine) and dilute (deionized water) (Khan et al., 2016). As ED removes only ions during the process, the fouling of membrane by uncharged ions is less severe in comparison with RO. However, ED is not capable of separating uncharged things such as bacteria or viruses.

#### 2 Design of ED Cell

The electrodialysis process is operated in either batch mode or continuous mode (Fig. 3). The setup can be organized in series or parallel form to achieve an expected degree of demineralization. Typically, the ED system is comprised of many elements, including a pre-treatment system, membranes, membrane stacks, spacers, electrodes, pumping systems, and post-treatment systems. The schematics of the typical ED stack design is shown in Fig. 4.

#### 2.1 Pre-treatment System

The purpose of pre-treatment system is to eliminate the coarse particles from water before initialization of ED process. The occurrence of metal oxides, microbes, suspended solids, and organic acids creates major issues and can alter the chemical composition while damaging the membranes. The suspended particles with an average diameter above 10 mm capable of clogging the membrane have to be removed. Large colloids, metal oxides, and organic anions are responsible for neutralizing the membranes while affecting the selectivity of the membrane. Depending on the nature of feedwater, the complexity and type of the pre-treatment method are determined. The pre-treatment



Fig. 2 Schematics setup of ED stack (Copyright 2019 Elsevier. All rights reserved, reprinted with permission) (Khan et al., 2016)



**Fig. 3** Process of electrodialysis in batch mode and continuous mode (Copyright 2010 Springer Nature. All rights reserved, reprinted with permission) (Chen et al., 2010)

methods include removal of impurities using flocculation (for colloids), filtration, and activated carbon filtration (for organic matter). It is well known that the large colloids and organic molecules (charged) such as humic acid are removed by filtration since they are too large in size to be removed during the ED process through the membrane (Chen et al., 2010). Microbial contamination and growth are other issues leading to membrane fouling. Hence, in order to disinfect these microbes, chlorine is generally used. However, the removal of chlorine from the inlet solution is important as it may damage the membrane. The fouling of membranes from soluble salts such as calcium sulfate is prevented by using sodium hexametaphosphate. After the completion of the pre-treatment, the feed water is supplied through the ED stack (Chen et al., 2010).

#### 2.2 Ion-Exchange Membranes

The IEMs are one of the main parts of ED cells. These membranes are arranged one after another between cathode and anode electrodes in the ED compartment to disperse two streams while carrying out the selective transportation of ions from the inlet (Strathmann, 2010). The IEMs are ion-selective, where the transfer of ions takes place, driven by the difference in the concentration called electro-osmosis,

**Fig. 4** Schematics and structure of ion-exchange membrane stack design (Copyright 2010 Elsevier. All rights reserved, reprinted with permission) (Strathmann, 2010)



i.e., electric potential. Moreover, the capacity of IEMs is based on electrostatic interactions (sorption) and dispersion of ions (Al-Amshawee et al., 2020; Palatý et al., 2009). The IEMs are used in applications such as high-water recovery, high desalination, hydrogen production (electrolysis of water), chemical conversion into electricity (energy conversion and storage), and chemicals synthesis (electrochemical reaction). Despite these applications, the technology is not favored at an industrial scale due to its energy-intensive nature, high resistivity, and poor selectivity. Membranes other than IEMs, known as conventional membranes, are characterized on the basis of pore size and the material; however, the IEMs are characterized on the basis of molecular charge interaction. Therefore, being non-selective and due to the ohmic barrier, the conventional membranes are not useful for ED applications.

IEMs are principally classified on the basis of their charged groups known as anion exchange membrane (AEM) and cation exchange membrane (CEM). They are made up of thin polymeric films such as polyethylene, polysulphone, polystyrene, etc., and have charged groups. The AEMs have a positive charge and mainly consists of secondary amine (- $NRH_2^+$ ), tertiary amine ( $-NR_2H^+$ ), quaternary amine (-NR<sub>3</sub><sup>+</sup>), and ammonium (-NH<sub>3</sub><sup>+</sup>, SR<sub>2</sub><sup>+</sup>, PR<sub>3</sub><sup>+</sup>) which are covalently attached to the backbone of the polymeric membrane and blocks the negatively charged ions and only permits the positively charged ions to pass. On the other side, CEMs have negatively charged groups attached, such as carboxylic acid (-COO<sup>-</sup>), sulfonic acid (-SO<sub>3</sub><sup>-</sup>, C<sub>6</sub>H<sub>4</sub>O<sup>-</sup>), phosphonic acid  $(-PO_3H^-)$ , phosphoryl  $(-PO_3^{2-})$  and these membranes carry out the exact opposite work of AEMs (Mei & Tang, 2018). These important characteristics originated and were described by Donnan (Galama et al., 2013). Considering the properties (physical) of IEMs such as active surface charge distribution, hydrophilicity, resistance to electricity, surface roughness, etc., along with the manufacturing methods of both AEM and CEM, they are classified into homogeneous and heterogeneous IEMs. The first homogeneous IEM was developed in 1933, which was fully bonded with uniform charges, while the heterogenous IEMs have uncharged binding polymer. Homogenous membranes are extensively used for the manufacturing of table salt (Sata, 2000).

#### 2.2.1 Homogeneous Membrane

In this type of IEMs, the membrane matrix is fully distributed homogenously with charged groups. The cross-linked polystyrene modified with sulfonate groups are components of cation-selective membranes. Also, the anion-selective membranes contain cross-linked polystyrene modified with quaternary ammonium groups. Three essential manufacturing methods are identified for homogeneous membranes (Nagarale et al., 2006; Strathmann et al., 2013; Xu, 2005); (a) monomer polymerization, (b) polymeric film introduction with charged moieties, and (c) incorporation of charged moieties into the polymer, accompanied by melting of polymer while forming into a type of film. The styrene-divinylbenzene-based membranes are an example of the use of monomers for homogeneous structures production. Such membranes are extensively utilized in ED and can from aminated or sulfonated be found styrenedivinylbenzene copolymers (Sata, 2000; Shahi et al., 2000). Also, these membranes are used for the production of table salt by concentrating seawater concerning a requirement of increased selectivity of monovalent ions (Sata, 2000). Considering another preparation process, fluorocarbon, polypropylene polymers, and polyethylene are employed for IEMs manufacturing (Xu, 2005). The functionalization of these films, in this case, is generally carried out by grafting acrylic monomers to get the weak acid CEMs (Choi & Nho, 2000; Gineste et al., 1993; Gupta & Chapiro, 1989; Hegazy et al., 1990; Horsfall & Lovell, 2003). Also, styrene, which is a vinyl monomer, is attached to the polymer film and then aminated or sulfonated to produce strong acid AEMs (Herman et al., 2003; Lee et al., 1993) or CEMs

(Gupta et al., 1996; Yamaki et al., 2003). The third method of preparation, is usually implemented for soluble polymers, including sulfonated polymers and polyether ketone (Nagarale et al., 2005a; Xu, 2005). These membranes are highly important as they are considered to be a promising possibility for manufacturing low-cost membranes for ED and other methods employed at stringent conditions such as high temperatures (Nagarale et al., 2006). The electrospinning process, coupled with hot-pressing, is a laboratory-scale method which is proposed for the manufacturing of nano-porous IEMs (Ran et al., 2017). In this process, it is possible to produce a nanofibrous structure consisting of large surface area and high porosity along with higher tensile modulus in comparison with the bulk material. These IEMs are found to have significant applications in diffusion dialvsis (Ran et al., 2017).

#### 2.2.2 Heterogeneous Membrane

Heterogenous IEMs contain ion-exchange (IX) groups in small areas that are spread all over the inert support matrix to provide mechanical support. One such simple form has highly fine cation and anion exchange resins distributed in polypropylene. Also, a much smaller heterogeneous distribution of IX is possibly made from polyvinyl chloride plastisol. The heterogeneous membranes have excellent mechanical strength and thickness, but possess poor electrochemical properties. Due to the use of low-cost binding polymers, these membranes are usually cost less (Nagarale et al., 2006). These IEMs are usually produced by the incorporation of resins (ion exchange) into polymer sheers with three different processes (Nagarale et al., 2005b, 2006); (a) incorporation of particles into polymer sheet, (b) molding (dry) of resins and inert polymer followed by milling, and (c) dissolution of resins in the mixture consisting of a binder (film-forming) accompanied by casting and vaporization of the solvent. Additionally, the new manufacturing processes have been developed with a target to improvise the IEMs structure. Hence, the blending of polymer and pore-filling processes (alternatives) was lately introduced (Ran et al., 2017). The particle size distribution is a crucial feature that heavily affects the mechanical behavior of heterogeneous membranes. More specifically, with the decreasing particle size, the flexibility increases, and with an increase in particle loading, brittleness increases (Campione et al., 2018; Vyas et al., 2001).

#### 2.2.3 Special Ion-Exchange Membranes

These are the types of membranes that are developed by putting a lot of effort into concentrating on the alteration of membrane properties. This leads to the development of special membraned categorized by artificial structure and specific preparative methods (Nagarale et al., 2006; Xu, 2005). The examples are interpenetrating (IPN) and semi-interpenetrating polymer (sIPN) networks (Choi et al., 2003; Lebrun et al., 2002, 2004; Lin et al., 2007; Wang et al., 2011; Wu et al., 2007). The preparation of IPN involves mixing two polymers cross-linked due to fixed entanglement. However, the sIPN consists of branched or linear polyelectrolyte immobilized in a polymeric matrix (cross-linked). Its specific structure permits mechanical and electrochemical attributes to be incorporated for particular applications (Chen & Chen, 2005; Vermaas et al., 2011). Grafting by radiation is another method for membrane preparation representing another class of IEMs. It allows discordant polymers (to be linked), hence enhancing their overall properties. Further, these membranes are readily altered by selecting the desired degree of grafting (Nagarale et al., 2006). Various studies dealing with the characterization and preparation of IEMs employing the grafting process while utilizing them in ED systems have been investigated (Brack et al., 2000; Kostov & Atanassov, 1993; Kostov & Turmanova, 1997; Kostov et al., 1992; Nasef et al., 2000). The former group of a special type of membrane is rendered by a mixture of inorganic or organic IEMs. By mixing the characteristic of polymers with inorganic or chemical materials, thermal and mechanical steadiness has been significantly magnified (Xu, 2005). Therefore, primarily for fuel applications, these membranes are used (Depre et al., 1998, 2000; Gautier-Luneau et al., 1992; Kim et al., 2004, 2005; Mauritz et al., 2004; Nagarale et al., 2004; Walcarius, 2001; Zou et al., 2004). The membrane hybrid structures are typically found through in situ polymerization, blending, intercalation, sol-gel process, or molecular self-assembling, the latter being the most widely used (Kogure et al., 1997; Ohya et al., 1995). Specific improvement has been developed in the special membranes by the incorporation of fixed charges of both negative and positive in the same membrane (Campione et al., 2018; Xu, 2005).

#### Bipolar membranes

The bipolar membranes (BPM) are one of the outstanding examples of special IEMs. BPM consists of a layer of anion exchange overlaid with a CEM layer. Such types of membranes are specifically employed in applications such as electrodialysis, where splitting of water is enhanced to produce acid-alkaline and acid solutions (Tanaka, 1999; Xu, 2005). These membranes are manufactured by various methods such as adhering commercial anion and cations exchange membranes (or vice versa) (Hao et al., 2001), molding polyelectrolyte solution of cation exchange on a membrane of anion exchange (Frilette, 1956), or functionalizing both planes of the standard membrane (Fu et al., 2003; Zhili et al., 1993). Two other exceptional types of membranes contain both classes of charges, the mosaic and amphoteric membranes (Xu, 2005). The earlier represent randomly dispersed weak basic and weak acid groups. The main characteristic of charged groups is to respond to pH variation, hence altering the anionic and cationic selective character of the membrane. It is helpful for the amphoteric membranes, specifically in distinct applications employed in industrial and biomedical sectors (Xu, 2005). These membranes are distinguished by cation and anion exchange zones agreed in a coextending (parallel) way inside a single membrane (Higa et al., 2008). Due to this, in each layer, there is a circulation of individual current, pointing to a salt permeability and negative osmosis greater than the neutral species. Considering these unique features, BMPs are considered as a capable mediator for the waste stream treatment in which organic compounds and salts are separated. However, these membranes are not commercially available (Campione et al., 2018; Xu, 2005).

#### 2.2.4 IEMs Properties and Characterization

The membrane performance evaluation is one of the essential factors and can be evaluated by grouping into three distinct sections; physicochemical, electrochemical, and mechanical. In order to employ ED systems in industrial applications, the mechanical stability of the membrane plays an essential role. Various such tests are developed to evaluate mechanical stability of the membrane (Wang et al., 2017a) as follows; (A) Creep and relaxation tests (Majsztrik et al., 2008), (B) The Uniaxial tensile test: used to estimate vield strength, Young's modulus, strain-softening/ strain-hardening, and elongation at break, (C) Dynamical mechanical analysis is extensively accepted to investigate the glass transition temperature and viscoelastic properties, and (D) Bursting test: a membrane is subjected to increasing pressure until the failure befalls. The mechanical characteristics of IEMs are widely affected by varying features of the membrane material such as water uptake, cross-linking, along with some operating conditions such as temperature and aging (Garcia-Vasquez et al., 2013). Precisely, with an increase in temperature, Young's modulus decreases (Kawano et al., 2002), and membranes turn to be stiffer due to an increase in the degree of cross-linking. On the other hand, if water uptake is significantly higher, membranes plasticize (Safronova et al., 2016).

The physicochemical properties of IEMs are characterized by IEC (ion-exchange capacity), swelling, the permeability of chemicals, ions stability (permeability to diffusion and conductivity), and neutral species (diffusion permeation). The degree of the swelling has an immediate impact on the perm-selectivity, specific conductivity (electrical), and dimensional stability. Further, the extent of swelling can be estimated by calculating the difference of weight between the swollen and dry membrane (Ataollahi et al., 2017; Liang et al., 2017). For the calculation of volumetric swelling, the density of membrane material is needed (Nagarale et al.,

2006). The capacity of ion exchange specifies the number of charges (fixed) in the IEM. The IEC is frequently revealed as milliequivalent (meq) of functional groups/cm<sup>3</sup> or gram of dry membrane and is beneficial to determine the concentration of the fixed charge. IEC is the titration method in which sodium hydroxide (NaOH) is used for negative groups, whereas hydrochloric acid (HCL) is used for positive groups (Ataollahi et al., 2017; Liang et al., 2017; Nagarale et al., 2005a). The electrodialysis and related processes involve the substantial concentration differences in two compartments. Therefore, the water and salt dispersed through the IEMs can facilitate an influential determinant defining the performance of the process. The diffusion of salt can be readily determined to employ the time-lag method or some other superior technique using tracers (radioactive) (Campione et al., 2018). The movement of water through the membranes depends upon two mechanisms: osmosis (due to osmotic pressure difference) and electro-osmosis. The water osmosis permeability can be determined by estimating the weight difference in the reservoirs, intermittently turning off the stack current to eliminate the electroosmotic effect (Galama et al., 2014). The critical influence on the performance of IEMs is mainly based on the electrochemical properties. Particularly two types of electrochemical properties to be estimated are electrical resistance and perm-selectivity. The selectivity of the membrane for the passage of counter-ions is indicated by perm-selectivity. The passage of co-ions between compartments is allowed by a low perm-selective membrane, thus affecting the separation efficiency negatively. The membrane perm-selectivity is determined rapidly by measuring the non-ohmic potential of membranes (Campione et al., 2018; Nagarale et al., 2006; Nemati et al., 2017).

#### 2.3 Spacers

The IEMs cation and anion are separated by the spacers, which are frequently made up of a polymer such as polyvinyl chloride, polyethylene, or speciality polymer. The main purpose of the spacer is to provide support to the membranes. Figure 5 shows the different types of conventional and non-conventional spacers. Generally, the spacers are made in a maze structure, ensuring that they are not pressed onto each other. The maze structure also provides flow dynamics or uniform flow by promoting turbulence (Bruggen, 2015). The spacers also keep the membranes apart. Along with this, due to the solution distribution by the presence of spacers, it helps to increase the limiting current density. The spacers are selected considering the interpretation of the following necessities (Tanaka, 2012, 2015): (a) ease in air release, (b) less electric current transmission effect, (c) lowered blockage of flow-pass (caused due to the



#### Non-conventional spacers



Fig. 5 Different kinds of conventional and non-conventional spacers (Copyright 2018 Elsevier. All rights reserved, reprinted with permission) (Campione et al., 2018)

formation of fine suspended particles in the feeding solution), and (d) lower friction head loss. Further, the structures of a spacer are classified as (i) wave porous plate, (ii) diagonal net, (iii) honeycomb net, (iv) expanded PVC, and (v) porous wave plate.

Spacers are usually used in reverse electrodialysis (RED) stacks to support IEMs, promote mixing and provide the flow channels (Vermaas et al., 2014). It can improve mixing, thus minimizing the concentration polarization while reducing the non-ohmic resistance (Kwon et al.,

2017). Also, the presence of non-conductive spacers minimizes the active membrane area for ionic conduction and makes the movement of ions more ancillary, which resulted in higher ohmic resistance, particularly in LS compartments (Długołecki et al., 2010; Mei & Tang, 2018; Post et al., 2008). Along with this, when the pumping solutions are filled in the spacer compartments, it can serve as the primary source of energy loss due to substantial pressure drops (Veerman et al., 2009; Vermaas et al., 2011, 2014). The significant impacts of spacers depend largely on its geometry and material used (Długołecki et al., 2010; Gurreri et al., 2012; Post et al., 2008; Veerman et al., 2009). Along with spacer configuration, it is also important to confirm proper feed flow circulation by modifying the flow supply. This can be attained via modification in the water supply pattern, such as broader feed manifolds and the addition of more water-inlet and outlet that could ultimately result in higher net power density.

#### 2.4 Electrodes

The anode material is made up of platinum-plated magnetite, graphite, titanium, whereas cathode is usually made using iron or stainless-steel material. The conductors used can be of any shape such as net, flat, or bar. The stacks and electrode chambers are partitioned to avoid solutions to get mixed. The oxidizing substances such as chlorine gas is usually get evolved in the anode chamber. Due to the evaluation of these oxidizing substances, the IEMs are easily depreciated. Hence, it is required to put a buffer chamber in between two sheets of partitions (Tanaka, 2015). The material used for the partition is an IEM (fluorocarbon membrane), a battery partition, or an asbestos sheet. The precipitation of magnesium hydroxides can be prevented in the cathode chamber by the addition of an acid solution into the cathode solution and operating ED under controlled pH condition of the cathode solution. The electrode chamber is supplied with a concentrated solution or feeding solution (Tanaka, 2015). However, sodium thiosulfate or sodium sulfite is used in the discharge solution to reduce the concentration of oxidizing substances in the anode solution. To achieve the neutralization of the solution, a sodium sulfate solution is supplemented to the cathode and anode chamber, by mixing the effluent of both chambers.

The electrode system consists of electrodes in which electrolytes are put in the electrode compartments. In these compartments, with the help of redox reaction, the ionic current is converted to electric current. The system is primarily divided into two groups, with and without reverse electrode reactions (Veerman et al., 2010). The latter typically involved redox reactions (e.g., water splitting) and gas formation (e.g., H<sub>2</sub>, O<sub>2</sub>, and Cl<sub>2</sub>) (Jagur-Grodzinskr & Kramer, 1986). These electrode systems usually lose the high voltage toward gas generation, and additional devices are needed for the collective toxic (e.g., Cl<sub>2</sub>) and/or explosive gases (e.g., H<sub>2</sub>) (Scialdone et al., 2012; Veerman et al., 2010).

In order to improvise the performance for the generation of power and safety issues, systems with electrodes carrying the opposite reactions are commonly used, where no net reactions take place. Further, two sub-groups are made for the electrode systems, systems with homogeneous redox 25

couples (e.g.,  $FeCl_3/FeCl_2$ ,  $[Fe(CN)_6]_3/[Fe (CN)_6]_4$  and Fe (III)-EDTA/Fe(II)-EDTA) (Scialdone et al., 2012) with reactive electrodes (e.g., Cu–CuSO<sub>4</sub> system and Zn–ZnSO<sub>4</sub>) and with inert electrodes (e.g., titanium mesh coated by Ru–Ir metal oxide electrodes, graphite electrodes, etc.) (Mei & Tang, 2018). There is a need to change the feed solution periodically and electric current inversion for reactive electrodes (Mei & Tang, 2018; Veerman et al., 2010). Hence, homogeneous redox couples with inert electrodes are preferred.

#### 2.4.1 DSA Preparation

Dimensionally stabilized anodes (DSA) were invented by Beer in the 1960s, and since then, it has become one of the most essential electrodes considered in electrochemical engineering (Chen & Chen, 2005). As the evaluation of Cl and O leads to the degradation of electrodes; Beer's invention has overcome this technical limitation affecting the chlorine-alkali industry. In numerous electrochemical processes such as water electrolysis, electroplating, metal electrowinning, and electro-floatation along with Cl evolution, substantial efforts have been carried out to study the evaluation of O, which is common in anodic reaction. In general, the DSA electrodes are made by using precious conducting metal oxides as electrocatalysts (Chen & Chen, 2005). The typical process of DSA preparation has been well explained by Chen et al. (2005). In the process, the combination of three metal oxides in various concentrations takes place. The detailed procedure is described below.

#### Precursor preparation

Tin chloride (1.227 g SnCl<sub>4</sub>5H<sub>2</sub>O, 98+%), antimony chloride (0.171 g SbCl<sub>3</sub>, 99+%) ruthenium chloride (0.161 g RuCl<sub>3</sub> · H<sub>2</sub>O, 47.0% Ru) with the molar ratio of 70:15:15 (Sn:SB:Sn) was dissolved in solution consisting of isopropanol (10 mL, 99.7%) and 0.5 mL hydrochloric acid (37%). It is advisable to prepare the precursor solution freshly before coating to be done.

#### Electrode preparation

In the next step of the electrode preparation, titanium disks, with 12.7 mm in diameter and 2 mm thickness with a working area of 1.27 cm<sup>2</sup>, were fabricated using titanium rod (99.2%, Grade 2, McMaster-CARR, CA) and utilized as the electrode material for coating. The thermal decomposition method was utilized for electrode preparation. Before coating, the titanium piece was pre-treated using sandblasting, washed with tap water followed by 10 min ultrasound cleaning using DI, 2 min etching in hydrochloric acid (37%), and further 10 min cleaning using DI under ultrasonic irradiation. After completing the pre-treatment process,

the titanium disks were subjected to the coating at ambient temperature with the precursor solution and kept for drying at 80 °C for 5 min to permit the evaporation of solvents. Further, the disk was subjected to calcination at 555 °C for 5 min. The coating procedure was repeated 20 times. After the coating process, the disk was galvanized for 1 h at 550 ° C. About 1.5 mg/cm<sup>2</sup> oxide was coated on the disk.

#### 2.4.2 Characterization

The successful glazing and composition of the metal oxides on titanium disks were confirmed and analyzed by X-ray photoelectron spectroscopy (XPS) (Fig. 6). The composition and concentrations of individual metal ions on the titanium disks of the coating material were obtained using an inductively coupled plasma spectroscopy (ICP-AMS). The titanium disk was dissolved in boiling HCL (37%), and the concentrations were measured using ICP-AMS. Scanning electron microscopy (SEM) (Fig. 7) and X-ray diffraction (XRD) (Fig. 8) was used to study the surface morphology and microstructure of the coating. The voltammetry behavior was inspected using a galvanostat (Fig. 9) (Chen & Chen, 2005).



#### 2.5 Solution Pumping System

The pumping system is one of the critical components in the ED setup. The pump used maybe a low-pressure, which can circulate the solution throughout the stack. In this type of system, the pump essentially requires sufficient power to minimize water resistance when it crosses through the small channels. However, when the feed solution salt concentration decreases, the solution power requirement increases. The pressure drops inside each stack ranging from 15 to 30 psi for flow cells, and in tortuous cells, it varies up to 70–90 psi. Sometimes, there might be a requirement of inter-stage pumps in order to pump water through 2–4 cells in series with respect to the degree of purification required (Chen et al., 2010).

#### 3 Post-treatment

The quality of desalinated water is highly influenced by the technology used for salt removal or desalination (i.e., ED or related processes) along with the design aspects of the same.





Fig. 7 SEM images of Ti/RuO<sub>2</sub>-Sb<sub>2</sub>O<sub>5</sub>-SnO<sub>2</sub> (Copyright 2005 Elsevier. All rights reserved, reprinted with permission) (Chen & Chen, 2005)



**Fig. 8** XRD analysis spectra of  $Ti/RuO_x$ -Sb<sub>2</sub>O<sub>5</sub>-SnO<sub>2</sub> with a scanning rate of 0.002/s (Copyright 2005 Elsevier. All rights reserved, reprinted with permission) (Chen & Chen, 2005)

The efficiency of the process is also associated with the presence of TDS and any particular species concentrations (Birnhack & Lahav, 2018; Delion et al., 2004; Withers, 2005). Although, in most of the cases, the desalinated water is soft (presence of very less magnesium and calcium ions), slightly acidic, and has very less buffering capacity. Hence, water with such qualities cannot be sent directly for irrigation and domestic consumption (Delion et al., 2004). The post-treatment is required and applied to improve the overall quality of desalinated water, targeting potential users. From the literature survey, it was found that there are no standards set for the water quality processed from the desalination



**Fig. 9** The cyclic voltammograms gained in a 0.5 M  $H_2SO_4$  at the scanning rate of 0.1 Vs<sup>-1</sup> at 25 °C (Copyright 2005 Elsevier. All rights reserved, reprinted with permission) (Chen & Chen, 2005)

plant in the world. However, recently Israel and Cyprus adopted some regulations for the desalinated water. In the year 2007, the Israeli Ministry of Health approved the water quality criteria specifically for desalinated water (Lahav & Birnhack, 2007; Martínez-Alvarez et al., 2016). The same identical set of standard parameters were used by the Water Development Department in Cyprus (Philippou, 2015). Apart from these regulations, there are a few authorized norms that are practiced in many countries leading to variations in the quality of water that barely meet the end-user requirements concerning the public health while maintaining the integrity of the distribution system. Moreover, the presence of calcium in processed water is recommended for public health concerning the CaCO precipitation index, which is crucial for water corrosivity potential, is not addressed (Birnhack & Lahav, 2018). Therefore, the post-treatment should consist of stabilization of water while making it delivered to the end-users. Also, an adjustment of pH while removing unwanted gases must be conducted. In the case of higher salt concentration, the combination of different processes such as RO and IX should be employed.

#### 4 Electrodialysis Reversal

The IEMs used in the ED process can get fouled after a specific period of time. Hence, the research efforts have been made to overcome this issue. The process of EDR was established to minimize the fouling that mainly occurred in membranes. In the early 1970s, a company based in the USA launched the commercial EDR process for electrodialysis. Typically, the EDR process is comprised of a self-cleaning continuous ED setup (Reahl, 2006). The EDR process was

developed on the basis of ED technology. During the EDR process, the reversal of membrane polarity can take place several times (up to an hour). The salt-water stream was converted to the water stream and vice versa. It resulted in the attraction of ions in the reverse direction across the membrane stacks (Chen et al., 2010). With an immediate effect of reversing the flow and polarity, the outlet water can be discarded until the lines and stacks are washed out while required water quality is attained. The flushing step takes about 1-2 min, and then the unit continues to produce water. This process of EDR is highly important in flushing and breaking out the slimes, scales, and other deposits from the cells to prevent problems that can occur in the near future. The flushing will allow the unit to function with fewer pre-treatment chemicals, which ultimately reduces the fouling of membranes. More specifically, the EDR system works

with a higher concentration in the brine or concentrates the streams with a more concise flow into waste (Tsiakis & Papageorgiou, 2005). The significant benefit of EDR is its capability of cleansing colloidal particulates, which are generated on the surface of membranes throughout the operations (Fig. 10). The removal of deposit on the membranes is done by periodically changing the DC power polarity in an EDR system, thus reversing the driving force for this deposition (Allison, 1995). EDR technology offers several benefits. It can help cleaning automatically every 10-20 min while reducing fouling, slimes, and scaling. It also helps in the production of freshwater while enhancing the life of membranes in the EDR system. The stacks in the EDR system can be easily cleaned. The EDR system is less sensitive than RO towards the removal of metal oxides and particulate matters. The EDR system is highly stable and can



**Colloidal Displacement** 

be operated continuously without having any limitation concerning silica. Moreover, the EDR system can function with an effective residual of chloramines, chlorine dioxide, and chlorine that ultimately prevents the biological fouling.

#### 5 Advanced Applications

There are wide processes related to ED using IEMs that are proposed and are increasing a lot of interest among the scientific community around the globe. Hence some distinct technologies, exploring the usage of ED promoting optimization and progress of system component are deliberated hereby:

#### Electrodialysis with bipolar membranes

The special IEMs are used in the electrodialysis process with BiPolar Membranes (EBPM), which consist of two layers of anion and cation exchange membranes adhered to each other mainly employed to manufacture bases and acids from a brine solution. The EBPM stack consisted of AEM, a BPM, and a CEM in an alternating manner. The EBPM unit stack consists of three channels and three membranes, placed one after another or in an alternating manner (Fig. 11). The brine solution passes among AEM and CEM, whereas the basic and acid solution flows two other channels, respectively (Strathmann, 2010). After the supply of electric potential to the conductors, water gets stuck beneath these bipolar membranes and is induced to split by catalytic mechanism into  $OH^-$  and  $H^+$  ions, which then travel through an anionic and cationic layer of BPM, to reach alkaline and acidic compartments correspondingly.

At the instant, both cations and anions travel through IEMs from the brine solution, electrically balancing the channel of OH<sup>-</sup> and H<sup>+</sup>, thus generating the basic and acid solutions and returning the electroneutrality. However, the arrangement or the use of all the three compartments diffused is difficult to operate, and hence many times, only a two-compartment scheme is used, in which the product obtained is either base or acid solution. The excess of H<sup>+</sup> and OH<sup>-</sup> generated is absorbed by the salt solution. The combination can only be used, specifically when it is not suitable to get a highly purified base and acid (Pourcelly, 2002). The EBPM process is considered as an industrial opponent of the electrolysis process for the manufacturing of basic and acidic compounds. There are several scientific works available confirming how EBPM could reach to very low-energy intakes, particularly considering conductive a perm-selective membrane. However, in everyday usage, the amount of



Fig. 11 The use of bipolar membranes in the electrodialysis cell in a conventional three cell compartment configuration (Copyright 2002 Springer Nature. All rights reserved, reprinted with permission) (Pourcelly, 2002)
energy required is significantly improved by the ohmic resistance of stack, the non-ideal perm-selectivity of the bipolar/homopolar membrane, and diffusional losses (Pourcelly, 2002). One of the particular and interesting applications of EBPM is the valorization and treatment of waste salt solution originated by desalination units (Badruzzaman et al., 2009; Fernandez-Gonzalez et al., 2017; Ibáñez et al., 2013; Koter & Warszawski, 2006; Mavrov et al., 1999; Reig et al., 2016; Wang et al., 2014; Yang et al., 2014). This process can also be used to change high intense NaCl (with some extra minor elements) solution into NaOH and HCL. A salt solution is diluted to weaken the removal issues while obtaining some important products. Though EBPM offers some environmental advantages, it has a few drawbacks related to a number of technical, economic concerns along with membrane performance (i.e., electro-osmosis and limited perm-selectivity).

#### Continuous electro-deionization

Continuous Electro-Deionization (CEDI) is measured as a hybrid-type process of blending ED and ion-exchange deionization. Typically, the CEDI unit resembles configuration to ED, with alternating IEMs, dilute, and concentrate units in which the feed solutions travel. Compared to ED systems, in CEDI, a minimum one course is occupied with ion-exchange resins. The CEDI can be arranged in two probable provisions; (a) the compartment (dilute) can be occupied with a mixture of cation and anion exchange resins (Fig. 12a) the cationic and anionic resins are kept into two separated channels divided by a bipolar membrane (Fig. 12b) (Strathmann, 2010).

The resins help ions to move faster from the channel from one membrane to another in the dilute compartment; hence the conductivity of a dilute solution is restricted (Alvarado & Chen, 2014). Therefore, CEDI finds practical usage in the manufacturing of solutions of dilute water for medium and small-scale utilization. To elaborate, CEDI is generally accepted for the synthesis of ultrapure water in pharma, electronics, and food industries (Alvarado & Chen, 2014; Dermentzis, 2008; Strathmann, 2010). Also, CEDI is explored for the elimination of toxic metals from wastewater treatment. The vital benefit of mixed bed resins is the simultaneous removal of cations and anions while preventing double passage solution need and lowering the residence time inside the CEDI unit (Campione et al., 2018).

#### Capacitive deionization

Capacitive Deionization (CDI) is an ion transfer and desalination method depending on the usage of materials (capacitive) to release/eliminate ions from/into mixture in a cyclic fashion (Strathmann, 2010). In the CDI process, the brine mixture passes from the compartment among two separate capacitive electrodes; those are frequently protected with an IEM to enhance the current efficacy, therefore, enhancing the performance of the process (Andelman, 2011). When an electric potential difference is applied to the electrodes, the ions travel, rendering to the produced field of electricity, ultimately captivated on the surface of electrodes





**Fig. 12** Representation of continuous electro-deionization stacks in two different layouts  $\mathbf{a}$  the conventional stack in which the dilute channel is mixed with anion and cation exchange resins,  $\mathbf{b}$  stack with

two different channels filled with cation and anion exchange resins which are separated by a bipolar membrane (Copyright 2010 Elsevier. All rights reserved, reprinted with permission) (Strathmann, 2010)

(capacitive), and eliminating brine from the feed water to produce desalinated water. The polarity of the electrodes gets reversed after reaching the highest capacity of electrodes followed by discharging of ions within the channel accompanying purge stream flowing, thus, reviving the electrodes to produce brine in concentrated form to be disposed of. CDI is yet to be fully developed method; however, real applications are reported, and some commercially manufacturers are already existed (Oren, 2008). Recently, CDI experiences from market entry concerns, which might be due to challenges in the upscaling concerning the huge quantity and cost of electrode material required for high volume plants (Anderson et al., 2010).

#### Shock electrodialysis

Considering the operating conditions in ED, the over limiting and limiting currents are possible, yet these are neglected in standard operation (Campione et al., 2018). Even though, shock electrodialysis concept is nowadays recommended for the desalination of water operated under limiting current conditions (Deng et al., 2013, 2015; Schlumpberger et al., 2015). The repeating unit of shock ED consisted of a porous medium (weakly charged) arranged in a compartment between two same-charged membranes of ion exchange (AEMs or CEMs) (Fig. 13). Conventionally, in electrodialysis, a number of repeating units are there next to IEMs, and electrodes are placed at both the end of piles. When the brine solution enters through channels, the electrodes are subjected to the application of potential. Due to this, a flux of cations and anions gets formed in opposite directions.

In the case, when CEMs are selected as a selective layer, sodium ions are discharged from the compartment to form a reduction region at the one side of the partition and concentrated enhanced zone at the opposing side. In contrast, anions transfer from the reduction to the fortified area, staving obstructed in the upper section of the channel by CEMs. In ED, the limiting current is reached, when the ion concentration at the membrane boundary move toward zero. Although, ions move in a much faster way when limiting current is applied in the presence of the porous medium that is weakly charged. A sharp boundary gets formed by shock waves among the depleted and un-depleted zones when the over limiting current reaches the endpoint of the zone of depletion circulated via pores. When the mixtures pass between the two distinct zones are divided by a splitter kept in the end section of the channel, both brine streams and desalinated water are obtained separately (Deng et al., 2015). The discussion is still going on regarding the phenomena of shock ED at over limiting current conditions (Dydek et al., 2011; Mani et al., 2009; Yaroshchuk, 2012). It is believed that two main phenomena occur, surface convention and surface migration (Dydek et al., 2011). In the first phenomena, shock ED occurs by electroosmotic flow leading in large-sized pores, whereas the second phenomena occur due to sub-micropore. Shock ED is one of the current technologies, which are still at the primary stage of progress. Even though there are no applications at the industrial scale,



Fig. 13 The shock electrodialysis diagram with an indication of system repeating unit (Copyright 2018 Elsevier. All rights reserved, reprinted with permission) (Campione et al., 2018) some scientists have shown a small and scalable model that is capable of eliminating more than 99% of salt from the feed water while obtaining the concentration of salt up to 0.1 M (Schlumpberger et al., 2015). Moreover, another group has produced a small setup explaining the opportunity to employ shock ED in disinfection, separation, and filtration (Deng et al., 2015).

#### RED-ED couplings for low-energy desalination

Recently, significant progress has been made in the field of ED for salt removal by linking with osmotic dilution or salinity gradient power devices for low-energy desalination (Vanoppen et al., 2016). In consideration of all the various alternatives, the coupling of ED with RED or, specifically, with a unit of desalination, CDI, or RO (Jande & Kim, 2014), resulted in a technological breakthrough, especially for seawater treatment (Campione et al., 2018; Feinberg et al., 2013; Kwon et al., 2015; Li et al., 2013; Mei & Tang, 2017; Wang et al., 2017b). Figure 14 represents the possibility of couplings in RED-ED setup. In its initial configuration of Unit I-II, the RED is employed for the pre-treatment process. As soon as a less salt solution, unfavorable for non-potable water synthesis, is available, it is used for the dilution of inlet feed-in RED unit and seawater being used as a concentrate. Power is produced because of the alteration in the salinity concentration. Exit mixture from RED seawater is further diluted, which is attributed to the transfer of salt into reduced water without being directly mixed with it. The initial less

concentrated seawater stream can further be fed to the salt removal compartment, where the requirement of energy to arrive at the target concertation will be significantly reduced. Along with this, to decrease the complete consumption of the whole process, the energy from the RED unit will be further used. Also, the demand for the overall process can be additionally reduced by using the RED energy generation. In the second type of configuration where units II and III are coupled together, the RED unit can be employed for the post-treatment while mixing the brine with impaired water. It helps to recover energy from the two waste streams along with the modification of the disposal issue. The two arrangements can also be practiced concurrently.

Additionally, in the operation mode of classical SGP, two different modes can be operated using the RED unit, thus helping as an improved device of osmotic dilution (Vanoppen et al., 2016); (a) scRED (short-circuited reverse electrodialysis), and (b) ARED (assisted reverse electrodialysis). In scRED method, enhancement in the rate of salt transfer from concentrated to dilute compartment occurs. The external load of the RED unit is short-circuited, thus, abandoning to energy production and hence enhancing the dilution effect. In ARED method, an extra external electric potential is applied to the RED unit to attain an enhanced current inside the scRED unit. This possibly consumes energy while supporting the process of dilution, thus, enhancing the highest possible current in scRED. Considering both scenarios, the coupling with the first configuration gains more advantages, in which the dilute plays an important function in decreasing the complete



**Fig. 14** The RED/ED integration possibilities. Unit I represent RED (scRED, ARED) used as the pre-treatment for the dilution before supplying the seawater inlet to ED (Unit II). Unit III shows RED used as the post-treatment for energy recovery from waste salt solution and

relevant dilution with the decreased water stream. Coupling Unit I, II, and III represents the most complete integration possibility (Copyright 2018 Elsevier. All rights reserved, reprinted with permission) (Campione et al., 2018)

consumption of energy, whereas in the second, the dilution is deemed advantageous, solely for environmental purposes. Considering the complexity of some methods, it is tough to evaluate which of the RED operations method is the most appropriate to get the complete energy utilization. Moreover, there is a significant increase in capital costs, when the dilution process is used at high levels, this mainly depends on the quantity of membrane area required to carry out the process. In view of this, for such hybrid processes, the optimization of some parameters is very important. In some literature, the coupling process of RED-RO has been studied by many researchers; though, all are at a conceptual level (Feinberg et al., 2013; Kwon et al., 2015; Li et al., 2013; Mei & Tang, 2017). Particularly, Li et al. (2013) explained the RO-RED method simply utilizing a mathematical model, proving that the method can possibly achieve  $\sim 50\%$  less energy requirement in comparison with the very high-level seawater RO. Vanoppen et al. (2016) have done a reasonable investigation for combined RED-RO operational modes, explaining and identifying the various advantages of reduction in energy consumption and operational ranges. In these scenarios, it has been found that there is a decrease in the specific energy  $(< 1 \text{ kWh/m}^3)$  and exhibited to be achievable theoretically, even though there is a requirement of an increase in overall membrane area (considering the RED membranes). The coupled RED-ED process is not much considered unless recently. A publication by Wang et al. (2017b) studied a very particular case of salt removal from high salinity containing phenols. They have found a significant reduction in the energy consumption up to 30% in comparison with a single ED process although, such results cannot be compared with processes such as RO, where there is very high consumption of energy than the base case (>  $20 \text{ kWh/m}^3$ ). It is very important to note that other processes, such as pressure retarded osmosis (Feinberg et al., 2013; Kim et al., 2013), an alternative to RED has been recommended in recent literature. Similar to the RED method, some other processes, such as pressure-assisted osmosis (Blandin et al., 2015) and forward osmosis (FO) (Blandin et al., 2015; Cath et al., 2010; Yangali-Quintanilla et al., 2011, 2015), have also been given importance to further increase the dilution while decreasing the overall energy utilization during the desalination (Campione et al., 2018).

# 6 Conclusions

Nowadays, electrodialysis is considered as one of the important processes for desalination of water. From the history of ED, it is consistently occurred in the market, specifically for desalination of brine water. However, recently there is considerable interest being developed for ED and related methods due to the implementation of new industries, evaluation of innovative applications, and manufacturing of new IEMs. The important factor in the ED process is IEMs. As IEMs are responsible for the transport of ions from one side to another, it is very necessary to consider the important design aspects of IEMs such as a manufacturer of high permeable, low resistance membranes for high salt concentration solutions and development of surface modification techniques with an improved property of IEMs. Along with this, the cost associated with ED is equally important; hence, the development of a low-cost model is important for growth in the market. Also, the electrodes which are required for the current dissipation are crucial. As anode gets degraded with the oxidizing agents, a dimensionally stabilized electrode preparation is very essential. Also, the process of DSA preparation is comprehensively discussed. The prolonged use of IEMs causes membrane fouling; hence, the removal of the deposits is pivotal to improve the ED performance. This can be done using the concept called EDR. Overall, the ED can be used as a promising technology for various applications in wastewater treatment, industrial effluent, food, and pharma.

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# **Membrane Fouling in Desalination**

Maryam Ahmadzadeh Tofighy and Toraj Mohammadi

# Abstract

Clean and safe water scarcity due to population growth and industries development is a very important challenge, worldwide. As the demand for clean water increases, policies such as seawater desalination and wastewater reuse have been implemented in some countries. Membrane-based desalination is recognized as one of the promising ways to address the global challenge of water scarcity. Today, membrane-based desalination using polymeric membranes with many advantages has been considered as one of the most important methods to produce clean water, worldwide. Fouling as the most important challenge facing polymeric membranes limits the performance of membrane in long-term operation. Occurrence of fouling leads to decreasing membrane life, permeation flux and rejection, and also increasing transmembrane pressure and operating costs. There are several ways to control fouling in the membrane desalination processes including membrane surface modification, novel membrane development, pretreatment, membrane monitoring, and membrane cleaning. The advancement of nanotechnology has revolutionized the membrane desalination industry. Novel nanomaterials, especially graphene oxide (GO) and carbon nanotubes (CNTs), have been identified as promising candidates for the modification of high-performance desalination membranes. Using these nanomaterials, higher water flux than that of the conventional RO membranes could be obtained, successfully. Therefore, fabrication of novel membranes using new materials and via novel methods can be effective in delaying fouling phenomena and therefore increasing efficiency of the membrane desalination processes. Also,

membrane surface modification by coating and grafting methods can delay the membrane fouling by altering the properties of membrane surface including hydrophilicity and roughness. In this chapter, after recognizing fundamentals of membrane fouling and fouling types, recent advances in preparation and evaluation of new desalination membranes with improved antifouling properties are being reviewed.

#### Keywords

Desalination • Membrane technology • Fouling • Modification • Nanomaterials

# 1 Introduction

All living things on the earth need clean water as vital resource. Also, clean water should be used in most of the industrial processes such as petrochemical, gas and oil, food and beverage, electronics, and most importantly in agriculture (Jiang et al., 2017; Teow & Mohammad, 2019; Tofighy & Mohammadi, 2010, 2012a). Therefore, lack of clean water can negatively affect human health, economic growth, and industrial productions. On the other hand, continuing population growth, urbanization, and industrialization increase global water demand, significantly (Hibbs et al., 2016; Jiang et al., 2017; Tofighy & Mohammadi, 2011a). Since the saline water resources (oceans and seas) make up about 98% of the total water resources of the Earth, desalination of the saline water can be considered as the best solution to overcome the clean water scarcity. Research into the development of desalination technology is now being seriously pursued around the world. About 61% of water treatment processes worldwide are desalination plants (Goh & Ismail, 2015a; Goh et al., 2017; Qu et al., 2013; Tofighy & Mohammadi, 2011b).

Membrane technology with desirable properties such as cost-effectiveness, energy-efficient, easy scale-up, no phase

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change, easy operation, low pollution, low environmental footprints, low investment and operating costs, and high ability to combine with other separation processes has been considered as the essential technology to solve the problem of water shortage around the world (Goh & Ismail, 2015b; Kiran et al., 2016; Tofighy & Mohammadi, 2015a; Tofighy et al., 2011). Membrane technology can be widely used for clean water production through brackish water and seawater desalination and industrial effluents treatment. The most important membrane processes are nanofiltration (NF), ultrafiltration (UF), microfiltration (MF), reverse osmosis (RO), pervaporation (PV), membrane distillation (MD), forward osmosis (FO), electrodialysis (ED), and gas separation (GS) (Nasseri et al., 2018; Tofighy & Mohammadi, 2015b, 2020a; Tul Muntha et al., 2017). Among these processes, RO with high rejection and water flux and energy consumption of 1.8 kWh/m<sup>3</sup>, which is much less than that of thermal desalination technologies, is widely used in desalination (Jiang et al., 2017). About 60% of desalination plants worldwide are RO (Drioli et al., 2011). However, one of the most important challenges in the RO process is undesirable adsorption, accumulation, and precipitation of foulants inside the membrane pores or on the membrane surface as membrane fouling (Mansouri et al., 2010; Matin et al., 2011). There are several ways to control fouling in desalination process including membrane surface modification, novel membrane development pretreatment, membrane monitoring, and membrane cleaning (Henthorne & Boysen, 2015; Jiang et al., 2017; Robinson et al., 2016).

Polymeric membranes with desirable properties such as high processability, low cost, and excellent mechanical strength are used in water treatment and desalination commonly and their commercial market is impressively huge. One of the most important challenges in using polymeric membranes in the desalination processes is the trade-off relationship between salt rejection and water permeation flux and also fouling tendency that limits their separation characteristics (Goh et al., 2013; Shannon et al., 2008). Membrane fouling significantly reduces membrane flux and rejection and also increases operating costs by increasing energy consumption (increasing pressure drop), reducing membrane life (due to the need for membrane washing (cleaning)) as well as required pretreatment (due to the need for additional pretreatment processes) (Chang et al., 2019; Goh et al., 2018). Therefore, improving the polymeric membranes properties through various modification methods is very important. In this field, developing of thin-film nanocomposites (TFN) membranes and mixed matrix membranes (MMMs) and also surface modification of membranes via coating and grafting methods have been proposed to increase the desalination processes efficiency and solve the fouling problem (Azelee et al., 2017; Jalali et al., 2019; Yin & Deng, 2015). It is expected that the use of nanomaterials with unique properties in the membranes structure can improve desalination performance, significantly. As reported in the literature, using carbon nanomaterials, especially graphene oxide (GO) and carbon nanotubes (CNTs) can create high permeation flux and salt rejection and also delay membrane fouling (Ismail et al., 2009; Valamohammadi et al., 2020).

Recently, many articles have been published in the field of membrane fouling control, which shows the importance of this issue and the great interest of researchers in this field of research. As a result, writing a chapter in this research field is very important. In this chapter, after recognizing fouling phenomena, fouling types and fouling control strategies, recent advances in preparation and evaluation of the new desalination membranes with improved antifouling properties are being reviewed.

# 2 Membrane

Membrane is a semipermeable barrier through which the feed components pass selectively. In other words, membrane is a selective barrier which allows some components of feed to pass through but stops the others (Ho & Sirkar, 2012). The most common examples of membranes are the inner membrane of egg and also phospholipid bilayer cell membrane that surrounds every cell in our bodies. The membrane thickness can be as small as 100 µ to several millimeters. In the membrane processes, the part of the feed which passes through the membrane is named permeate, and the other part is called residual or retentate. Permeate or retentate can be considered as the membrane product, separately (Scott, 1995). The required driving force in the membrane processes can be the difference in concentration (°C), pressure (°P), temperature (°T), and electric potential (°E) as shown in Fig. 1. According to the transport mechanism, membranes are classified into solution-diffusion (non-porous) membranes and porous membranes. In solution-diffusion membranes, permeation of a component depends on its permeability that is related to diffusivity and solubility of the component within the membrane material, directly. In porous membranes, based on the size-exclusion mechanism, permeation flux depends on the applied pressure and membrane pores diameter (Scott, 1995; Tofighy & Mohammadi, 2020a).

Membranes can be fabricated using inorganic materials or organic materials (polymers) or a combination of both as polymer nanocomposite membranes (PNMs) (Bassyouni et al., 2019). This combination overcomes the drawbacks of both polymeric and inorganic membranes and enhances separation performance and antifouling properties (Bastani et al., 2013; Goh et al., 2016; Wong et al., 2015).



**Driving force:**  $\Delta P, \Delta C, \Delta T, \Delta E$ 

Fig. 1 Membrane separation processes with different driving forces

Generally, the membrane desalination efficiency is assessed by two parameters including salt rejection and water permeation flux. The permeation flux passing through the membrane is defined as the mass of permeate leaving the effective membrane surface per unit of time and obtained using the following equation:

$$J = \frac{m}{A.t} \tag{1}$$

where t is the permeation time (h), m is the permeate mass (kg), and A is the membrane effective surface area  $(m^2)$ .

Rejection is calculated using the following equation:

$$R(\%) = \left(1 - \frac{C_{\rm p}}{C_{\rm f}}\right) \times 100 \tag{2}$$

where  $C_{\rm p}$  and  $C_{\rm f}$  are the salt concentrations of permeate and feed, respectively.

# 3 Membrane Fouling

The unwanted accumulation, precipitation, and surface adsorption of foulants on the surface of membrane or inside the membrane pores are called fouling (Mansouri et al., 2010; Matin et al., 2011). Fouling is very important challenge in membrane processes with pressure driving force including RO, MF, UF, and NF (Goosen et al., 2005). The lifetime and performance of the membrane depend on its fouling. Over time, fouling can worsen the membrane performance (reduce flux and rejection) and increase pressure drop, resulting in higher maintenance and operating costs. Fouling rate can be influenced by several parameters including membrane type, concentration and nature of foulants, hydrodynamics of membrane module, and surface characteristics, pore size distribution and materials of membrane (Anand et al., 2018; Jiang et al., 2017; Malaeb & Ayoub, 2011).

Fouling may be irreversible or reversible, hydraulically. In irreversible fouling, the foulants are bonded on the membrane surface and pores tightly and therefore can be eliminated only by chemical cleaning. In reversible fouling, the foulants are loosely attached to the surface of membrane and can be eliminated by backwashing, controlling flow velocity or chemical cleaning (Anand et al., 2018; McCloskey et al., 2012; Mustafa et al., 2014).

Fouling is classified into internal fouling and surface fouling. Surface fouling is usually more reversible than internal fouling and can be controlled more easily via using chemical cleaning or enhancing feed water hydrodynamic conditions (Hoek et al., 2008; She et al., 2016). Nevertheless, depending on the foulants interactions with the membranes and the feed compositions, both internal and surface foulings can be irreversible (Jiang et al., 2017). It should be mentioned that the fouling mechanism of NF and RO (high pressure-driven membranes) is different from that of UF and MF (low pressure-driven membranes). Due to the relatively compact nature of RO and NF membranes compared to the porous nature of UF and MF membranes, surface fouling is more frequent for NF and RO membranes, while internal fouling is more common for MF and UF membranes (Gao et al., 2015; Jiang et al., 2017).

For porous membranes, pores are the membrane active area. Therefore, fouling mechanism of the porous membranes is related to their pores. Generally, for the porous membranes, four fouling mechanisms can be considered including internal pore blocking, partial pore blocking, complete pore blocking, and cake filtration as can be observed in Fig. 2. If the pores of the porous membrane are partially blocked due to fouling, the effective surface of membrane and as a result permeation flux of the membrane decreases. However, if a cake layer forms on the surface of membrane, the cake layer acts as an additional resistance which is in series with the membrane resistances (Belfort et al., 1994; Gao et al., 2011; Goosen et al., 2005; Zambare et al., 2017).

Fouling is caused by complex physical and chemical interactions between various foulants in the feed and the surface and pores of the membrane (Field, 2010). Fouling may be as following forms:

- Adsorption: Formation of a monolayer of foulants on the surface of membrane due to specific interactions between the surface of membrane and foulants that leads to an additional hydraulic resistance.
- **Pore blockage**: Partial closure or complete closure of the membrane pores during filtration that results in a reduction in permeation flux.
- **Cake formation**: Growing foulants deposition layer by layer on the surface of membrane as an additional hydraulic resistance (cake resistance).

**Fig. 2** Fouling mechanisms of porous membranes: **a** internal, **b** complete and **c** partial pore blocking, and **d** cake filtration



• Gel formation: Concentration polarization of a certain macromolecules solution (e.g., a concentrated protein solution) that leads to gel formation on the surface of membrane.

Fouling is also classified into inorganic fouling, organic fouling, colloidal fouling, and biofouling, in terms of the foulants types (Chang et al., 2019; Goh et al., 2018; Jiang et al., 2017).

# 3.1 Inorganic Fouling (Scaling)

Crystallization or precipitation of inorganic ions such as magnesium, calcium, sulfate, phosphate, and carbonate on the membrane surface is called inorganic scaling (Chang et al., 2019; Chun et al., 2016; Yang & Huang, 2010). During the desalination processes with pressure driving force, concentration of the dissolved salts increases 4-10 times (causing supersaturated solutions near the surface of membrane), which leads to the inorganic minerals crystallization on the surface of membrane (Antony et al., 2011; Karabelas et al., 2017). Stages of inorganic fouling in RO membrane are shown in Fig. 3. Nucleation sites availability is the most important factor in the scale formation. After nucleation and microcrystals formation that grows over time, the mineral scales deposit on the surface of membrane that damages the membrane selective layer and impairs the water transport. Also, the process conditions and the membrane morphology play very critical roles in scaling. Concentration polarization causes higher salt concentration at the surface of membrane, in high pressure membrane systems (Goh et al., 2018).

Inorganic fouling is classified into non-alkaline, alkaline, and silica-based scaling. Calcium carbonate as the most common type of alkaline-based scalant can exist in the form of bicarbonate and calcium ions in seawater, brackish water, and industrial water. Calcium sulfate is the most commonly non-alkaline-based scalant as a limiting factor to desalinate solutions with high concentration of phosphate ions. Membrane cleaning of the non-alkaline-based fouling is very challenging compared to alkaline-based fouling. Therefore, to prevent the sulfate scaling, the feed concentration should be below the saturation level. In inorganic fouling, the membrane cleaning through back-washing as a physical approach is not favorable due to the difficult removing of the scalants from the membrane surface and pores. Therefore, to delay the formation of crystals and inhibit the scaling occurrence, antiscalant addition is recommended. However, it should be mentioned that the chemicals addition is ineffective for the silica scaling removing. Therefore, it is more desirable to prevent the formation of silica through separate pretreatment processes (Goh et al., 2018; Landaburu-Aguirre et al., 2016; Lyster et al., 2010).

#### 3.2 Organic Fouling

Organic materials including oil, grease, proteins, polysaccharides, lipids, amino acids, nucleic acids, cell components, organic acids, and fulvic and humic acids can react and deposit on the surface of membrane and block the membrane surface pores as organic fouling (Chang et al., 2019; Tanudjaja et al., 2017). Bovine serum albumin (BSA) as a protein, alginate as a polysaccharide, and humic acid as natural organic matter (NOM) have received research interests as RO organic foulants. It should be mentioned that four important factors affecting organic fouling are interactions between foulant–membrane surface and foulant–foulant, feedwater chemistry as well as molecular weight of organic materials. Removing of the organic fouling is usually hard. Because, the dissolved organic materials can form



Fig. 3 Various stages of inorganic fouling in RO membrane (Piyadasa et al., 2017)

complex structures by combination with other substances (Ding et al., 2016; Naidu et al., 2014; Shen & Schäfer, 2015). Compared to organic matters with high molecular weight, elimination of low molecular weight organic materials through the conventional pretreatment methods is more difficult (Fabris et al., 2008; Jiang et al., 2017).

# 3.3 Colloidal Fouling

Inorganic or organic colloids are fine suspensions with particle size range from a few (nm) to a few (µm) which physically block the membrane pores and/or blind the membrane surface and thus hinder water transport through the membrane by developing a cake layer. Membranes can reject most suspended colloidal particles in the feed solution in the form of a cake layer as colloidal fouling (Al-Amoudi & Lovitt, 2007; Chang et al., 2019; Hwang & Ku, 2014). Aluminum silicate minerals, iron oxides/hydroxides, and silica are the major inorganic colloidal foulants in water, while proteins, polysaccharides, as well as some NOMs are the main organic colloidal foulants in water (Tang et al., 2011). The colloids shape, size and charge, interactions between foulants and surface of membrane and also the feed water characteristics affect colloidal fouling, significantly. The colloidal fouling forms a cake layer on the surface of membrane that leads to a serious concentration polarization and makes an additional hydraulic resistance, which increases the operating pressure and decreases the membrane permeate flux (Ang & Elimelech, 2007; Jiang et al., 2017).

# 3.4 Biofouling

One of the most challenges in the membrane technologies is biofouling that leads to lower membrane selectivity, less permeation flux, shorter membrane lifetime, and higher energy consumption. In biofouling, colonies of bacteria and other microorganisms grow on the surface of membrane. In other words, the microorganisms adhesion and proliferation on the surface membrane and the formation of biofilm are named as biofouling. The complexity of the biofouling mechanism is greater than the other foulings. The microorganisms produce extracellular polymeric substances (EPS) during their metabolism processes that accumulate on the surface of membrane, results in a sticky gel-like biofilm layer formation on the surface of membrane that decreases permeation flux, significantly. EPS can have different substances including proteins, polysaccharides, lipoproteins, glycoproteins or nucleic acids, and lipids, depending on bacteria community and different water environment (Anand et al., 2018; Hegab & Zou, 2015; Hoek et al., 2008; Mahmoud et al., 2015). Several main steps of the biofilm development on the surface of membrane are as follows:

- Attachment of the feed microorganisms on the surface of membrane
- Growing of the microorganisms colonies on the surface of membrane
- Production of EPS by the microorganisms and their accumulation on the surface of membrane
- Formation of the sticky gel-like biofilm layer.



Many other factors can also affect biofouling including the membrane surface characteristics, the microorganisms properties, and the membrane surface–microorganisms interactions, as well as the operational conditions as shown in Fig. 4, briefly. Also, the time-dependent stages of the biofilm formation on an impermeable membrane surface are presented in Fig. 5.

Biofouling mitigation is very important. So far, several attempts have been performed to mitigate the membrane biofouling as given as follows:

- Removal of nutrients for microorganisms from feed water in order to limit their growth.
- Continuous dosing of disinfectants such as chlorine, chlorine dioxide, ozone, antiscalants, and acids.
- Fabrication of engineered polymeric membranes:
  - Improving hydrophilic properties of membranes by incorporating hydrophilic modifiers.
  - Incorporating biocidal materials into membranes.

# 4 Rectifying Approaches

The influence of fouling can be rectified by several approaches that are divided into direct and indirect methods as shown in Table 1. For RO and NF membranes with very small or even no pores on their surfaces, back-washing cannot be used to control the membranes fouling, and chemical cleaning and the membrane surface modification can be more effective (Zhao & Yu, 2015).

# 4.1 Membrane Cleaning

Regular washing of membranes during desalination processes is very important. There are several ways to clean the membranes, including physical and chemical methods. The efficiency of these methods in the membrane cleaning is assessed by flux recovery. In the chemical method, the **Table 1**Direct and indirectmethods to reduce and/or preventfouling (Field, 2010)

Indirect methods	Direct methods
More hydrophilic membranes fabrication	Pulsed or reverse flow
Modification of membrane surface	Turbulent promoters
Pretreatment by filtration	Stirred cells with rotating blades close to membranes
Optimum operating conditions selection	Vibrating or rotating membranes
Appropriate mode selection	Ultrasonic enhancement
	Applying external fields
	Periodic backwash with gas or permeate
	Periodic cleaning • Mechanical cleaning • Chemical cleaning • Hydraulic cleaning

choice of chemical type is very important and based on the types of fouling and foulants and the economic considerations are determined. The most important chemicals used in chemical cleaning are acids, bases, and surfactants. Acids such as hydrochloric acid (HCl) and sulfuric acid  $(H_2SO_4)$ are used to remove inorganic fouling, while alkaline solutions such as solution of sodium hydroxide (NaOH) are used to organic and biological foulings removal. Operating conditions such as time, flow rate, and temperature can influence the efficiency of the process of membrane cleaning. Surfactants can also be used effectively to clean the membranes by having both hydrophilic and hydrophobic groups. A common surfactant used in chemical cleaning is sodiumdodecyl sulfate (SDS) that can be used effectively to remove organic and colloidal foulings. In the physical method, the membranes are washed regularly with water. A combination of physical and chemical methods is used to clean the membranes, practically (Filloux et al., 2015; Jiang et al., 2017; Varin et al., 2013; Yang et al., 2013).

# 4.2 Membrane Modification

Polymeric membranes are usually prepared by hydrophobic polymers including polysulfone (PSf), polyethersulfone (PES), and polyvinylidene fluoride (PVDF) with good thermal, mechanical, and film-forming properties. However, hydrophobic membranes have tendency to fouling. The membrane hydrophilicity can be enhanced by modification of the membrane material (e.g., inorganic nanomaterials incorporation into membrane matrices) to improve the antifouling properties of membrane (Zambare et al., 2017). Increasing the membrane hydrophilicity causes strong attraction between the surface of membrane and water molecules leads to a protective water layer formation on the surface of membrane which prevents adhesion of foulants on the membrane surface and as a result decreases the membrane fouling as shown in Fig. 6 (Ahmad et al., 2013). An efficient way to enhance the membranes antibiofouling properties is incorporation of biocidal nanoparticles into the membranes materials (Rahaman et al., 2014). Copper and silver nanoparticles have been widely used to microorganisms inactivation as well as prevention of biofilm formation on the surface of membrane. However, as the antimicrobial activity of these nanoparticles decreases over time, these nanoparticles are not suitable for long-term applications (Celik et al., 2011). Therefore, recent research has focused on the applications of carbon nanomaterials to disable the biofoulants on the membrane surface (Anand et al., 2018).

Among carbon nanomaterials, CNTs have been shown great potential in enhancing hydrophilicity of the membrane surface and antifouling properties as shown in Fig. 7. The antifouling properties of CNTs are related to their hydrophilic functional groups and smooth and frictionless interior walls that can accelerate the water molecules transport through the membranes. CNTs with biocidal properties can break the microorganisms cell membranes and thus microorganisms inactivation and/or death can take place (Farahbakhsh et al., 2017; Tofighy & Mohammadi, 2012b).

In recent years, GO nanosheets with abundant oxygen-containing functional groups on their structure have received increasing attention to improving the membranes antifouling properties. Sharp edges of the GO nanosheets act as cutters which physically damage the cellular membranes of the microorganisms. The release of intracellular content of the cell membranes leads to the cell membranes death as shown in Fig. 8 (Perreault et al., 2014; Tofighy & Mohammadi, 2020b).

Membrane surface chemistry has also a major role in reducing the membrane fouling. Increasing smoothness of the surface of membrane also decreases the foulants deposition on the membrane surface and therefore improves the membrane antifouling properties. Therefore, modification of the membrane surface can decrease the fouling potential, significantly (Gao et al., 2011; Zambare et al., 2017).



Fig. 6 Hydration layer formation on the membrane surface (Ahmad et al., 2020)



Various methods including blending, coatings, grafting have been proposed to enhance the antifouling properties of the polymeric membranes (Ahmad et al., 2013; Belfort et al., 1994; Bruggen, 2009; Kumar et al., 2015; Mohammadi et al., 2003a, 2003b; Nabe et al., 1997; Pieracci et al., 2002; Shi et al., 2014; Zambare et al., 2017; Zhao et al., 2013).

# 4.2.1 More Hydrophilic Membranes Fabrication (Blending)

In recent years, polymer nanocomposite membranes (PNMs) with having a combination characteristics of both polymeric and inorganic membranes have attracted considerable

attention (Peng et al., 2007; Smaihi et al., 1996; Souza & Quadri, 2013). PNMs have been fabricated through incorporation of inorganic nanofillers including metal nanoparticles (CaCO<sub>3</sub>, Ag, SiO<sub>2</sub>, TiO<sub>2</sub>, and ZrO), carbon nanomaterials (carbon nanotubes (CNTs) and graphene oxide (GO)), and hybrid nanoparticles (GO-SiO<sub>2</sub> and GO-TiO<sub>2</sub>) with superior separation performance into polymeric matrices with well processability (Bastani et al., 2013; Goh et al., 2016; Mallakpour & Naghdi, 2018; Papageorgiou et al., 2017; Wong et al., 2015). PNMs can be fabricated in two configurations: mixed matrix membranes (MMMs) and thin-film nanocomposite (TFN) membranes.



Fig. 8 Antifouling effect of GO (Perreault et al., 2014)

#### Mixed Matrix Membranes (MMMs)

Tremendous improvements have been made in efficiency of the membrane process in terms of flux, rejection, antifouling characteristics and mechanical, thermal, and chemical stability by mixed matrix membranes (MMMs) fabrication as a combination of polymeric and inorganic membranes (Ahmad et al., 2011; Wang et al., 2013). MMMs due to the use of processability of polymeric membranes and unique of inorganic properties nanomaterials including hydrophilicity, excellent mechanical stability, and antimicrobial properties have become the focus of many researchers (Esfahani et al., 2018; Shirazi et al., 2011; Vinh-Thang & Kaliaguine, 2013). In MMMs, various inorganic nanomaterials as nanofillers are directly incorporated into polymeric matrices which can improve membranes physicochemical properties including hydrophilicity, thermal, chemical and mechanical stability, porosity, charge density and antifouling properties and overcome the trade-off relationship between permeability and selectivity during desalination (Badawi et al., 2014; Kim & Bruggen, 2010; Pendergast & Hoek, 2011; Robeson, 1991; Wong et al., 2015).

The MMMs fabrication is similar to regular polymeric membranes fabrication (Guillen et al., 2011). The selection of proper technique for the polymeric membranes fabrication is restricted by the choice of polymer and the desired membrane structure. The most commonly used process for polymeric membranes fabrication is phase inversion. Using this technique, a variety of morphologies from very porous structure membranes that are suitable for MF applications to denser RO membranes can be obtained. In MMMs fabrication, controlling nanofillers distribution within the polymer matrices is essential (Lalia et al., 2013; Strathmann, 1985; Wienk et al., 1996; Witte et al., 1996). Membrane materials (polymer, solvent, non-solvent, and nanomaterials) selection

is important in fabricating membranes with desirable properties and structure (Boom et al., 1992; Choi et al., 2006; Smolders et al., 1992).

Rahimpour et al. (2012) chemically treated CNTs using 1,3-phenylenediamine (mPDA) and strong acids (H<sub>2</sub>SO<sub>4</sub>/ HNO<sub>3</sub>) to synthesize amine-functionalized CNTs. They reported preparation of amine f-CNTs/PES MMMs via the phase inversion method with the enhanced antifouling properties and BSA rejection. Gul et al. (2017) reported fabrication of cellulose acetate (CA)/CuO/Ag<sub>2</sub>O/PES MMMs with the enhanced antibacterial properties against Escherichia coli. Chen et al. (2013) reported fabrication of halloysite loaded chitosan (CS)/Ag/PES MMMs with the enhanced antibiofouling properties against E. coli and Staphylococcus aureus than the neat PES membrane. Vatanpour et al. (2011) reported fabrication of oxidized CNTs/PES MMMs via the phase inversion method with the superior antifouling properties. Their results demonstrated that due to migration of the f-CNTs with negatively charge acidic functional groups to the surface of membrane during the phase inversion process, the hydrophilicity of membrane enhances. In another work (Vatanpour et al., 2014), they reported preparation of amine f-CNTs (NH2-CNTs)/PES MMMs via the phase inversion method. In another work (Vatanpour et al., 2012), they coated oxidized CNTs by anatase titanium dioxide (TiO<sub>2</sub>) nanoparticles and reported preparation of CNTs-TiO<sub>2</sub>/PES MMMs with superior antibiofouling properties. Peydayesh et al. (2018) reported preparation of positively charged Triethylenetetramine (TETA) f-CNTs/PES MMMs via the phase inversion method with enhanced antifouling properties. In another work (Peydayesh et al., 2019), they reported preparation of positively charged hyperbranched polyethyleneimine (HPEI) functionalized CNTs/PES MMMs via the phase inversion method with the improved antifouling properties and durability. Yu

et al. (2012) reported fabrication of SiO<sub>2</sub>@N-Halamine/PES MMMs via the phase inversion method with the improved antifouling and antibiofouling properties. Shaban et al. (2018) incorporated titanium dioxide nanoribbons (TNRs)/CNTs as nanofiller into PES matrix and prepared TNRs/CNTs/PES MMMs via the phase inversion method with improved antifouling properties. Zinadini et al. (2017) incorporated ZnO/CNTs as nanofiller in PES matrix to prepare ZnO/CNTs/PES MMMs with the well antibiofouling characteristics. Koseoglu-Imer et al. (2013) reported fabrication of Ag/PSf MMMs with the improved antifouling properties than the neat PSf membrane.

#### Thin-Film Nanocomposite (TFN) Membranes

Thin-film composite (TFC) membranes as standard polymeric membranes have received considerable attention worldwide. TFC membranes are usually composed of a thin selective polyamide (PA) layer (with thickness of  $0.1-1 \mu m$ ) deposited on a microporous support membrane (with thickness of about 50 µm) and widely applied to water treatment applications (Dong et al., 2015; Samadi et al., 2014; Sharma & Chellam, 2005). Trade-off between permeability and rejection is an important challenge facing TFC membranes. A new generation of TFC membranes, where nanomaterials have integrated as nanofillers within the surface thin layer and/or the support layer, is called thin-film nanocomposite (TFN) membranes. TFN membranes are commonly prepared via the interfacial polymerization (IP) technique which is based on polycondensation reaction between two-functional amine and tri-functional acid chloride as monomers (Morgan & Kwolek, 1959; Zargar et al., 2017). Recently, the nanotechnology advancement has opened the doors to fabricate TFN membranes with improved properties. During the TFN membranes fabrication, the nanoparticles are introduced into the structure of the thin active layer and/or the support layer that enhances hydrophilicity, surface charge density, and antifouling properties without decreasing the efficiency of membranes separation (Lau et al., 2015). Optimizing ability of the TFN membranes skin and support layers discretely is the main advantage of TFN membranes than MMMs, and this allows for more diverse membranes to be designed (Lau et al., 2012; Sorribas et al., 2013).

Chan et al. (2016) incorporated zwitterion functionalized CNTs into PA TFN membranes to improve surface biofouling resistance and membrane selectivity. Their results showed that compared to the neat membrane, the antifouling properties of the fabricated membranes were significantly improved. Son et al. (2015) reported preparation of a TFN membrane with f-CNTs containing support layer via the phase inversion and IP methods and studied the effect of f-CNT on the performance of membrane. Their results showed that compared to the neat TFC membrane, TFN membrane with more negatively charged surface have 50% higher initial water flux and higher fouling resistance without sacrificing NaCl rejection. Azelee et al. (2017) reported fabrication of PA TFN membrane incorporated with CNT-TNT hybrid via the IP method over porous commercial PSf UF support for desalination application. It was found that the properties of the membrane surface in terms of surface charge, surface roughness, and contact angle are improved with addition of the acid-treated CNT-TNT as filler to the PA membrane layer. Zarrabi et al. (2016) reported fabrication of a modified TFC membrane by IP between trimesoyl chloride (TMC) and piperazine (PIP) monomers and NH<sub>2</sub>-CNTs as hydrophilic modifier on asymmetric porous PSf support. Their results showed that the fabricated modified membranes have higher hydrophilicity and smoother surface and improved fouling resistance compared to the neat membrane. Lee et al. (2017)used CNTs to tune the microporous PSf membranes surface properties (surface porosity) that play an important role in RO TFC membranes performance. Compared to the neat PA-TFC membrane, water permeability of the PA-TFC membranes fabricated with CNTs/PSf nanocomposite supports increased up to 35% (without salt rejection reduction). Shen et al. (2013) reported preparation of PA TFN membranes containing PMMA f-CNTs via the IP method. It was found that the PMMA f-CNTs significantly improve selectivity and permeability of the prepared membranes.

Farahbaksh et al. (2017) demonstrated that CNTsembedded TFC membranes exhibit better antifouling performance. He et al. (2015) reported fabrication of TFC membranes with the excellent antibiofouling performance by incorporating GO nanosheets into PA films through the IP method.

#### 4.2.2 Modification of the Membrane Surface

#### Coating

Physical adsorption or hydrophilic-hydrophobic interactions can be used to membranes coating to improve their antifouling properties. Wang et al. (2010) coated chitosan on polypropylene non-woven membrane using glutaraldehyde (GA) as cross-linking agent to improve the antifouling and antibacterial properties. Surface smoothness and hydrophilicity were improved, and therefore, foulants adhesion to the membrane surface was reduced. Ben-Sasson et al. (2014) to endow the antimicrobial properties, coated copper nanoparticles on TFC PA RO membranes by the dip-coating method. The copper nanoparticles-coated PA membrane with improved hydrophilicity and roughness showed the superior antibacterial properties against various microorganisms. Silver nanoparticles (AgNPs) were also used to coat on the membranes surface by various research groups to improve the membranes antibiofouling properties (Dong et al., 2017; Huang et al., 2016; Liu et al., 2015). Reis

et al. (2015) using low-pressure plasma technique, introduced amine functional groups onto the commercial PA-TFC membranes surface, and then bonded AgNPs to the amine-rich TFC membranes. The fabricated membranes showed the enhanced antimicrobial properties.

#### Grafting

Membrane surface chemistry plays a major role in reducing fouling. The surface of membrane can be modified chemiby attaching cally (grafting) macromolecules and co-polymers by various treatment methods including the plasma and UV irradiation treatments (Chang et al., 2019). Grafting of hydrophilic groups such as carboxylic acid group, hydroxyl group, zwitterionic betaine group, and ethylene glycol group to the membrane surface improves the antifouling properties, significantly (Holmlin et al., 2001). Chou et al. (2016) reported grafting of a zwitterionic sulfobetaine-based copolymer onto a substrate. Their results showed that adsorption propensity of fibroblast cells, bacterial cells, and human blood cells to the surface of membrane decreases due to improving the membrane surface hydrophilicity. Mauter et al. (2011) treated PSf membrane surface with plasma to activate carboxylic acid, carbonyl, and hvdroxvl functional groups. Then. polvethyleneimine (PEI) encapsulated AgNPs were grafted on the plasma-treated PSf membrane surface by electrostatic interactions to improve the antimicrobial properties. Li et al. (2013) grafted PVDF membrane by poly(acrylic acid) (PAA) and then immobilized AgNPs in the grafted membrane to improve hydrophilicity and the antifouling and antibiofouling properties. Zhang et al. (2013) grafted 3-allyl-5,5-dimethylhydantion (ADMH) on commercial aromatic PA RO membranes and cross-linked them by N,N'-Methylene bis (acrylamide) to improve the antifouling properties. Yu et al. (2009) grafted (3-(methacryloyl amino) propyl)-dimethyl(3-sulfopropyl)ammo-niumhydroxide inner salt (MPDSAH) on PSf membrane to antifouling properties improving. Their results showed that with increasing the grafting degree, the antifouling properties enhance. Wang et al. (2014) also UV-grafted N-(5-methyl-3-tert-butyl-2hydroxy benzyl) acrylamide (MBHBA) and acrylic acid (AA) on PSf membrane surface to improve the antifouling and antibiofouling properties. They found that the modified membrane shows enhanced hydrophilicity and antifouling and antibiofouling properties than the unmodified membrane.

# 5 Conclusion

Lack of clean water can negatively affect human health, economic growth and industrial productions, worldwide. Membrane-based desalination is known as one of the promising ways to address the global clean water scarcity challenge. Polymeric membranes with desirable properties such as low cost, high processability, and excellent mechanical strength are commonly used in desalination. One of the most important challenges in using polymeric membranes in the desalination processes is fouling that significantly reduces membrane flux and salt rejection and also increases operating costs. Fouling is classified into organic fouling, inorganic fouling, colloidal fouling, and biofouling, in terms of the foulants types. There are several ways to control fouling in desalination process including membrane surface modification, novel membrane development, membrane monitoring, pretreatment, and membrane cleaning. Increasing the membrane hydrophilicity causes strong attraction between the surface of membrane and water molecules leads to a protective water layer formation on the surface of membrane which prevents foulants adhesion and therefore reduces the membrane fouling. Using carbon nanomaterials, especially CNTs and GO can create high permeation flux and salt rejection and also delay membrane fouling. An efficient way to enhance the membranes antibiofouling properties is biocidal nanoparticles incorporation into the membranes materials. Membrane surface chemistry also plays a major role in reducing the membrane fouling. Therefore, modification of the surface of membrane via coatings and grafting methods can decrease the fouling potential, significantly. Researchers have devoted their efforts to develop new desalination membranes with low fouling tendency. It is predicted that in the coming years, many advances will be made in the field of fabricating membrane desalination and solving fouling challenge.

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# **Zeolite Membranes for Desalination**

# Catia Algieri, Enrico Drioli, and Francesca Macedonio

# Abstract

Today, membrane technology is largely utilized in desalination process. Saline waters are generally treated with polymeric membranes. However, the Achilles heel of the membrane processes used in water treatment remains fouling. The latter causes flux decline in the time and so a lowering in productivity and an increase in energy consumption. Therefore, a significant interest was devoted to the recourse to inorganic membranes thanks to their excellent thermal and chemical resistance and high mechanical stability. Recently, the scientific interest was focused on zeolite membranes which offer the advantages of the inorganic ones in addition to a well-defined pore size at molecular level: these characteristics make them appropriate for separation processes. In this chapter, initially, the zeolite properties and their application at industrial level will be dealt with. Subsequently, the main methods for the zeolite membrane preparation will be discussed. Finally, the use of these membranes in desalination will be deeply presented.

# Keywords

Zeolite • Zeolite membrane • Desalination • Membrane distillation

Abbreviat	ions
RO	Reverse osmosi
NF	Nanofiltration
MD	Membrane disti
LTA	Linde type A
MFI	Mobil-Type Fiv
TPAOH	Tetrapropylamn

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ZSM-5	Zeolite Socony Mobil-5
FAU	Faujasite
DDR	Deca-dodecasil 3R
OFF	Offretite
ERI	Erionite
SAPO	Silicoaluminophosphate
GO	Graphene oxide
CNT	Carbon nanotube
MD	Membrane distillation
DCMD	Direct contact membrane distillation
AGMD	Air gap membrane distillation
VMD	Vacuum membrane distillation
SGMD	Sweep gas membrane distillation
PV	Pervaporation
MF	Microfiltration
UF	Ultrafiltration
NF	Nanofiltration
FO	Forward osmosis

# 1 Introduction

Freshwater scarcity is a worldwide growing problem aggravated by overpopulation and pollution. Only 2.5% of the global water is freshwater, whereas the rest is saline (Isaka, 2012). In this scenario, water desalination represents an interesting route for obtaining drinking water or water for agricultural and industrial use for overcoming the water shortage (Xia et al., 2020). Traditional desalination processes for obtaining freshwater present very high operation and capital costs (Alkaisi et al., 2017; Youssef et al., 2014).

Today, membrane technology governs the industry of sea and brackish water treatment owing to their high-energy efficiency (Cay-Durgun & Lind, 2018). Various membrane processes were employed for saline waters desalination, such as reverse osmosis (RO), nanofiltration (NF) and membrane distillation (MD) (Cay-Durgun & Lind, 2018). These

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processes offer several advantages such as easy operation, easiness of process automation and high productivity in perfect agreement with the process intensification strategy (Drioli & Curcio, 2007; Drioli et al., 2011). Polymeric membranes are widely used in water desalination at industrial level owing to their well-known interesting properties (Bassyouni et al., 2019). The main challenge in the polymeric membrane field is the fouling. In particular, it determines the deposition of organic (colloids, proteins, polysaccharides, etc.), biologic (viruses, algae, bacteria and other microorganisms) and inorganic constituents (e.g., salts) on the membrane and into its porous structure (Saqib & Aljundi, 2016). This phenomenon significantly reduces the flux and the quality of the permeate. Consequently, there is a poor plant operation with the necessity to pretreat the feed, to repeat membrane cleaning, and inevitably, there is a reduction of the membrane life (Goh et al., 2018). On the other hand, inorganic membranes for their high thermal, mechanical and chemical stabilities are appropriate for water treatment and desalination process (Fard et al., 2018) and can operate when polymeric membranes fail. Inorganic membranes comprise different materials (e.g., ceramic, silica, zeolite, carbon and palladium) and have dense and porous structures. Alumina, silica, zirconia, titania and carbon membranes are mainly used in wastewater treatment and desalination (Verweij, 2012). Lately, the attention was also focused on the zeolite membranes application in desalination process (Drobek et al., 2012; Garofalo et al., 2014, 2016; Zhu et al., 2014a). These membranes present all the typical characteristics of the inorganic ones, but in addition, also pore size at molecular scale and so they are appropriate for separating species with very similar dimension. These membranes, for their peculiar properties, could be used in gas separations, desalination, pervaporation as well as in membrane reactors and sensors. However, their wide industrial application is hindered by high cost and the presence of defects in the zeolite layer. In fact, nowadays, only Linde Type A (LTA) zeolite membranes are commercialized and used for the dewatering of different organic solvents (Garofalo et al., 2014). Consequently, many other efforts must be made in order to have a wide commercialization of zeolite membranes to be used for separations of gaseous and liquid species.

In this chapter, the application of zeolite membranes in desalination process is reported and deeply discussed. The zeolite properties and application at industrial level are also dealt with. In addition, the principal methods used for the membrane preparation are discussed.

#### 2 Zeolites: Properties and Applications

Zeolites are aluminosilicate crystalline materials with canals and cavities. The presence of the tetracordinated aluminum confers to the structure negative charges balanced with metal cations (Armbruster & Gunter, 2001; Pavelic et al., 2018). For this reason, zeolites with aluminum exhibit hydrophilic properties. In fact, zeolite hydrophilicity increases by raising the aluminum amount. It is possible to modulate their adsorption capacity by changing the Si/Al molar ratio of the reaction mixture. These minerals exhibit also elevated thermal stability and chemical resistance. Today, about 40 natural zeolites are known and the most common are clinoptilolite, chabazite and mordenite, whereas over 200 are synthetic (Borai et al., 2009). Natural zeolites showed very high adsorption properties, but this capacity can decrease due to the presence of impurities (as quartz) (Borai et al., 2009); hence, the need to produce synthetic zeolites. They are synthesized by hydrothermal route in aqueous media at temperature more than 100 °C and 1 bar (Sazali et al., 2019). The main producers in the world are China, Japan, South Korea, Jordan, Turkey Slovakia, United States and Cuba (Bernhardt & Reilly, 2019). These materials display high adsorption properties, excellent ion exchange capacity and molecular sieving ability. For these peculiar characteristics, they are used as chemical sensors (Bento Ribeiro et al., 2015), as medical monitoring (Byrappa & Yoshimura, 2001), for the separation of linear from nonlinear hydrocarbons (Boettinger & Ming, 2002), in industrial air-quality monitoring and in air separation (Kosinov et al., 2016). In addition, they are exploited in veterinary and human medicine (Wang et al., 2010). About the wastewater treatment, zeolites are utilized for removing heavy metals (Tran et al., 2018; Wingenfelder et al., 2005), for reducing the ammonium excess (Zhang et al., 2016) and for adsorbing organic pollutants (Braschi et al., 2010; Peng et al., 2015). In addition, these materials are used as ion exchangers in laundry detergents and water softening (Ates & Akgül, 2016). The different industrial applications of the zeolites are reported in Fig. 1.

In arid countries, the water presents elevated salinity and therefore is not useful for irrigation. Several international environmental organizations and authorities gave guidelines for regulating the safety and quality of water for irrigation (FAO, 1992; US EPA, 2012). The zeolites are used for decreasing the amount of Na<sup>+</sup> by exploiting the cation exchange property of the zeolites. The use of zeolites in desalination process was investigated by various scientists with complete reduction of the salt content (99.99%) by



Fig. 1 Industrial application of zeolites

means of a series of columns filled with zeolites (Caputo & Pepe, 2007). However, the salt reduction depends on zeolite type and salinity of water (Ghaly Abdel & Verma, 2008). Recently, it was demonstrated the possibility to desalinate seawater by using the clinopliolite (a natural zeolite) (Wajima, 2019). The experimental results evidenced the possibility to reduce the NaCl concentration from seawater by using a Ca<sup>2+</sup>-clinoptiolite. Wibowo et al. (2017) utilized modified natural zeolite for reducing the seawater salinity; good results were obtained at 225 °C for 3 h.

# 3 Zeolite Membrane Preparation

Zeolite membranes open up the possibility to operate in a continuous way favoring the separations of liquid and gas mixtures on the basis of different adsorption properties and different molecular sizes (Tavolaro & Drioli, 1999). These membranes show both elevated stability at high temperatures

and in chemical aggressive environments, and so different research and industrial groups have focused on their synthesis since 1980 (Nair et al., 2003). However, the preparation of low-cost, reproducible and very thin (<100 nm) membranes without defects (grain boundary-free, pinhole-free and crack-free) and with both high selectivity and permeance are the biggest challenges. Indeed, the application of the zeolite membranes on large scale is limited to some zeolite topologies in pervaporation process for removing water from organic solvents (Morigami et al., 2001). Different companies industrialized the Linde Type A (LTA) zeolite membranes for solvent dehydration (Gascon et al., 2012) considering their interesting performance in terms of separation values. The performance of NaA membranes produced by Mitsui Engineering & Shipbuilding Co. for solvent dehydration is reported in Table 1. However, LTA zeolite membranes present two disadvantages: Dealumination under very strong acidic conditions (by limiting its application at pH value lower than 6) and the membrane

**Table 1** Performance of NaAmembranes produced by Mitsui inPV (Sommer & Melin, 2005)

Solvent	<i>T</i> (°C)	Feed (ethanol/water)	Separation factor	Permeate flux (kg m <sup>2</sup> h <sup>-1</sup> )	
Methanol	60	90–10	10,000	0,46	
Ethanol	70	90–10	18,000	1.12	
Isopropylalcohol	75	90–10	30,000	1.58	
n-Butanol	75	90–10	90,000	1.39	
Ethylene glycol	100	91–9	162,000	0.03	
1,4-Butandiol	99	87–13	11,000	2.23	
THF	60	90–10	12,000	1.78	

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contraction (Wenten et al., 2017). These limits are owing to the high aluminum amount in the LTA structure. Jamieson and coworkers (Jamieson et al., 2015) reported as under acidic conditions the degradation of the LTA zeolite is attributable to the removal of the aluminum from the structure.

Zeolite membranes are synthesized on supports for improving mechanical resistance because self-supported zeolitic layers are very brittle. Typically, are used flat or tubular porous supports in different materials mainly in alumina and stainless steel (Jiang et al., 2015; Ma et al., 2014; Martinez Galeano et al., 2016; Sommer & Melin, 2005).

They are very expensive compared to polymeric ones (about  $5.000 \text{ C/m}^2$ ), and this cost is due to by 50% of the cost of the support and by 50% of the chemicals. Consequently, different research groups tried to prepare membranes by using cheaper supports (Achiou et al., 2018; Bouazizi et al., 2016).

Two main methods are used for their preparation: one step (called too traditional) (Vroon et al., 1998) and secondary growth (Lovallo et al., 1998). In the first one, the porous support is in contact with a solution or gel (in an autoclave), and during the hydrothermal treatment, nuclei formation and crystal growth occur on the support. As a consequence, this method suffers from poor reproducibility owing to the difficulty of controlling the nucleation and the crystal formation. The secondary growth is more complicated than the traditional one having some steps, but at the same time, it allows better control of the zeolitic layer formation on the support, so it is more reproducible. In this method, at first, pre-synthesized zeolite nuclei are deposited on the support (having tubular or flat configuration). The seeded support is then put in autoclave in contact with a synthesis mixture and during the hydrothermal treatment both the growth of the zeolite layer and a covalent bond among crystals and support occurs, thus by determining the membrane stabilization (see Fig. 2).

Among the several procedures used for depositing a thin layer of zeolite seeds on the support (Chen et al., 2013; Chun et al., 2002; Huang et al., 2010; Pera-Titus et al., 2005; Schillo et al., 2010), the most used are dip-coating and rubbing, but they present some drawbacks. For example, during the dip coating, when the support is extracted from the suspension, the crystal seeds come off very easily (for the gravitational force action), and therefore, it is difficult to obtain continuous layers (Wang et al., 2011). The rubbing does not allow uniform seed coverage since it is made with small brushes immersed in a zeolite paste which is then rubbed inside or outside the support surface (Liu et al., 2011). When the seeding is performed by means of the filtration (mainly cross-flow or dead-end) of a zeolite suspension through flat porous supports, a more uniform layer is obtained (Pera-Titus et al., 2005). This is due to the possibility of well controlling the operating conditions of these procedures being widely consolidated membrane processes. When a tubular support is used, the cross-flow filtration is coupled with a very slow rotation of the support along its longitudinal axis for obtaining the formation of a more uniform film (Algieri et al., 2009).

The secondary growth method is more reproducible than the traditional one by allowing to separately optimize the operating conditions of nucleation and formation of the crystals. Different research groups tried to prepare oriented zeolite films with the aim to reduce the intercrystalline defects (Pham et al., 2013). For example, the b-oriented



Fig. 2 Scheme about the steps of the secondary growth method



Scheme 1 MFI structure

silicalite (Mobil-type five (MFI)) membranes (see Scheme 1) showed better separation performance by using substances as xylene isomers (Lai et al., 2003; Shete et al., 2017; Tung et al., 2011).

Different methods were developed for the preparation of MFI b-oriented membranes as Langmuir–Blodgett deposition (Rangnekar et al., 2015), Langmuir-Schaefer deposition (Rangnekar et al., 2015), dynamic interfacial assembly (Liu et al., 2010) and polymer-mediated assembly (Zhang et al., 2008). Despite all these methods, the oriented membrane

synthesis remains in the developmental phase. In the last years, significant progress was also accomplished on microwave synthesis of zeolite membranes providing an elevated reduction of the synthesis time (Li & Yang, 2008). A comparison between the actions of the microwave heating on the membrane formation with respect to the conventional one is reported in Fig. 3.

During the synthesis of the zeolite membranes, some organic molecules (called templates) are used for orienting the formation of the desired topology. For example, the template used for the MFI topology is the tetrapropylammonium hydroxide (TPAOH) (Algieri et al., 2003). When the synthesis is completed, the template removal is required because it blocks the zeolite pores. This chemical species is removed by thermal treatment (about 500-550 °C) (Algieri et al., 2003). However, this process causes the formation of defects in the zeolite layer for thermal stress owing to the two different thermal expansion coefficients of zeolite and support material. Different post-treatment methods are used for reducing the defects present in the zeolite layer as coke deposition, surface coatings, chemical vapor and liquid depositions and hydrothermal treatments (Maghsoudi, 2016).

Among the different zeolitic topologies used for the preparation of the membranes, the most studied are (MFI) and Linde type A (LTA). The first one presents interconnected channels: the right and sinusoidal channels having an opening of  $0.54 \times 0.56$  and  $0.51 \times 0.55$  nm<sup>2</sup> (see Scheme 1). In addition, it presents two structures: one



Fig. 3 Influence of traditional and microwave heating on the zeolite layer production

**Table 2**Zeolite topologies usedfor membrane preparation

Zeolite topology	Pore size (nm)	Si/Al	References
Faujasite (FAU)	0.74	1–1.5	Ikemoto et al. (2000)
Faujasite (FAU)	0.74	>1.5	Zhang et al. (2008)
Deca-dodecasil 3R (DDR)	0.36 × 0.44	(All silica)	Bergh et al. (2010)
offretite (OFF)-erionite (ERI) intergrowth	0.36 × 0.51	3–4	Shafiei et al. (2014)
Silicoaluminophosphate (SAPO) (Similar CHA structure)	0.38	-	Sun et al. (2018)
	Zeolite topology Faujasite (FAU) Faujasite (FAU) Deca-dodecasil 3R (DDR) offretite (OFF)-erionite (ERI) intergrowth Silicoaluminophosphate (SAPO) (Similar CHA structure)	Zeolite topologyPore size (nm)Faujasite (FAU)0.74Faujasite (FAU)0.74Deca-dodecasil 3R (DDR)0.36 × 0.44offretite (OFF)-erionite (ERI) intergrowth0.36 × 0.51Silicoaluminophosphate (SAPO) (Similar CHA structure)0.38	Zeolite topologyPore size (nm)Si/AlFaujasite (FAU)0.741-1.5Faujasite (FAU)0.74>1.5Deca-dodecasil 3R (DDR)0.36 × 0.44(All silica)offretite (OFF)-erionite (ERI) intergrowth0.36 × 0.513-4Silicoaluminophosphate (SAPO) (Similar CHA structure)0.38-

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containing all-silica (called silicalite) with hydrophobic properties and another containing aluminum and silicon (called Zeolite Socony Mobil–5, (ZSM-5)) with hydrophilic properties. The LTA topology has a three-dimensional pore structure (Meier et al., 2007). The LTA pore size opening depends on the extra-framework cation presents in the zeolite channels. In particular, the pore size is near 4 Å with the cation Na<sup>+</sup>; it is about 3 Å with K<sup>+</sup> and when the sodium cation is only in part exchanged with Ca<sup>2+</sup> the pore size is about 5 Å (Montanari et al., 2008). Table 2 reports the other zeolite topologies used for zeolite membrane preparation.

# 4 Zeolite Membranes in Desalination Process

The fresh water available in the world is limited and in 2025 two-third of the earth's population will be forced to live in high water stress areas (W. H. Organization, 2018). This big problem can be overcome by means of desalination technology. The highest number of desalination plants is located in the Middle East (Clayton, 2011; Isaka, 2012). Figure 4

shows the distribution of desalination plants (in percentage) in the World.

Thermal and membrane processes are the most used for saline water desalination. However, this process is very expensive considering both requests high energy for water heating and the high plant maintenance costs (Anis et al., 2019). Today, a membrane process widely used for producing clean water (appropriate for agriculture, industries and drinking) is the reverse osmosis (RO) (Macedonio, 2017). RO process exhibits different advantages with respect to the traditional one: Lower energy use associated with the utilization of inexpensive construction materials, the versatility in feed-water with excellent quality of water and high efficiency (Peñate & García-Rodríguez, 2012). In fact, the installation of RO plants has grown a lot reaching more than 90% of the total desalination plants (Macedonio & Drioli, 2017). However, fouling remains the biggest unsolved problem in this process. This is explained because in seawater are present salts, organic and inorganic substances that can be adsorbed on the membrane by lowering the process efficiency (Badruzzaman et al., 2019). Consequently, for restoring the membrane performance, cleaning procedures



are required and also an increase of the feed pressure for maintaining high flux. The fouling could be reduced by using pretreatment technologies. Very common is the use of the microfiltration/ultrafiltration process (Voutchkov, 2010).

Polymeric membranes are widely used in RO process because they are easy to prepare and at the same time inexpensive. Nevertheless, as reported before, in the polymeric membrane field, the fouling is a very critical issue. In addition, they also show high selectivity associated with low permeability and vice versa (Hailemariam et al., 2020). Inorganic membranes seem to be a good alternative to the polymeric ones in desalination process because they possess interesting performance in chemical aggressive environment, high-temperature resistance and long lifetime (Zakrzewska-Trznadel, 2013). In the inorganic field, there are many possible solutions to prepare membranes with ultra-fast water transport and/or excellent rejection: from ceramic membranes based on metal oxides to carbon-based materials, from organic metal structures to zeolites (Goh & Ismail, 2018). Various are the benefits offered by inorganic membranes:

- less membrane maintenance and replacement cost because the high chemical and thermal stability of the inorganic materials allow performing cleaning at high temperatures and with chemicals for high flux recovery.
- less biofouling problems owing to the lower susceptibility of inorganic membranes with respect to polymeric membranes (Amin et al., 2016).

Depending on the nature of the inorganic material employed for the fabrication of the membranes, the resulting structures can be either self-supporting or may need support. In both cases, the resulting membrane, be it a multilayer or a single thin sheet, is sufficiently resistant and highly permeable (Feng et al., 2015). Historically, due to their high production costs, the use of inorganic membranes has not spread on at industrial level for wastewater treatment (Lorente-Ayza et al., 2017). On the contrary, inorganic membranes have found application in those fields where polymeric membranes are either inapplicable or inefficient (such as at high temperature and/or with highly contaminated feed streams due to material instability and/or fouling problems) (Hedfi et al., 2016; Kaur et al., 2016; Lorente-Ayza et al., 2015, 2016; Mustafa et al., 2016; Rana et al., 2013; Zakrzewska-Trznadel, 2013; Zhu et al., 2014b). Different inorganic membranes used in desalination and water treatment are reported in Table 3.

Furthermore, hydrophobized inorganic membranes can find an even wider range of applications, such as in membrane distillation (MD). The latter is a relatively new membrane operation that finds applications in the sea and brackish water desalination. In MD a hydrophobic membrane is used and vapor passes through the membrane (permeate side) and all non-volatile species remain in the retentate side. Usually, MD is carried out at reduced temperatures (lower than 100 °C) with respect to traditional thermal processes, and lower pressure with respect to RO (Camacho et al., 2013). MD can be performed via, at least, four configurations as described below:

Direct contact membrane distillation (DCMD), in which the membrane puts in contact feed and permeate and both are in liquid state (Lawson & Lloyd, 1996). This configuration is the most used being very simple and the flux value is reasonable.

Air gap membrane distillation (AGMD), in which among the membrane and the condenser surface is positioned an air gap for increasing the flux and reducing the loss of the heat by conduction across the membrane (Meindersma et al., 2006). In this configuration, the flux is very low due to the air gap that determines an additional resistance to the mass transfer (Al-Zoubi et al., 2018).

Vacuum membrane distillation (VMD), in which the vacuum permits the removal of the permeate that condenses by means of an external device (Sarti et al., 1993).

Sweep gas membrane distillation (SGMD), in which a gas is utilized for the removal of the vapor (Basini et al., 1987).

In membrane distillation, the membranes must have very controlled properties (Khayet, 2011). First, the hydrophobic layer and the pore size distribution must be very narrow and with sizes ranging from nanometers to micrometers for avoiding the passage of liquid feed. In addition, the tortuosity must be very small and the hydrophobic layer very thin. Furthermore, the thermal conductivity of the membrane material must be very low for achieving high thermal efficiency. Polyvinylidene fluoride, polytetrafluoroethylene and polypropylene are polymers often employed in MD because apart from their hydrophobicity, they have sufficiently low thermal conductivity (from 0.14 Wm<sup>-1</sup> K<sup>-1</sup> of PP to 0.29  $Wm^{-1} K^{-1}$  of Polyvinylidene fluoride). In the case of inorganic membranes (characterized by high thermal conductivity), the thermal efficiency can be improved either by reducing the membrane thickness or by increasing the transmembrane flux (Al-Obaidani et al., 2008). Finally, MD membranes must possess good thermal resistance and chemical stability and with interesting permeability and selectivity values for long time.

As before described, polymeric membranes present different disadvantages as reduced stability at elevated temperatures and in the presence of aggressive substances (as acids, alkals and organic solvents) and thus short lifetime (Jamalludin et al., 2016). For that reason, the use of inorganic membranes (especially ceramic ones) in MD could be

 Table 3 Examples of inorganic membranes used for desalination and wastewater treatment

Membrane material	Membrane process	Application (feed solution)	Flux	References
$\gamma\text{-}Al_2O_3/\alpha\text{-}Al_2O_3$ inorganic hollow fiber	NF (nanofiltration)	Salt solution of 2000 ppm	$17.4 L m^{-27} h^{-1} bar^{-1}$	Wang et al. (2016b)
SiO <sub>2</sub>	PV (pervaporation)	3.5 wt% NaCl	9.5 kg m <sup>-2</sup> h <sup>-1</sup> , 6.6 kg m <sup>-2</sup> h <sup>-1</sup>	Elma et al. (2013), Elma et al. (2015a)
CoO-SiO <sub>2</sub>	PV (pervaporation)	3.5 wt% NaCl	7.7 kg m <sup>-2</sup> h <sup>-1</sup>	Elma et al. (2015b)
TiO <sub>2</sub>	PV (pervaporation)	10 wt% NaCl	$6.0 \text{ kg m}^{-2} \text{ h}^{-1}$	Yacou et al. (2015)
CMS-Al <sub>2</sub> O <sub>3</sub>	PV (pervaporation)	3.5 wt% NaCl	$25.0 \text{ kg m}^{-2} \text{ h}^{-1}$	Song et al. (2016)
Zirconium (IV)-based metal-organic framework (Zr-MOF—i.e., UiO-66) membranes on alumina hollow fibers	UF (at 10.0 bar)	Saline water solutions (containing KCl, NaCl, CaCl <sub>2</sub> , MgCl <sub>2</sub> , or AlCl <sub>3</sub> ) with concentration 0.20 wt%		Liu et al. (2015)
$\gamma$ -Al <sub>2</sub> O <sub>3</sub>	UF (at transmembrane pressure of 5 bar)	Oil/water separation	112.7 kg m <sup>-2</sup> h <sup>-1</sup>	Bayat et al. (2016)
Al <sub>2</sub> O <sub>3</sub>	MF (at transmembrane pressure of 0.15 MPa) Oil/water separation (oil concentration in water about 5 g/L)		450 kg m <sup>-2</sup> h <sup>-1</sup>	Zhong et al. (2013)
Mullite-titania composite ceramic hollow fiber	te-titania composite ceramic MF 200 w fiber emu		$150 L \\ m^{-2} h^{-1} bar^{-1}$	Zhu et al. (2016)
Commercial tubular $Al_2O_3$ membranes with nano-TiO <sub>2</sub> coating	MF (trans-membrane pressure 0.16 MPa)	Oil/water emultion (with an average droplet size of 6 $\mu$ m with 90% of the oil droplets being in the range of 0.7–20 $\mu$ m)	$320 L m^{-2} h^{-1} bar^{-1}$	Chang et al. (2014)
Graphene oxide (GO) modification on $Al_2O_3$ membrane	MF	Oil/water emulsion (with an oil concentration of 1 g $1^{-1}$ , size range of oil-droplets within 0.5–15 $\mu$ m with a mean size of 1.8 $\mu$ m)	${}^{667}_{m^{-2}}{}^{L}_{h^{-1}}{}^{bar^{-1}}$	Hu et al. (2015)
Graphene Oxide (GO)-Palygorskite Nanohybrid Membrane	Dead-end UF	Oil-in-water emulsions (100 mg or 1 g hexadecane, 100 mg pump oil, or 100 mg soybean oil and a certain amount of sodium dodecyl sulfate)	1867 L m <sup>-2</sup> h <sup>-1</sup>	Zhao et al. (2016)
Carbon nanotube (CNT) network implanted in the pores of a ZrO2Emulsified oil filtration (TMP from 1 to 2 atm)Emulsified oil surfa and c and c L of		Emulsified oil feed: 1.6 g of surfactant-sodium dodecyl sulfate and oil (concentrations of 100, 300 and 600 ppm, soluble blue dye,) to 1 L of water $26 L m^{-2} h^{-1}$		Chen et al. (2016a)
Poly(acrylic acid)(PAA)/poly(vinyl alcohol)(PVA)/glutaraldehyde(GA) assembled on to the inner surfaces of tubular Al <sub>2</sub> O <sub>3</sub> microporous substrates	NF	Congo red dye desalination	$25 \text{ Lm}^{-2} \text{ h}^{-1}$	Wang et al. (2013)
SiO <sub>2</sub>	Membrane adsorption	Methylene blue aqueous solution	$40 \text{ L m}^{-2} \text{ d}^{-1}$	Tolba et al. (2015)
ZIF-8 membranes synthesized on porous polyethersulfone (PES) supports	NF	17.5 μM Rose Bengal solution in water	90 L $m^{-2} h^{-1} bar^{-1}$	Li et al. (2017)
Reduced graphene oxide (rGO)– carbon nanotubes (CNT) hybrid membrane	NF	Aqueous solutions with nanoparticles, dyes, BSA, sugars and particularly humic acid	$\begin{array}{c} 20-30 \text{ L} \\ \text{m}^{-2} \text{ h}^{-1} \text{ bar}^{-1} \end{array}$	Chen et al. (2016b)

(continued)

Table 3 (continued)

Membrane material	Membrane process	Application (feed solution)	Flux	References
MCM-41, MCM-48	UF	Aqueous solutions with 1000 ppm $HCrO_4^-$ at pH ~ 2.35	$\begin{array}{c} 6.05 \times 10^{-8} \text{ m}^3 \\ \text{m}^2 \text{s} \cdot \text{kPa}, \\ 4.18 \times 10^{-8} \text{ m}^3 \\ \text{m}^2 \text{ s} \cdot \text{kPa} \end{array}$	Basumatary et al. (2016)
MCM-41-ceramic composite membrane	Pressure driven membrane operations (applied pressure 69– 345 kPa)	Aqueous solutions with 1000 ppm $HCrO_4^-$ at pH ~ 2.35	$1.1007 \times 10^5 \text{ m/s}$	Basumatary et al. (2015)
Analcime-C ceramic	UF	Aqueous solutions with 1000 ppm $HCrO_4^-$	$4.53 \times 10^{-8} \text{ m}^3/\text{m}^2 \text{ s}\cdot\text{kPa}$	Kumar et al. (2015)
TiO <sub>2</sub>	UF	Solutions with NaH <sub>2</sub> PO <sub>4</sub> from 0 to 1.0 mg P $L^{-1}$	-	Shang et al. (2014)
SiO <sub>2</sub>	FO	Synthetic wastewater	$69 \text{ L m}^{-2} \text{ h}^{-1}$	You et al. (2017)
$\alpha$ -Al <sub>2</sub> O <sub>3</sub> (with Fe <sub>3</sub> O <sub>4</sub> nanoparticles for NF)	Hybrid UF/NF	Stream with 70 ppb As(III) + As(V)	$75 \text{ Lm}^{-2} \text{ min}^{-1}$	Sklari et al. (2015)
TiO <sub>2</sub> -ZrO <sub>2</sub>	NF	Synthetic radioactive effluent containing cobalt chloride, strontium chloride and cesium chlorid	$\begin{array}{c} 40 \text{ L} \\ \text{m}^{-2} \text{ h}^{-1} \text{ bar}^{-1} \end{array}$	Lu et al. (2016)
SiO <sub>2</sub>	PV	Solution containing less than 1000 mg/L of ammonia	$1.7 \text{ Lm}^{-2} \text{ h}^{-1}$	Yang et al. (2014)

promising. However, ceramic membranes are hydrophilic and hence have to be modified in hydrophobic by using different methods (Ramlow et al., 2019). The hydrophobization of ceramic membranes is usually performed by direct grafting and using silane agents. The main grafting approaches are chemical vapor deposition, direct immersion and sol–gel methods (Kujawski et al., 2016; Vlassiouk et al., 2013; Yang et al., 2016). Different studies reported that the hydrophobic membranes, prepared with the first two methods, are stable up to 100 days in MD process, while those prepared with the sol–gel method are less stable especially at high temperature (Khatib & Oyama, 2013; Yang et al., 2010, 2017). Table 4 reports a list of inorganic membranes tested in the various possible MD configurations.

In the field of inorganic membranes, there are the zeolitic ones which not only exhibit high chemical and thermal stability but also well-defined pore size at molecular level and very attractive for desalination process. In particular, ions are separated from aqueous solution with a mechanism of electric and steric exclusion (Kujawski et al., 2016). A first application occurred in 2004 by Li and coworkers (2004a) where MFI zeolite (silicalite, aluminum-free) membranes were prepared on disc-shaped alumina by using the traditional method. The prepared membranes were characterized in RO desalination process. In particular, by using a NaCl solution (0.1 M) a flux of 0.112 kg m<sup>-2</sup> h<sup>-1</sup> and a rejection of 76.7% were obtained (Li et al., 2004a). In addition, a higher rejection was obtained with a divalent cation by using feed containing different ion species. Li et al. (2004b) found that ion charge, size and diffusivity of the hydrated ions influence the rejection and permeate flux in reverse osmosis process. In addition, a different mechanism controls the ion and water transport through the zeolitic pores and intercrystalline defects. The ion rejection depends on the exclusion of the big ions by the zeolite pores. Moreover, ion repulsion into the defects is owing to the strong interactions between the ion and the charged double layers that determine a limited diffusion of the ions into the defects. Other authors prepared MFI supported membranes (using disk alumina support) with different Si/Al ratios and their performance was evaluated in reverse osmosis by using sodium chloride solution as feed (Li et al., 2007). The experimental results showed flux and rejection improved with the increase of the aluminum content owing to the high surface charge and so higher affinity with water. Duke and coworkers (2009) prepared MFI membranes on alumina flat support by using the secondary growth method. They synthesized silicalite (aluminum free) and ZMS-5 (different Si/Al) membranes and then characterized them in water desalination by means of pervaporation (PV) processes. Better results were found with ZSM-5 membranes. In particular, silicalite and ZSM-5 exhibited a rejection value of 97% and 99%, respectively.

MFI (silicalite and ZSM-5) membranes synthesized on tubular alumina support (length 5 cm) were characterized for long time in water desalination (using different salt solution concentrations) by PV process (Drobek et al., 2012). Also, in  
 Table 4
 Examples of inorganic
 membranes used in MD

Sialon  $ZrO_2/T$ TiO<sub>2</sub>

PMSQ aerogel, alumina

DCMD

DCMD.

VMD

Inorganic membrane	Application	References
FAS grafted Al <sub>2</sub> O <sub>3</sub>	DCMD desalination	Ren et al. (2015)
Organosilica	VMD desalination	Chu et al. (2013)
MOF-functionalized Al <sub>2</sub> O <sub>3</sub> tube	VMD desalination	Zuo and Chung (2016)
FAS-CNT	DCMD desalination	Ali et al. (2017)
Al <sub>2</sub> O <sub>3</sub>	SGMD, DCMD, VMD	Ren et al. (2015), Gu et al. (2016), Hendren et al. (2009), Wang et al. (2016a), Fang et al. (2012), Huang et al. (2018)
ZrO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub>	AGMD	Krajewski et al. (2006)
ZrO <sub>2</sub>	AGMD	Khemakhem et al. (2013)
Titania	AGMD, DCMD	Kujawa et al. (2014)
ZrO <sub>2</sub> /Ti	VMD, AGMD	Hendren et al. (2009)
Si <sub>3</sub> N <sub>4</sub>	VMD, DCMD, SGMD	Zhang et al. (2014), Zhang et al. (2012), Wang et al. (2017a)
Sialon	DCMD	Wang et al. (2016a)
ZrO <sub>2</sub> /Ti	DCMD	Cerneaux et al. (2009)

Fan et al. (2017)

Ko et al. (2018)

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this case, ZSM-5 membranes showed better water flux for their high hydrophilicity. In addition, for all the prepared membranes, a decrease in water flux and rejection was detected by increasing salt solution concentration. However, silicalite one seemed to be more stable under the effect of temperature. In 2012, for the first time, thin membranes were prepared from natural zeolite (clinoptiolite) and used for water desalination via PV process (Swenson et al., 2012). These membranes displayed high flux and a good ion rejection values (see Table 5).

Algieri and coworkers (Donato et al., 2020; Garofalo et al., 2014, 2016) prepared silicalite membranes on tubular support (having a length of 30 cm) by secondary growth method and using the cross-flow coupled with the rotation of the support as seeding procedure. The prepared membranes were characterized, using different NaCl solution concentrations, in VMD process. For various salt concentrations (0.2-0.9 M), the ion rejection values were equal to 99.9%. The best performance was obtained by using a salt solution of 0.2 M and working at 70 °C and 120  $Lh^{-1}$  (see Table 5).

The literature data of the inorganic membranes performance in desalination are reported in Table 5.

Hydrophobic alumina membranes exhibited very interesting results. However, the ceramic membranes modified superficially for obtaining hydrophobic systems are not

stable for long-term operation, and at the same time, the different methods present reproducibility problem (Ramlow et al., 2019). Good results were also obtained with the silicalite membranes synthesized by Algieri and coworkers (Wang et al., 2013) even in comparison with the other zeolitic ones (Drobek et al., 2012; Duke et al., 2009; Li et al., 2004a, 2007; Swenson et al., 2012; Zhou et al., 2016; Zhu et al., 2014a). A possible explanation is owing to the synthesis methods used for their preparation: some membranes are produced by using the traditional method and other the secondary one (preferring dip-coating for seeding). It is well known that membranes prepared with the traditional method present high concentration of defects (Caro et al., 2000; Li et al., 2004a; Zhou et al., 2016; Zhu et al., 2014a). On the contrary, the secondary one permits the defect reduction for the optimization of both nucleation and growth of the crystals. However, the membranes prepared by using dip-coating as seeding procedure do not exhibit interesting desalination performance because the coating of the support is not uniform (Drobek et al., 2012; Duke et al., 2009). The zeolite membranes prepared by means of the cross-flow filtration coupled with the rotation of the tubular support present a very low concentration of defects (Garofalo et al., 2016). In addition, this seeding technique is reproducible and scalable (Garofalo et al., 2014). These data are

Sample	Configuration	Length (cm)	Process	Feed (g/l)	T (° C)	Flux (kg $m^{-2} h^{-1}$ )	R (%)	References
Hydrophobic Alumina	Tubular	9.2	VMD	90	70	31	99.9	Pagliero et al. (2020)
Hydrophobic Alumina	Tubular	11	VMD	30	70	30	99.9	Chen et al. (2018b)
Hydrophobic Alumina	Hollow fiber	-	DCMD	58	-	14	99.9	Chen et al. (2018a)
Hydrophobic Titania	Tubular	-	AGMD	14.6	-	2	98.5	Kujawa et al. (2017)
Hydrophobic Alumina	Tubular	-	AGMD	14.6	-	3.9	98.3	Kujawa et al. (2017)
Hydrophobic Titania	Tubular	-	AGMD	11.7	-	4.0	>98.5	Kujawa et al. (2016)
Hydrophobic Alumina	Flat sheet	-	VMD	35	-	29.3	99.9	Huang et al. (2016)
Hydrophobic Alumina	Capillary	15	VMD	29	60	4.71	99.8	Da et al. (2016)
Hydrophobic CMS	Tubular	7	DCMD	35	60	32.3	99.9	Song et al. (2016)
FAU (zeolite)	Tubular	7.5	PV	35	75	3.84	> 99.8	Zhu et al. (2014a)
MFI (ZSM5)	Tubular	5	PV	35	75	5.10	98	Drobek et al. (2012)
MFI (silicalite)	Flat	_	RO	5.8	25	0.11	90.6	Li et al. (2007)
MFI (ZSM5)	Flat	_	RO	38	70	0.55	99	Duke et al. (2009)
MFI (ZSM5)	Flat	_	RO	5.8	25	0.12	77	Li et al. (2004a)
FAU zeolite	Tubular	_	PV	35	90	5.64	99.8	Zhou et al. (2016)
Natural zeolite	-	-	PV	-	93	2.50	97.5	Swenson et al. (2012)
MFI (silicalite)	Tubular	30	VMD	35	70	19.0	99.9	Donato et al. (2020)

Table 5 Comparison with literature data

encouraging for the possibility to use zeolite membranes in desalination process. However, extensive efforts need to be required by researchers for improving their flux and selectivity, the manufacturing methodology and to reduce their production costs. The achievement of these goals will make it possible to practically apply the zeolite membranes in water desalination.

# 5 Conclusions and Future Outlook

The extraordinary research activity on the preparation of the zeolite membranes has brought a significant progress on the understanding of the separation mechanism and of the preparation of thin zeolite layer with high flux and good separation factor. However, despite the progress reached in the zeolite membrane preparation field, their application is limited to the dehydration of various organic solvents by using hydrophilic zeolite membranes (LTA zeolite) and

showing very interesting separation factors (see Table 1). Nevertheless, in gas separation processes, the zeolite commercial membranes exhibit a Knudsen diffusion mechanism by indicating the presence of defects in the layer.

These membrane types are prepared via traditional and secondary growth methods. The second one is more promising by permitting the preparation of membranes very thin, with a low defect concentration. In addition, this method is more reproducible than the traditional one since it is possible to optimize the operating conditions of each step independently.

Initially, zeolite membranes were applied in separation of gas species and organic solvent pervaporation (Bedard & Liu, 2018). In 2001 Lin and Murad (2001) via computational study demonstrated the possibility to use zeolite membranes for saltwater treatment. In 2004, Li and coworkers prepared zeolite membranes for water desalination (Li et al., 2004b). Over the years, other papers were published with better performance in desalination process. The results present in the open literature demonstrate as desalination process could represent an interesting market for the zeolite membranes though some problems have to be solved like the high membrane cost and the reduction of defects present in the zeolite layer. The achievement of these goals might place zeolite membranes in the forefront of water desalination process.

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# **Integrating Desalination Systems**

# Mashura Shammi

# Abstract

Considering the challenge of global water-energy nexus, an integration and systematic inspection of the integrated desalination is a growing research interest. This chapter aims to discuss numerously integrated and hybrid desalination technologies found in the entire globe. The tremendous research interest for integration and hybridization of different renewable technologies with desalination is being developed to achieve water security and improve seawater reverse osmosis (RO) desalination. Researchers are utilizing state-of-the-art membranes and developing nanocomposite materials to enhance the RO process. Technical advancements of renewables such as solar photovoltaics, wind, geothermal, and hybridization of photovoltaics-wind-RO for a broader capacity are being researched and developed replacing greenhouse gas-emitting fossil fuels. Multiple solar irradiation technologies have been developed that focuses on the enhancement of solar still or panel geometry, solar thermal collectors. Furthermore, research attention has also shifted on the development of portable and micro-desalination based on electrodialysis. Forward osmosis, pressure-retarded osmosis, and other hybridized system such as membrane distillation and the uses of nanomaterials have also been explored for the improvement of RO. Besides, the potential application of integrated desalination that may be applicable in the near future has also been investigated. The integration of desalination technologies is required for identifying cost-effective, commercially viable sustainable water, and energy production with less environmental impacts. It can also help the water and energy researchers, policymakers, and legislators to identify which integrated

technologies have opted for sustainable water and energy security.

# Keywords

Reverse osmosis (RO) • Forward osmosis • Renewable energy (RE) • Solar still • Solar collectors • Nanocomposites • Membranes • Micro-desalination • Electrodialysis • Membrane distillation • Hybrid

# 1 Introduction

Growing water demand and over-exploitation of surface water and groundwater in many regions caused severe water scarcity. The ever-increasing population growth and industrialization of the society's rising demand for freshwater led to faster growth of desalination system capability in many countries. Desalination technology disposes of dissolved solids and salts from saline water, seawater, industrial wastewater, or brine water (Demir & Dincer, 2017). Nearly half of the world's total water removals are consumed by electricity generation, whereas seawater desalination devours much of the electricity (Wang et al., 2019).

The conventional category of desalination methods is thermal and membrane processes. Based on phase variation, thermal desalination comprises multi-phase flash distillation, multi-effect distillation, vapor compression distillation, humidification-dehumidification. Thermal desalination processes use a lot of energy and electricity (Lee et al., 2019). On the other hand, membrane processes use and develop filtration membranes of different pore sizes with exclusive characteristics such as electrodialysis and electrodialysis reversal, RO, membrane distillation, and others (Abdelkareem et al., 2018; Gorjian & Ghobadian, 2015).

Desalination technologies can provide water security for the entire globe where fresh water is a scarcity. Nevertheless, the energy and carbon footprint of the current desalination

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technologies are incredibly high. For instance, the thermal desalination plants which work through multistage flash method or multi-effect distillation plants are typically positioned with power plants run by fossil fuels that have high environmental pollution, greenhouse gas emission, and higher carbon footprint (Abdelkareem et al., 2018; Ahmed et al., 2019). Moreover, the limited fossil fuels in the future may have a dramatic price hike if the resource is done for (El-Said et al., 2016). Besides, uncompromising power-to-water output limits are another major drawbacks that cannot satisfy the required demand pattern (Agashichev & Kumar, 2017; González-Bravo et al., 2017).

On the other hand, membrane desalination consumes less power than thermal desalination processes. However, the amount of energy use is still sizeable (Lee et al., 2019). A hybrid power system is, therefore, an adequate substitute for the supply of clean water and electricity at minimum cost to the remote regions (Wu et al., 2018).

It is here that integrated desalination methods, which are often called the hybrid system, pop up. By utilizing renewable energy (RE) resources and techniques of energy production, broad sections of traditional desalination techniques can be powered (Esmaeilion, 2020). Furthermore, in off-grid coastal zones and island regions where a substantial amount of power is needed to deliver freshwater by extraction and treatments, it may generate a complicated socioeconomical, environmental, and geopolitical crisis (Gökçek, 2018; Omar et al., 2020; Padrón et al., 2019). Water and energy policymakers need to pay more emphasis on the regional potential of using desalination processes and power generation simultaneously. This also emphasizes using locally available resources such as seawater, saline groundwater, polluted surface water, industrial wastewater, and available RE sources such as solar, wind, and geothermal power.

Therefore, the universal research activity is thriving connecting between water and electricity demand that drives the desalination plants powered by RE or by other methods such as membranes (Alhaj & Al-Ghamdi, 2019). The multitude of desalination methods is typically run by various renewable and non-renewable energy sources to produce electricity and freshwater simultaneously (Esmaeilion, 2020). Grid-less hybrid RE systems, in particular wind and solar power, have gained considerable attention (Maleki, 2018). This chapter, therefore, discusses the various integrated and hybrid desalination technologies presently at practice around the globe and the growing research interest in this field that will stimulate the mind of the researchers, water and energy policymakers, and legislators for a more sustainable future for water and energy security.

# 2 Various Types of the Integrated Desalination System

Considering the challenge of global water-energy nexus, an integration and systematic inspection of REs and different water desalination methods should be investigated (Alhaj & Al-Ghamdi, 2019). The desalination marketplace is governed by the methods of seawater RO (SWRO) globally, and the demand is rising (Ganora et al., 2019). Nevertheless, crucial technological problems linger to be unaddressed includes minimal water recovery (near about around 50%), a considerable amount of energy consumption (3-5 kWh/m<sup>3</sup>), and discharge of brine (Bitaw et al., 2019; Lee et al., 2019; Tufa et al., 2019). Among the seawater desalination processes, RO is the most effective and dependable source to produce drinking water, which uses a pressure gradient to separate dissolved solids from water. It uses a semipermeable membrane (Monnot et al., 2018). But the highly concentrated brine water from the seawater desalination plant can be damaging to the marine ecosystems (Bitaw et al., 2019). However, RO desalination centered on hybrid RE devices has surfaced as a tidier and sustainable option of desalinated water (Ganora et al., 2019; Khan et al., 2018).

Technical advancements of renewables such as photovoltaics, wind, and hybridization of photovoltaics-wind-RO for a more comprehensive capacity are being researched and developed. The substitution of fossil fuels with REs for desalination will drastically reduce carbon footprints and greenhouse gas emissions (González-Bravo et al., 2017; Khan et al., 2018). Moreover, several blending of desalination methods has been implemented, linking forward osmosis, RO, assisted reverse osmosis (ARO), microfiltration, mechanical vapor compression, and membrane distillation (Osipi et al., 2018). Hybrid integrated systems can outpace a RO stand-alone system. It can reduce water costs and alleviate the environmental discarding of brines (Choi et al., 2017). Figure 1 depicts the different system integration of desalination technologies, which includes integration of REs, integration of membrane process development, and material development.

#### 2.1 Renewable Desalination

Elimelech & Phillip, et al. (2011) asked a critical question —"Is seawater (or brackish water, wastewater, etc.) desalination justified to worldwide water scarcities?". There are various concerns with desalination methods as they possess some harmful environmental impacts. The principal power



Fig. 1 State-of-the-art research on integrated or hybrid desalination technologies

supply for seawater reverse osmosis (SWRO) component is the thermoelectric power. When thermoelectric power is used, the outcomes include particulate matter emission, greenhouse gas emissions that contribute to climate change (Bitaw et al., 2019). RO is linked with solar photovoltaic systems is an economically viable option to brackish water (and seawater) desalination. Moreover, it can overcome the current water and energy crises for water desalination (Taha & Al-Sa'ed, 2017). Furthermore, RE can reduce the environmental and ecological impacts of seawater desalination in off-grid rural communities (Alhaj & Al-Ghamdi, 2019). Wind energy and solar energy, coupled with a desalination plant, can enable ever-increasing water needs with less carbon footprint than standard methods in a sustainable manner (Aminfard et al., 2019; Jones & Olsson, 2017). Moreover, most of the desalination processes generate much-wasted heat at low temperatures, retrieved (González-Bravo et al., 2017). Considering the environmental and operational efficiency perspectives, solar, geothermal, wind, and tidal/waves are the primary sources of typical RE of desalination concern (Ali et al., 2018a).

The application of hybrid RO device could be an economical way in remote areas with strong wind and solar power (Gökçek, 2018). Off-grid renewable technology, from the household level to the village or neighborhood level, may form an essential part of the solution. Not only lighting but also electricity can be provided by small off-ride systems to pump power, clean water, and fuel. Strips can, but must be augmented with decentralized energy supplies, be accessible in increasingly increasing peri-urban areas. The new kind of hybrid energy sources will include solar and wind. In the following section, we will mainly focus on the integration of solar desalination technologies.

#### 2.2 Solar Desalination Technologies

The rapid advancement of up-to-the-minute photothermal material fabrication has enhanced the possibilities to develop varied photothermal-assisted desalination and treatment know-hows (Zhang et al., 2019). Desalination plants can be operated by solar energy directly or indirectly to decrease the desalination impacts allied with energy costs (Zhang et al., 2018). Solar desalination significantly reduces energy costs and carbon footprints. It provides a sustainable solution to meet water needs (Ahmed et al., 2019). There are two types of desalination using solar power: (a) solar collector or direct desalination and (b) solar thermal collector or thermal heat transfer to membrane-based desalination (Fig. 2). In the following segments, the types are described with their advantage disadvantage along with blended applications.

#### 2.2.1 Solar Still

Solar power can directly prepare to distill water using a solar collector, which is a straightforward solar desalination process. "Solar still" is the inducing example of the solar collector (Zhang et al., 2018). Solar stills supply potable water, and its functionality and upkeep are easy to maintain and user friendly. Integrated photovoltaic thermal solar still has excellent applications for the off-grid areas, villages, and

#### **Solar Desalination technologies**



Fig. 2 Two types of solar desalination technologies

communities facing an electricity crisis and water scarcity (Muthu Manokar et al., 2018). However, solar stills usually produce a meager amount of freshwater distillate (Samuel Hansen & Kalidasa Murugavel, 2017).

Samuel Hansen and Kalidasa Murugavel (2017) developed an integrated desalination system by merging a tilting solar still with a sink and heated water loading tank. The system performed and tested various geometrical positions of absorber plates under real-time climatic conditions. The absorbers developed had increased surface area for water availability for the evaporation in the tilted solar still. Waste heat energy could be recovered from the un-evaporated production drained from the inclined still. Tilted solar stills could produce 25.75% more water productivity while utilizing fin-shaped absorbers increased 74.5% water productivity integrated with basin still (Samuel Hansen & Kalidasa Murugavel, 2017).

#### 2.2.2 Solar Thermal Collectors

Solar collectors convert solar power to thermal power. There are multiple types of parabolic collectors used in the desalination systems, which is a solar cooling device focused on solar absorption and heat adsorption, the development of solar electricity, solar desalination, and solar disinfection (Mortazavi & Maleki, 2019). "Solar thermal collector" or photovoltaic panel captures solar energy for heat transport to

operate various types of membrane desalination technologies (Zhang et al., 2018). An example includes the application of compound parabolic collectors transferring solar energy to multi-effect distillation systems. The system is more energy-efficient than other evaporation techniques (Mortazavi & Maleki, 2019).

For the efficient application of humidification-dehumidification solar desalination systems, solar collectors or tube collectors with a parabolic trough are suggested. The high temperature of the feed water considerably increases water production (Shalaby et al., 2017). Essential considerations to enhance the device efficiency includes nano-heat transfer, fluid flow rate, enforced convection process, multi-pass technology, recycling method, and phase shifting material fabrications. Furthermore, there are studies of the impact on the watershed efficiency of solar power, air temperature, wind speed, heating, and cooling stages. The simplicity of construction of flat plate collector and the high-efficiency indices in the evacuated tube range makes them the choice of technologies in solar humidification-dehumidification desalination system, which are used to heat water and air heat transfer fluids (Santosh et al., 2019). Moreover, a twostage humidification-dehumidification desalination system configured with a single-stage flashing evaporation unit works efficiently (El-Said et al., 2016). An Al<sub>2</sub>O<sub>3</sub>/H<sub>2</sub>O nano-fluid solar water heater constructed it. The mass flow

rate of feed water and cooling water of both single-stage flash unit and humidification–dehumidification units influence the system productivity. Besides, inlet cooling water temperature and nanoparticle volume fraction also affects the productivity of the system.

# 2.2.3 Membrane-Based Hybrid Solar Photovoltaics

Revolutionary innovations in nano-membrane fabrication and error-free production procedures are crucial to efficient solar desalination technologies (Alvarez et al., 2018). In the conceivable future, membrane-based separation and desalination techniques may stay critical instruments in water treatment. Besides, nanotechnology will assist in overcoming working interferences, namely foulant and poor selectivity in the separation of ions or molecules. In nanoscale grafting and doping, membrane efficiency and reliability will be improved, while selective membranes for the specific desalination tasks can be built using nanotubes. The next-in-line desalination membranes would be antifoulant and steady against oxidizing substances such as chlorine.

Many of photovoltaics-RO systems function spasmodically to cut the expenditures of RE storage. Consequently, the influence of sporadic function influences the membrane life, develops fouling and scaling, and increases operational maintenance costs (Freire-Gormaly & Bilton, 2018). Nevertheless, a simple procedure of rinsing the membrane before shutdown can significantly uphold the membrane lifespan and preserve higher membrane permeability compared to the membranes without rinsing. Freire-Gormaly and Bilton (2018) further designated that using antiscalant and rinsing can significantly lower scale deposits on the membrane, which leads to membrane fouling.

Currently, the majority of the solar-water treatment processes are yet to be implemented for real-time applications. For example, Jun et al. (2019) examined the uses of biofouling resistance in photothermal-induced inactivation of microorganisms in RO and ultrafiltration procedures (Jun et al., 2019). Ultrafiltration as a pretreatment of RO is a promising scheme of integrating RO-multistage flush desalination. Seawater at raised temperature can be applied as feedwater in this method (Agashichev & Kumar, 2017). The pretreatment device can be segmented into a sequence of functions which are ultrafiltered, backwashed, chemically strengthened backwashing, cleansing and dried, and disposal.

Again, up-to-the-minute technologies often need complex systems with several parts that result in low efficiency yet at a too high price (Yang et al., 2018). Consequently, economic affordability is a significant reason for scaling and marketing (Zhang et al., 2018). For instance, the price of water from tiny to moderate-scale PVRO plants is in the range of US \$0.2–22/m<sup>3</sup>, which was costlier than traditional fossil-fueled

plants (Zhang et al., 2018). However, the evaluated small price of water (US $0.9-2.2/m^3$ ) for large-scale solar-powered reverse osmosis plants imply alternative options will be available soon.

# 2.2.4 Portable and Micro-desalination Based on Electrodialysis

photovoltaics-electrolysis Hybrid solar and micro-desalination have been developed to enhance water and energy security (Kim et al., 2018). A portable desalination unit based on direct contact membrane distillation powered by solar photovoltaics can be a good option for micro-desalination plants and off-grid houses (Kadi et al., 2019). Based on membrane distillation powered by solar photovoltaics, the portable desalination devices are less energy-intense and low carbon-emitting and the considerable concern is rising to integrate it with RE (Ali et al., 2018a). However, the determination of the most efficient-type RE form is yet to be configured to maximize desalinated water yield while consuming a minimum volume of energy (Abdelkareem et al., 2018). The main reason for this is the existence of diverse methods of desalination and renewable energies. The selection of RE as a source is affected by many parameters, for instance desalination plant size, place, feeder pressure, feed water features, and anticipated water price (Ali et al., 2018a).

A portable solar desalination unit consisting of  $0.042 \text{ m}^2$ membrane at 40 °C and permeate temperature of 25 °C can produce 2.31 L/day of freshwater without any external heat (Kadi et al., 2019). This technology seems highly promising to explore comprising both ideal assumptions and practical computational fluid dynamics validated by modeling.

However, the sporadic traits of the mentioned sources encompass severe obstacles to the techno-economic feasibility of the process. Renewable sources are typically utilized for small-scale desalination plants but do not apply to power big plants because they are sporadic and unstable (Mito et al., 2019). Moreover, the RO plant's close link to renewables may require swiftness and flexible operation to complement the necessary power load. More solar intensity does not, however, deduce decreased carbon dioxide emissions from solar desalination plants (Alhaj & Al-Ghamdi, 2019). Environmental factors such as seawater temperature and salinity gradients also limit different factors of desalination.

Independent photovoltaic-RO wastewater treatment plants are also promising to secure freshwater in locations that require a functional electricity network (Abdelkareem et al., 2018). The practical arrangement of photovoltaic clusters and the addition of solar tracking, adjustment of tilting angle, photovoltaic array, cleaning technologies can effectively expand the effectiveness of independent photovoltaic-RO desalination. With feedwater for cooling the photovoltaic system, increased efficiency and freshwater output in the RO unit raised cumulative productivity while decreased total costs of function.

# 2.2.5 Forward Osmosis, Pressure-Retarded Osmosis, and Other Hybridized System

Forward osmosis and pressure-retarded osmosis generates energy which depends on the use of a semipermeable membrane (Anissimov, 2016). The membrane is permeable to water while impermeable to salt. It is a potential membrane-based technique of high-salinity seawater desalination and water reuse where RO is incompetent or uneconomic (Blandin et al., 2016; Hsu et al., 2019). In forward-osmosis, water flows over semipermeable membranes that change the osmotic pressure gradient between the membrane sides. This method does not cost transmembrane energy except for those required to diffuse the system solutions (Safder et al., 2018). Forward osmosis reduces fouling and scaling, energy recovery of the brine, minimizes power consumption, and the use of chemicals. It is potentially applicable for wastewater treatment that has severe fouling propensity and when the draw solution is needed both in pretreatment and post-treatment. In addition, it requires minimal electricity. Mostly, it is usually integrated into other separation procedures to separate the draw solution from the output water or used in the pretreatment process to conventional desalination technologies (Chekli et al., 2016).

The use of forward osmosis, high pressure-retarded osmosis, and membrane recycling can significantly reduce the carbon footprint of desalination. Enhanced membranes could decrease energy claim by 15-20% (Fane, 2018). Moreover, energy decrease is the primary driving force in using RE for desalination purposes in countless remote communities (Freire-Gormaly & Bilton, 2018). The significant drawbacks of forward osmosis are the draw solution, which are usually salts and challenging to recycle energy and water recovery. Hsu et al. (2019) developed a hybrid ionic liquid or hydrogel to improve the overall forward osmosis process. The hydrogels can recover the water without heating the draw solution. It can be used in a continuous recycling process to recover water. It also alleviates the water flux drop of the ionic liquid as the draw agent (Hsu et al., 2019).

An integrated forward osmosis RO hybrid process developed by Choi et al. (2017) showed that water carrying factor and inner strength of polarization resistance are crucial issues affecting the execution in the forward osmosis process, where the salt transportation factor was not most influential. The permeation flux rate and the membrane cost of the forward osmosis are similar to RO, but the electricity cost is expensive. This hybrid system can be economically frugal if permeation flux and membrane recovery are achievable (Blandin et al., 2016; Choi et al., 2015).

Progress in the forward osmosis membrane includes the application of pressure-assisted osmosis. Membrane improvement had established noteworthy upgrading in the permeation of water flux. However, this improvement is not without fouling and the low rebuff of trace organic compounds (Blandin et al., 2016). Likewise, the forward osmosis membrane also needs to improve the mechanical resistance, focusing on future research.

## 2.2.6 Electrodialysis and Other Hybridized Method

Ion exchange membranes are unique in their combination of ion exchange resin, their electrochemical properties, and membrane permeability. Commonly used for the municipal wastewater treatment and desalination of fresh and brackish water, capacitive membrane deionization is an energyefficient and emerging technology that selectively transfers counter ions toward carbon electrodes in ion exchange membranes (Hassanvand et al., 2017).

Integrating electrodialysis with RO has several technical advantages: no driving force limitations, when nanofiltration is used, and low energy requirement. Consequently, electricity intakes of 1.3 kWh/m<sup>3</sup> accomplished < 30% recovery. However, the process is expensive than the RO system (Bitaw et al., 2019). Coupling membrane distillation with pressure-retarded osmosis or reverse electrodialysis boosts the process performances. It offers a smooth and viable route to produce freshwater and sustainable electricity from a range of wastewaters, together with brines, which would otherwise be considered for environmental obligations (Ali et al., 2018a). The decarbonization of membrane desalination is an outstanding trial to decrease carbon discharges to alleviate climate change (Fane, 2018). Owing to more significant membrane areas, the hybrid electrodialysis-RO system could reduce carbon dioxide emissions from 63 to 26%, while brine concentrations reduced from 42 to 34%, respectively (Bitaw et al., 2019).

# 2.2.7 Hybrid Membrane Distillation and Related Desalination

Steam production using solar power is the foundation for various sustainable desalination methods (Cooper et al., 2018). The steam price for membrane distillation plays a significant part in defining the profitability and feasibility of RO membrane distillation/pressure-retarded osmosis integrated/hybrid systems (Choi et al., 2017). Membrane distillation has demonstrated the ability to produce electricity and fresh water simultaneously by itself. Plenty of inexpensive renewables are an alternative way to contemporary power procedures of desalination. Furthermore, developing a

scalable and economic "photothermal membrane" for RO, ultrafiltration and membrane distillation, and solar steam generation has prospective powering for an energy-efficient membrane distillation (Jun et al., 2019). Moradi et al. (2019) informed about synchronized energy generation and thermal waste retrieval scheme in a multi-effect desalination system. For steam generation using heat energy from the vent, a heat recovery steam generator is applied. The emitted steam moves through a multistage desalination device to yield freshwater (Moradi et al., 2019).

The vacuum membrane distillation process and adsorption desalination units are coupled to designate a "VMD-AD cycle." It maintains the low vacuum atmosphere in the high-pressure gradient across the microporous hydrophobic membrane. Furthermore, this system supported with solar power and temperature modulating unit showed a higher total water production (21%) and water recoveries (23%) compared to the conventional system (Kim et al., 2016). Additionally, an amalgamation of membrane distillations with pressure-retarded osmosis or reverse electrodialysis can improve efficiency and deliver a sustainable path for freshwater and energy production. High-pressure RO has functioned at a hydraulic pressure greater than  $\sim 100$  bar has also been applied to desalinate hypersaline brines competently (Stevens et al., 2017). It has a potential application for high-salinity industrial wastewaters, and this can decrease energy needs for desalination. Membrane distillations with pressure-retarded osmosis or reverse electrodialysis have a potential industrial application of wastewater management by zero liquid discharge. An economical replacement for more excellent salts concentration could be microfiltration blended with assisted RO as well as coupling with mechanically based vapor compression or forward osmosis-based vapor compression (Osipi et al., 2018).

#### 2.2.8 Nano-enabled Desalination

Nanotechnology provides a novel, innovative products for water treatment systems by regulating the material, size, morphology, and chemical composition. Nano-enabled water treatments possess excellent catalysts that increase the cost-effectiveness of water treatment with adsorption, optical, quantum, electrical, and antimicrobial properties (Alvarez et al., 2018). As an alternative to polyamides, many innovative nanoporous materials are developed (Stevens et al., 2017), which are exclusively used for RO and nanofiltration membrane production. The development of distinct kinds of nanomaterials such as graphenes, graphene oxide, block copolymers, liquid crystals, aquaporins, and other biologically inspired molecular channels and thin-film composites, carbon nanotubes (Stevens et al., 2017; Yang et al., 2019) is increasing field of research aimed at meeting the cumulative requirement of freshwater around the globe. These carbon-based nanomaterials often have a more

significant surface area, flexibility, and have intrinsic adsorption and sieving ability to eliminate waterborne contaminants and minerals. Because of their lightweight nature, these materials are readily transportable for household and business-scale portable water purification appliances (Alvarez et al., 2018; Yang et al., 2018).

Ali et al. (2019) revealed that the bench-scale implementation of nanocomposite membranes of carbon nanotubes could enhance water penetration, high selectivity, and antifouling capacity (Ali et al., 2019). Moreover, membranes containing multi-walled carbon nanotube-polyamide nanocomposite segments above 12.5 wt% improve the degradation of RO membrane's resistance against chlorine exposure. It increases water flux and maintains salt rejection performance (Ortiz-Medina et al., 2018).

Nano-enabled treatment methods indicate that energy and chemical needs, and solid waste and wastewater residuals generation, the associated cost and likely ecological effects may be considerably reduced. For instance, nanophotonicbased technology can be combined with RE sources to harvest freshwater in the distillation process. Besides, to reduce interference and improve effectiveness and interruptions, nanotechnology can strengthen membranes and photocatalysts (Alvarez et al., 2018). However, the evolving upsurge of the invention can ultimately enable nextgeneration modular water treatment technologies to considerably enhance water security and resilience in water supply (Alvarez et al., 2018). Besides, functional carbon nanotubes (f-CNT) membranes further revealed superior antifoulant repellant because of the characteristics of elevated conductivity of electrical charge and the presence of negative surface charge. By creating reactive oxygen species, f-CNTs impair microbes and repulse biofilm growth responsible for fouling (Ali et al., 2019). Graphene is another illustration of a carbon-based substance that functions as a solar-thermal converter. 3D cross-linked honeycomb graphene foam material captures and converts solar energy into heat. Subsequently, the heat distills water into steam and generates freshwater effectively, even under low sun intensity (Yang et al., 2018). Besides, the hydrophilic quartz glass fibrous, often known as QGF membrane composite, dramatically lowers the cost of investment by minimizing land size and increasing freshwater production (Wang et al., 2019).

# 2.2.9 Other Interesting Methods for Future Potentiality

 Pervaporative desalination is a membrane-based procedure which is not commercially explored yet but has tremendous potentiality. The pervaporative membrane material is produced from polyvinyl alcohol, maleic acid, and tetraethyl orthosilicate. The considerable amount of heat energy (2609 MJ/m<sup>3</sup>) is needed to recirculate the fluid flux at 65 °C. It required electricity as low as <0.2 kWh/m<sup>3</sup>, and the vacuum pump mainly supplied the electricity. Pervaporation might be an alluring desalination method when it is combined to recover waste heat and salt (Xie et al., 2018). Pervaporative desalination is one of the most influential and encouraging membrane technologies where the nanocomposite membranes play the most crucial role. Nanocomposite membranes also regulates procedural efficacy (Roy & Singha, 2017). In pervaporation desalination, the efficiency of photovoltaics depends on strength, and mechanical features of the membrane, along with operational parameters. Therefore, advancement on the high-performance and cost-effective nanocomposite membranes having the

 The latent heat of the water vapor can be condensed and retrieved in each distillation using photovoltaics with a multifaceted membrane distillation unit at the flip side of solar cells (Wang et al., 2019).

given emphasis (Singha et al., 2019).

benefits of both organic- and inorganic-membranes have

- 3. Nuclear energy proposes a viable choice for simultaneous thermal energy generation and freshwater coproduction by desalination. A significant quantity of heat is recovered during the process, which can be utilized for freshwater reclamation. The recovered heat is employed to harvest vapor and make thermal electricity onsite to provide thermal power and facilitate membrane desalination. Different sizes of nuclear power reactors can be coupled with a variety of desalination methods. The charge was calculated in the range of \$0.4–1.8/m<sup>3</sup> using nuclear power based on the reactor category and the procedures for desalination (Al-Othman et al., 2019). Small modular reactors appear to be the safest selection in nuclear desalination and suitable for the newcomer nations adopting nuclear energy.
- For a coastal region, RE-pumped storage structures can be merged with desalination plants to form a RE pumped storage-seawater desalination system (Liu et al., 2019). RE-operated units have gained substantial importance.
- 5. Membrane distillation produces high-purity water indifferent to a salinity gradient. Membrane distillation was employed to condense geothermal brackish water using RO units after the cooling of the treated feedstock. Integrating membrane distillation-RO for treating geothermal water resulted in a substantial advantage. The integrated system recovered 30% to 40% water. The price of energy per m<sup>3</sup> varied from 0.4 to 0.9 \$ depending on the differences in RO operating pressures (which varied between 6 and 40 bar). It was found that brine recycling also improved the recovery rate and

performance ratio only when brine was reused around the RO unit (Ali et al., 2018b).

- 6. A combination of geothermal and solar energy was used in the humidification–dehumidification system. Geothermal energy provided constant heat supply (24-h), and the operating system is efficient in areas with geothermal water temperature above 60 °C and a flow rate below 0.15 kg/s. The average percentage of humidity at the humidification unit outlet increased by 46% for an increase in geothermal water flow rate from 0.05 to 0.1 kg/S. Simultaneously, the solar energy increased the temperature of induced air by 3–4 °C, which heightened the evaporation level within the humidification unit. Daily potable water output was near about 192 L/m<sup>2</sup> and cost 0.003 USD/L (Elminshawy et al., 2016).
- In many studies on integrated/hybrid methods of desalination algorithms and mathematical models and modeling software such as HOMER have been utilized to improve different RO coupled with other desalination systems (Agashichev & Kumar, 2017; Ahmed et al., 2019; Ali et al., 2017; Anissimov, 2016; Bitaw et al., 2019; Esmaeilion, 2020; Khan et al., 2018; Maleki, 2018; Monnot et al., 2018; Rabiee et al., 2019; Roy & Singha, 2017; Santosh et al., 2019; Shalaby et al., 2017). By the optimization of mathematical models and algorithms, hybrid desalination systems can perform better.

#### **3** Concluding Remarks

The rising water demand has resulted in a dramatic increase in the research of global desalination efficiency. Through an attempt to reduce the carbon footprint due to energy-intensive fossil fuel-based desalination, the focus has been given to integrate REs and improve the efficiency of membranes using different types of nanocomposite materials. A combination of renewable energies and desalination techniques is being tried to achieve economically viable water and energy output as well as reduce environmental impacts. Multiple solar irradiation technologies have been developed to integrate solar energy. It has also focused on the development of portable and micro-desalination based on electrodialysis for the remote off-grid islands and communities. Forward osmosis, pressure-retarded osmosis, and other hybridized systems are being improvised to for leveraging RO processes, overcome its limitations such as increasing water flux, decrease membrane fouling, enhance lifetime of the membrane, waste heat, and water recovery with potential future applications. The integration of desalination technologies is required for identifying cost-effective, commercially viable sustainable water and energy production with fewer environmental impacts. It can also help the water and energy researchers, policymakers, and legislators to identify which integrated technologies to have opted for sustainable water and energy security.

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# **Reverse Osmosis Desalination**

Ashish Kapoor, Muthamilselvi Ponnuchamy, and Sivaraman Prabhakar

# Abstract

In the last few decades, desalination processes have played a crucial role in meeting the growing demand for water. Desalination process consists of raw water, desalination device, product water and energy. Among the desalination processes, membrane-based reverse osmosis desalination has become popular because of its characteristics such as ambient temperature operation, low specific energy consumption, modularity, and tolerance to intermittent operation. The chapter discusses the mechanism of desalination using preferential sorptioncapillary flow model. A brief description of membrane preparation techniques is followed by a discussion on the merits and demerits of the membrane element configuration based on tubular and flat sheet geometries. The various steps involved in the design of RO desalination plants are indicated with the reason for each operation including pretreatment involving fouling and scaling control, design considerations for fixing the recovery, the arrangement of modules including the concept of stages and passes. The monitoring methods including silt density index are indicated. The need for post treatment and the methods used are discussed including control of boron concentration in the permeate. The developments of various types of pumps and the corresponding energy recovery devices over the years resulting in the reduction of specific energy consumption to less than 3 kWh/m<sup>3</sup> are highlighted. While discussing inland brackish water desalination, it is indicated that the design strategy should focus on resource conservation and sustainability rather than minimisation of cost. The specific energy consumption of reverse osmosis desalination plants nowadays is

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around or less than about 3 kWh/m<sup>3</sup> and further improvement in this direction will be only marginal. Major improvements are expected in the development of membranes that resist fouling.

#### Keywords

Desalination • Reverse osmosis • Pretreatment • Membrane arrangement • Post treatment • Energy recovery

# 1 Introduction

Though two thirds of the earth's surface are covered with water, only less than one percent is available as fresh water suitable for direct use. Even this quantum of water has a skewed distribution across the world resulting in many water stress or water-scarce regions. Seawater accounts for more than 96% of the available water resources but is not suitable for many human activities because of high salinity levels coupled with the challenges in tapping the resources in a cost-effective manner. Several inland ground water sources are also saline which are not fit for direct human use. To meet the growing demand for water it is necessary to convert saline water to sweet water. Desalination refers to the processes which enable the separation of water from saline sources mostly using physico-chemical methods. In the absence of any sustainable source of water, it is imperative to look for desalination of seawater and brackish water to augment the fresh water sources. In the early twentieth century, pure water was obtained from saline water by distillation process. Even though the phenomenon of osmosis was known in the eighteenth century, its practical application for the separation of water from solution gathered momentum after, ambient temperature casting of cellulose acetate membrane in the early sixties by Loeb and Sourirajan (1963). Thermal processes require thermal energy and are

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limited by thermodynamic efficiency. Further inverse solubility of calcium carbonate and sulphate solutes limits the operational temperature and recovery (Roy et al., 2017). On the other hand, reverse osmosis desalination processes use mechanical energy at ambient temperature. Sustainability of the membrane surface with reference to physical, chemical, and biological deterioration is a challenge. In the last four decades, both the thermal and reverse osmosis desalination processes have undergone many modifications towards reliability, reduction in specific energy consumption and cost of water. About 16,000 desalination, plants are in operation across the world accounting for about 95 billion litres per day of desalinated water (Jones et al., 2019). However, as seen in Fig. 1, the reverse osmosis (RO) desalination has grown more significantly compared to thermal processes with respect to capacity and number of plants in the last two decades in view of specific features with reference to flexibility in operation and less energy consumption.

The basic components of a desalination plant include input resources such as water source, energy, desalination device and outputs such as purified water and reject stream as shown in Fig. 2. Desalination systems can remove either water or dissolved salts preferentially, depending on the characteristics of the system. Many desalination devices have been developed including reverse osmosis (RO), electro-dialysis (ED) and a number of variants of thermal desalination (multi-stage flash (MSF), multi-effect distillation (MED), mechanical vapour compression (MVC) and thermo vapour compression (TVC)) in commercial scale. A few more devices such as forward osmosis (FO), membrane distillation (MD), capacitative deionisation (CDI) (Zhang et al., 2020), electro-deionisation (EDI) and freeze desalination (FD) are evolving.

Energy is a major requirement of a desalination plant, whose actual consumption depends on energy type used, process design, feed characteristics and product purity. Thermal, mechanical, and electrical energy have been commercially used for desalination of sea water and brackish water. Solar energy has been used directly as well as in derived forms: thermal, electrical, mechanical and ocean thermal energy. Thermodynamically (irrespective of the desalination process), the minimum energy required is about 0.7 kWh for extracting 1000 L of pure water from an infinite source of seawater at 25 °C. Complete recovery of pure water is not possible in any of the devices, because of the physical and chemical properties of the solutes present resulting in irreversible loss of a fraction of energy used in the process. Constant improvement in all the processes have resulted in the reduction of energy consumption from about 10-20 kWh range to about 2.5-11 kWh per 1000 L, depending on the type of energy used and process design. Local logistics and operational sustainability considerations have in specific instances resulted in adopting novel energy-intensive processes such as ocean thermal energy conversion (OTEC) (Davis & Schubert, 1974).



**Fig. 1** Growth of desalination over years 1960–2020, **a** desalination capacity and number of desalination plants and **b** capacity of various types of desalination plants (reproduced from (Jones et al., 2019) with permission from Elsevier)

desalination



Reject Brine Concentrate

The characteristics of the feed both with respect to the dissolved solids and suspended matter influence the design of a desalination system, as the heat and mass transfer efficiencies are seriously affected by fouling and scaling. Hence, the feed requires pretreatment and the rigour depends on the process.

Product quality and the fraction of water obtained with respect to the original feed (recovery) varies depending on the process and to some extent on process design. Thermal desalination processes normally yield high purity water with less solute content as the separation involves phase change. In all the other processes, the product water has some dissolved solutes. In any case, post treatment is necessary either for supplying safe drinking water or for transmission through pipelines (to minimise corrosion) for other uses.

Reverse osmosis desalination is a pressure-driven process requiring mechanical energy, working at ambient temperature with a membrane capable of preferential permeation of water. Thermal processes are energy-intensive involving phase change. Electro-dialysis process has drawbacks including high energy consumption and poor product quality. In view of these factors, most of the newly constructed desalination plants are based on reverse osmosis technology. In the succeeding sections, different aspects of reverse osmosis desalination technology are discussed.

#### 1.1 **Fundamentals of Reverse Osmosis**

Osmosis, a natural phenomenon, describes the preferential flow of water from dilute solution to concentrated solution through a semipermeable membrane separating both solutions. The driving force is the osmotic pressure gradient arising out of the chemical potential difference of water across the membrane and is dependent on the concentration of the dissolved solutes. Osmotic pressure can be visualised as the water-solute binding energy which must be overcome to remove water from a solution and the binding energy increases with an increase in dissolved solutes per unit volume of water. In the reverse osmosis process, water is forced through the semipermeable membrane by the application of hydrostatic pressure above the osmotic pressure of the solution (Nagy, 2019). The applied pressure can be considered as the sum of the binding energy (osmotic pressure) and the driving force for the Poiseuille flow of water.

#### 1.2 **Basic Mechanisms of Reverse Osmosis** Process

Amongst the approaches suggested in the literature to explain the transport processes through membranes, preferential sorption-capillary flow (PSCF) and solution diffusion mechanisms are two extremes. The former (Kimura & Sourirajan, 1967) considers the transport of water as capillary flow through the pores of the membranes. Further, it recognises interfacial adsorption of water on the membrane surface. The second approach (Riley et al., 1967) considers that the water and solutes dissolve in the membrane matrix and diffuse through the membrane and does not recognise the preferential sorption at all. The separation is attributed to the combination of two factors solubility in the matrix and diffusivity through the matrix. Third approach is based on the principles of

Product

Fresh water

irreversible thermodynamics (Spiegler & Kedem, 1966) considering the membrane as a black box. The passage of water and the solutes are governed by the respective concentration gradients. Further, the mechanism assumes that water flow is coupled with solute flow and vice-versa.

For the description of reverse osmosis process, preferential sorption capillary flow mechanism is considered suitable. Accordingly, as shown in Fig. 3, water from the solution gets preferentially adsorbed onto the membrane surface. The thickness of the sorbed layer depends on the physicochemical nature of the membrane. Under the application of pressure, greater than the osmotic pressure, water flows through the capillaries. As the pores in the membrane are not uniform, a salt passage occurs through the larger pores by diffusion. Since the boundary layer formation is inherent due to the preferential removal of water, osmotic pressure of the boundary layer and concentration of solutes at the boundary layers are considered for the water flux and salt flux. Since boundary layer concentration is not directly measurable, a third equation is derived to relate the bulk concentration with boundary layer concentration and permeate concentration based on film theory. The basic equations are

$$N_w = A_m [P - \Delta \pi] = A_s [C_2 - C_3]$$
  
=  $k(1 - C_3) \ln \left\{ \frac{(C_2 - C_3)}{(C_1 - C_3)} \right\}$  (1)

 $N_w$ : water flux in L/m<sup>2</sup> s.

 $A_m$ : membrane constant, L/s m<sup>2</sup> atm defined as water permeating through the membrane per unit area and unit pressure for pure water (zero osmotic pressure) as feed.

P: Applied pressure atm.

 $\Delta\pi$ : difference between osmotic pressure of feed solution and product solution in atm.

 $A_s$ : membrane surface area in m<sup>2</sup>.

k: mass transfer coefficient m/s.

*c*: solution concentration and suffices 1, 2, 3 represent bulk feed, boundary layer and permeate respectively.

The above discussions lead to the inference that the membranes for RO desalination should have the property conducive for interfacial adsorption of water molecules and structural pores to allow water transport with minimal resistance.

# 2 Preparation of Membranes and Membrane Modules

#### 2.1 Preparation of Membranes

Membrane is the heart of the process and has the characteristics to control the preferential separation of water. At the same time, the membrane should offer less resistance for the flow of water to achieve reasonable quantity of pure water at moderate operating pressures. Hence, the reverse osmosis membrane should be highly semipermeable and strong enough to withstand the operating pressure and offer less resistance for the flow of separated water.

To achieve the requisite semi-permeability, the membrane surface should have the ability to preferentially attract water, leaving out the solutes which are present in the hydrated state in the solution. This is achieved by balancing the presence of hydrophilic and hydrophobic moieties of the membrane surface as well as keeping the pore size distribution in a narrow range with the average pore size around a few Angstrom units. Cellulose acetate membrane was the first membrane used for the demonstration of the RO desalination process albeit at high operating pressures (Merten et al., 1967). Later, synthetic polymeric membranes were developed, to achieve a high degree of separation. The average molecular weight of the polymers used for the membrane preparation was high enough to provide, inherent strength to withstand the operating pressures at least up to 80 bars. Since separation occurs only on the surface of the membrane, porous matrix support coated with an active polymeric membrane layer has been developed resulting in high water flux and better salt removal efficiencies at lower operating pressures.

Membranes can be of different types made of different materials: porous or non-porous, organic, or inorganic, synthetic, or natural. The membranes can be made from a single polymer or from two different polymers one coated over the other. Depending on the structure and morphology they are classified as asymmetric or symmetric, homogenous



Fig. 3 Working mechanism of reverse osmosis membrane process

(isotropic) or heterogeneous (anisotropic). Asymmetric membranes can be of the same material or made of two different materials. Symmetric membranes are uniform throughout, while asymmetric membranes exhibit changes in density in its structure (porosity will be changing suddenly or gradually). Homogeneous membranes are highly dense membranes. For reverse osmosis, only asymmetric membranes are used to ensure minimum resistance for water flow without sacrificing the selectivity of separation. Accordingly, the active layer requires to be very thin with small surface pores (with average pore sizes around a few angstroms). Commercial asymmetric membranes were earlier prepared from the same polymer viz. cellulose 2.5 acetate, cellulose triacetate and aromatic polyamides. Later, Cadotte et al. (1980) developed a technique for in situ polymerisation over a porous matrix, normally an ultrafiltration membrane, popularly known as thin film composites (TFC).

Normally reverse osmosis and ultrafiltration membranes are prepared by phase inversion technique. In phase inversion method, a dope solution consisting of a mixture of membrane polymer, solvent and a pore-forming additive, is prepared and its homogeneity is ensured by thorough mixing and degasification. The solution is cast in 'sheet' or 'tubular' form and immersed into a gelling medium, which is normally water. Between the casting and gelation, the film gets exposed to the environment during which evaporation of solvent takes place leading to the formation of smaller pores on the surface. During the gelation, all additives and solvents present in the film are exchanged with water molecules thus allowing the formation of the pores. The pores formed during exposure to air are smaller compared to those formed during gelation. Since the surface structure is different from the rest the membrane, these types of membrane are known as 'asymmetric membrane'.

The membranes formed by in situ polymerisation of active monomers over a porous support material (0.01 micron average pore size and asymmetric in nature) results in a thin active layer and the membrane thus formed is a thin film composite. As shown in Fig. 4, the support is immersed into a solution of water soluble monomer (mostly aqueous m-phenylene diamine), drip dried, and immersed into an organic reactive monomer solution (tri-mesoyl chloride) and cured at temperatures between 80 and 150 °C (Ali et al., 2020). The membrane formed has a thin 'active' layer resulting in high solute removal. As the thickness of the active layer is less compared to asymmetric membranes, permeate water flux is also higher.

There are other methods available for the preparation of membranes including track etching (Apel, 2001) and melt casting (Erukhimovich & de la Cruz, 2004) but these membranes are not used for reverse osmosis or nanofiltration. Thin film composite membranes are used commercially for reverse osmosis desalination. It is to be noted that the average pore sizes of the reverse osmosis membranes are in the order of a few Angstrom units. The pores are not uniform and there exists a pore size distribution on the membrane surface.

# 2.2 Membrane Configuration

Water separation from saline water through reverse osmosis membranes is essentially a surface phenomenon. The water that is adsorbed on the membrane surface permeates through the pores under the applied pressure. The permeation rate at constantly applied pressure depends on the resistance, the pores offer. Thus, separation of water from the solution is governed by the top surface of the membrane while the water flux depends on the support layer resistance and hence the thickness. Compactness, defined as 'surface area of the membrane per unit volume' is an important consideration. Total production of the membrane desalination process is proportional to membrane area. The footprint required for the plant is therefore depends on the compactness of the membrane unit. As the separation takes place progressively with the passage of feed, the concentration of dissolved species increases, particularly near the membrane surface affecting the performance. The fluid flow characteristics play an important role in reducing the impact of the 'concentrating feed' on the membrane performance.

Membrane configuration refers to the packing of the prepared membrane into different geometries catering to different requirements of compactness and hydrodynamics. There are basically four commercial membrane configurations, each one with unique hydrodynamic characteristics, and varying compactness meeting different requirements with reference to feeding quality and design constraints.

Membranes are generally prepared as long sheets with maximum width of one metre. Alternately, they are prepared in tubular forms with inner diameters varying from about 6 mm to about 25 mm or as capillaries of about 0.5 mm to about 2 mm inner diameters. Hollow fibre membranes used for reverse osmosis have less than 100 micron inner diameter and up to about 150 micron outer diameter.

Two configurations are prepared from sheet membranes viz. plate and frame and spiral wound and the remaining two are based on cylindrical membranes viz. tubular and hollow fibre or capillary. Each of these configurations has distinct characteristics and are useful under certain specific environment (Belfort, 1988).

#### 2.2.1 Plate and Frame Configuration

Plate and frame configuration akin to filter press consists of small membranes in circular or hexagonal shapes with appropriate arrangement for the passage of feed both in series and parallel mode (Balster et al., 2015). As shown in



Fig. 5, the membranes are placed on either side of a pressure plate. A separator plate placed in between the pressure plates separates the membranes on each surface. The plate surface is designed with a slope and random projections on the surface to induce turbulence to the feed. The feed flows along the surface of pressure plates contacting the active surface of the membranes while the permeate is transported by capillary action through the non-woven cloth placed between the membrane and the separator plate. The maximum surface area of each membrane is less than 900  $\text{cm}^2$ . The advantages include facile construction of unit modules with different membrane areas, easy replacement of damaged membrane and less waste generation after use. Earlier, plate and frame RO modules were used for ship borne RO desalination plants but discontinued due to high unit cost because of the requirement of pressure plates for every pair of membranes used. At present, plate and frame configuration is mostly used for nano-filtration applications in dairy industries (Kumar et al., 2013).

#### 2.2.2 Spiral-Wound Configuration

Spiral-wound configuration consists of long sheets of membrane spirally wound over a porous tube. Each pair of membrane sheets is made into an envelope sealing three sides of the membrane leaving one width side open, exposing the active surface of the membrane. Non-woven cloth pieces longer than the membrane envelope and with a slightly smaller width is inserted into the envelope. The open end of the envelope is then sealed and fixed to the porous central tube to enable the outflow of the permeate water into the central tube. Number and length of the envelopes vary depending on the unit capacity. The maximum width of the membranes manufactured is around one metre. The membrane envelopes thus prepared are spirally wound over a central porous tube such that the permeate drains into the central tube. Feed spacers are introduced between the envelopes to facilitate the feed flow through the annular space. The whole assembly is sealed with an outer cover mostly of fibre-reinforced plastic (FRP). The permeate water flows out through the central tube, while the feed enters through the annular space available between the envelopes and exits through the other end as shown in Fig. 6. Commercial elements are available in different standard sizes from several manufacturers (Hoek et al., 2013).

#### 2.2.3 Tubular Configuration

Hollow cylindrical membranes of inner diameter varying from 0.25" to 1" are inserted into a porous support tube and assembled akin to a shell-tube heat exchanger (Fig. 7). Porous tubes should be of sufficient strength to withstand the operating pressure. Feed passes through the tube side and the permeate is collected in the shell side.

#### 2.2.4 Hollow Fibre Configuration

Hollow fibre capillary element is a micro-version of the tubular configuration (Fig. 8). The basic difference is that the membranes can operate without any support for withstanding the operating pressure. Hollow fibre membranes are asymmetric membranes prepared by phase inversion technique and has a very high surface to volume ratio, resulting in higher specific production and less footprint requirement compared to all the other configurations. Several RO desalination plants were installed using Dupont's aromatic polyamide based *Permasep* hollow fibre membrane elements up to mid-nineties. The basic disadvantage of hollow fibre modules arises from its vulnerability for heavy scaling and



Fig. 5 Plate and frame membrane module



Fig. 6 Spiral-wound membrane module

fouling due to constrained hydrodynamics, requiring frequent maintenance and replacement of membrane elements. Consequently, the use of these elements was discontinued. After nearly a decade, Toyobo's cellulose triacetate based *Hollosep* membrane elements were installed in Middle-East (Khawaji et al., 2007) around 2007 for seawater desalination. Later 'Hollosep' membrane manufacturing unit has also been set up in Rabigh, Saudi Arabia. The challenge appears to have been lessened by chlorination as CTA membranes can tolerate chlorine, unlike polyamide membranes.



#### 2.2.5 Comparison of Membrane Configurations

Comparisons of these module configurations based on compactness, hydrodynamics, ease of maintenance and economics have been reported in the literature (Belfort, 1988). For large scale applications as in desalination, where the installed capacities are in the order of few hundreds of million litres per day (MLD), footprint requirement becomes an important consideration. Desalination process, being a surface phenomenon, the sustainability depends on providing a conducive hydrodynamic environment, whereby the fouling and scaling become minimum. Tubular membrane elements are amenable for maintaining turbulent flow conditions for minimising the fouling and scaling concerns. In the plate and frame module turbulence is created by plate surface geometry and frequent changes in the direction of flow. In the case of spiral elements turbulence is induced by the network geometry of the feed spacers, while in hollow fibre membrane elements only laminar flow is possible as the diameters of the fibres are extremely small (<100 µm internal diameter) and no scope exists for introducing turbulence promoters.

With reference to fouling and scaling resistance, the ranking of membrane elements is (bad to good): hollow fibre, spiral, plate and frame and tubular. Hollow fibre membrane elements were found to be most challenging with frequent failures while tubular membranes were more suitable.



Fig. 8 Hollow fibre membrane module



Desalination plant capacities are normally huge in the range of a few hundreds of million litres per day (MLD). The production of water depends on the surface area of the membrane. Compactness is defined as the area per unit volume that should be high to reduce the footprint required for the desired capacity. Hollow fibre, spiral, plate and frame, and tubular have decreasing compactness, respectively. Since hollow fibre membranes are prone to scaling and fouling, spiral element is the most preferred and exploited configuration for desalination. Hollow fibre membrane based on polyamide membranes have not stood the test of time with reference to sustainability and was discontinued for reverse osmosis applications. Recently CTA based hollow fibre units are installed in RO plants in Saudi Arabia. The performance is under assessment and not much information is available in the public domain. Tubular and plate and frame modules are not suitable for large scale desalination because of the economic considerations but they find use in a number of industrial applications more for dewatering rather than for pure water production and for experimental studies (Le & Nunes, 2016).

# 3 Role of Feed Water Chemistry in Reverse Osmosis Desalination

Reverse osmosis desalination process is a surface phenomenon. For sustained performance, it is necessary to keep the membrane surface in a clean condition. Feed water depending on its compositional chemistry may lead to many challenges in the operation and performance of the reverse osmosis desalination plants with reference to corrosion, scaling and fouling, and degradation of construction materials of equipment and membranes.

# 3.1 Feed Characteristics

Water has different salinities depending on the location and has been classified by international water association into different categories as given in Table 1. Many large capacity reverse osmosis plants use seawater as the source. Where an acute shortage of fresh water is encountered, brackish water is used for desalination. Industries use their wastewater streams as a feed to recover water for reuse.

Seawater contains all elements present in nature. In general, seawater salinity in terms of total dissolved solids is considered as 35,000 ppm (total dissolved solids). However, the salinity has been found to vary depending on the geographical location as shown in Table 2 and depth of the ocean. Besides, the seawater contains small amounts of dissolved organic matter, particulates and dissolved atmospheric gases (Talley, 1978). The relative ratios of major components such as sodium, chloride, calcium, magnesium, and sulphate are nearly constant even though the salinity values vary widely. The pH of the surface seawater is in the range of 7.8–8.2 thanks to the buffering action of carbon-dioxide and weak base (calcium hydroxide) interaction. However, with depth, the pH changes towards the acidic side.

Brackish water is mostly found in hinterland as ground water, depending on the geo-hydrological environment. The salinity may vary both with the depth of the well and the withdrawal rate. Composition of water in terms of dissolved solutes vary widely. Other than salinity, most of the inland sources have other contaminants either geo-genic (emerging from geological environment like fluoride, arsenic, iron, nitrate, etc.) or anthropogenic (emerging from human activities related to industrial, agricultural and domestic like pesticides, heavy metal ions, etc.) in nature.

# 3.2 Scaling and Fouling

Raw seawater has suspended matter, microorganisms, and dissolved solutes. Microorganisms and suspended matter may settle on the membrane, masking its surface, leading to a loss in productivity and damage to the membrane in the long run. As the desalination process involves continuous withdrawal of pure water, the feed solution gets progressively concentrated and at some level of concentration, sparingly soluble mineral salts get deposited on the membrane, reducing its performance. The scale forming components include calcium salts such as carbonates, sulphates, fluorides, phosphates, strontium & barium (sulphates), and compounds of iron, aluminium, manganese, and silica. **Table 1**Salinity characteristicsof feed water for desalination

Classification of water	Salinity (TDS)		
Water (Potable)	<1000 ppm		
Brackish	1000–5000 ppm		
Highly Brackish	5000–15,000 ppm		
Saline water	15,000–30,000 ppm		
Sea Water	30,000–45,000 ppm		
Brine	45,000–300,000 ppm		

**Table 2**Seawater compositionat different locations (all valuesare in mg/L)

Constituents	Normal	Mediterranean	Gulf (Arabia)	Red Sea
Chloride (Cl <sup>-</sup> )	19,000	21,000	23,000	22,200
Sodium (Na <sup>+</sup> )	10,600	11,800	16,000	14,300
Sulfate $(SO_4^{2^-})$	2650	2950	3200	3100
Magnesium (Mg <sup>2+</sup> )	1260	1400	1800	750
Calcium (Ca <sup>2+</sup> )	400	420	500	230
Potassium (K <sup>+</sup> )	380	460	460	210
Bi carbonate (HCO <sub>3</sub> <sup>-</sup> )	140	-	140	150
Strontium (Sr <sup>2+</sup> )	15	-	-	_
Bromide (Br <sup>-</sup> )	70	160	80	70
Borate (BO <sub>3</sub> <sup>3-</sup> )	25	70	-	_
Total dissolved solids (TDS)	34,540	38,260	45,100	41,010

https://www.lenntech.com/composition-seawater.htm#ixzz52UEOVZwd; accessed 04 June 2020

Microorganisms whose physical sizes being higher than the pore size of the membrane would also settle as foulants along with suspended solids (Marian et al., 2015).

# 3.3 Chemical Deterioration

The presence of oxygen and oxidising agents lead to oxidative degradation of polyamide polymers resulting in the loss of structural integrity and properties. Chlorine is more dangerous as it can also directly attack the nitrogen of the amide group by 'N-chlorination'. Cellulose acetate membranes are vulnerable to hydrolysis and loss of its semi-permeability over time. The rate of deterioration can be controlled by operating the system at mildly acidic conditions (pH around 4.5) where the hydrolysis constant is minimum (Vos et al., 1966).

#### 3.4 Corrosion

The corrosive nature of seawater is attributable to salinity, microbiological activity, dissolved oxygen concentration, and temperature. Presence of high chloride concentration may promote localised corrosion like pitting, crevice and stress corrosion. At high seawater velocities, erosion may also occur (Larché & Dézerville, 2011). In certain instances,

bio-corrosion may also occur when the operation is suspended for a longer duration. Careful choice of material of construction is called for trouble-free operation. SS316 was considered earlier as suitable but experience has shown that it is vulnerable for pitting corrosion (Abu-safiah, 1991). Better varieties of stainless steel such as duplex and super duplex steel containing more percent of chromium and molybdenum are used for improved corrosion resistance.

#### 4 Seawater Intake System

Several options are available for seawater intake. However, the final choice is based on economics which in turn depends on the quantum of seawater required, environmental aspects and locational constraints. The possible intake systems are *'beach wells'*, *'open seawater'* and *'deep seawater'*. Beach well is the most economical source and the quality of seawater is excellent with very few suspended solids and microorganisms. However, the available water is limited by the geological characteristics of the location. This type is suitable mostly for small volume requirements, say about 1000 m<sup>3</sup>/day or less. The open seawater intake requires less investment compared to deep seawater intake, but the quality of water would be poor in terms of microbial load and suspended matter. Further sufficient safeguards must be provided to protect the intake pump from damage from sand

and particulate matter. Deep seawater intake has the benefits of better quality in terms of surface pollutants and suspended matter but requires high investment. The intake mouth is normally located at least 500 m away from the shore to take care of low tides as well as to draw water from a depth of about 5 m or more from the surface to ensure the quality. Deep seawater intake is viable in most instances for capacities equal to or higher than 500,000  $\text{m}^3$ /day (Gille, 2003). The source for most of the brackish water desalination plants is underground water and secondary treated industrial effluents. In both cases, the volumes available are much less.

# 5 Pretreatment of Seawater for RO Desalination

For sustainable performance of reverse osmosis desalination plants, pretreatment of the feed is critical to make the feed water compatible for membrane processing in terms of physical, biological and chemical characteristics. This is necessary for the following reasons:

- Membrane separation is a surface phenomenon and hence the membrane surface requires to be preserved with respect to its physico-chemical characteristics to ensure sustained performance.
- Suspended matter and foulants can deposit on the membrane surface under applied pressure and mask the surface thus reducing its performance. Hence, physical pretreatment is carried out at least to minimise if not eliminate the foulants including particulates, suspended matter and colloids.
- Desalination process involves continuous removal of water resulting in a progressive increase in the concentration of the feed as it flows over the membrane surface. The presence of sparingly soluble salts such as calcium carbonate, calcium sulphate would precipitate on the membrane surface when their solubility limits exceed, thus affecting the performance. This can be achieved by a combination of design strategy and the addition of anti-scalants.
- All the commercial membranes used in reverse osmosis desalination are made of aromatic polyamides except cellulose triacetate and are vulnerable to oxidative and chemical degradation due to chlorine. The damage can be minimised by maintaining a reducing environment in the feed as it enters into the reverse osmosis module.
- Reverse osmosis membranes with pore sizes in the range of a few angstrom units are smaller compared to the dimensions of microorganisms. Hence, during the process of desalination, these microorganisms deposit on the surface of the membrane and grow, damaging the whole surface warranting the replacement of the membrane.

Hence, it is necessary to ensure the absence of bio-organisms in feed. This can be achieved to some extent by chlorination of the feed. At the same time, one must ensure the complete removal of chlorine from the feed before entry to the membrane device. Ultrafiltration which can efficiently and physically remove microorganisms has become a popular pretreatment step for RO desalination (Wolf et al., 2005).

The pretreatment for reverse osmosis desalination of seawater or brackish water is similar and has three segments namely removal of particulate matter including suspended matter and colloids (foulants), scale control measures and chemical control measures to prevent deterioration of membrane both chemically and biologically.

#### 5.1 Particulate and Foulant Removal

Screening, coagulation and flocculation, clarification, sand filtration, activated carbon filtration and micron cartridge filtration are some of the unit operations conventionally used for removing the foulant materials (Sohn et al., 2011). Depending on the original quality of seawater some of the operations may be skipped. For example, if seawater is drawn from a depth of more than 5 m or from the beach wells, suspended load will be less (normally less than 10 NTU) and hence one may skip, coagulation and clarification step and use only sand filtration, activated carbon filtration and cartridge micro-filtration. Activated carbon filters have been found to remove high molecular weight organics, most of the microorganisms and the residual chlorine present in the water. The advent of ultrafiltration has afforded the replacement of micron cartridge filters which require frequent replacement due to plugging. Ultrafiltration effectively removes microbes, both living and dead cells ensuring high quality of treated water and is amenable for in service cleaning by programmed backwash cycle (Monnot et al., 2016). Particulate removal for underground brackish water is much simpler as the source water may not have much load of suspended solids or microorganisms. It may suffice to use only sand and dual media filters for particulate removal.

# 5.2 Scale Control

Scaling is a major problem in desalination plants where water is preferentially removed from the saline feed (Kapoor et al., 2020). With the continuous removal of water, the ions present in the solution gets concentrated resulting in increased ionic strength and concentration of sparingly soluble salts. The major scale forming species include carbonates and sulphates of calcium, strontium, barium, chemical compounds of having iron, aluminium and silica. Inverse solubility of calcium carbonate and sulphates with temperature may not be much of a concern, as reverse osmosis desalination plants work at ambient conditions and the temperature changes are only marginal even when part of the reject stream is recycled along with the fresh feed. Two approaches are together followed as a part of scale control strategy: careful process design and addition of proprietary polymeric anti-scalants. Despite the adopted strategies, formation of scales cannot be altogether avoided due to the process limited inherent formation of the boundary layer. Thus, apart from preventive measures, cleaning of the membranes periodically may also have to be adopted as a curative strategy.

#### 5.3 Chemical Control

Chemical control is necessary to minimise the chemical deterioration of the membrane as the commercial membranes are susceptible to chlorine attack and deterioration under oxidising conditions. Chlorine is necessarily dosed at the intake mouth to minimise biofouling of the feed flow through the entire pretreatment system. The dosing of chlorine is decided by trial and error method with a goal of having a little amount of residual chlorine at the exit of all filtration units. The chlorine required would depend on the biological load present in the feed water.

Commercial polyamide based membranes which are the work horses of the desalination industry are susceptible to chlorine attack and suffers oxidative degradation in the presence of oxygen or oxidising agents. In view of this sodium meta-bisulphite (SMBS) is dosed and the water quality checked before entry to the suction of the high-pressure pump.

# 5.4 Monitoring of Pretreatment System Performance

The pretreatment performance is checked, with respect to fouling potential and oxidation potential of the treated water. With reference to scaling only the dosing rates of anti-scalants are maintained as per design specifications. Earlier years pH values of the feed were maintained around 6 by controlled acid dosing to minimise carbonate scaling but with the availability of efficient anti-scalants, the addition of acid is no more in practice for seawater desalination.

#### 5.4.1 Fouling Potential

Silt constitutes all types of suspended particulates that accumulate on the membrane surface and are quantified by nephelometric turbidity units (NTU). NTU is dependent on particle size and the number of particles and is not very much dependent on microbial load. Assessment of NTU is done to ascertain the performance of particulate filters such as sand filters.

Silt Density Index (SDI) relates to the simulated performance of the membrane with reference to the treated feed water quality and hence to the overall pretreatment efficiency. SDI considers the microbial load by using standard 0.45  $\mu$ m filter paper (normally used to filter microorganisms in bio-chemical laboratories). The empirical testing method (ASTM, 2002) is carried out in situ to assess the fouling potential of the pretreated feed being introduced into the membrane element and is widely accepted (Fig. 9). The SDI values indicate the extent of particulate fouling that may occur on the membrane and serve as a warning signal for initiating necessary action (Rachman et al., 2013).

The basic principle involved is to measure the change in water flow rate through a standard 0.45  $\mu$ m filter during a fixed interval of time (5 or 15 min) physically under standard conditions of 30 psig pressure. The water under testing is allowed to flow through the filter for the specified duration, 5 or 15 min. The test involves measurement of time required for collecting 100 mL of filtered water at the zeroth time 't<sub>0</sub>' (at the start) and later after 5 min (t<sub>5</sub>) or 15 min (t<sub>15</sub>). SDI<sub>5</sub> or SDI<sub>15</sub> is calculated using the formula

$$\mathrm{SDI}_T = \left(\frac{\left[1 - \frac{t_0}{t}\right]}{T}\right) 100\tag{2}$$



Fig. 9 Slit density index test skid

where  $t_0$  is the time taken for collecting 100 mL at the beginning, while *t* refers to the time required for collecting 100 mL after the time interval '*T*'.

For example.

 $t_0 = 10 \min$  and  $t = 30 \min$  and  $T = 15 \min$ 

By using the above equation  $SDI_{15}$ , works out to be 2.2.

For a test duration of 15 min, the maximum SDI value is 6.67, as 't' tends to infinity ( $t_0/t = 0$ ), SDI becomes 100/ T. For other intervals, the maximum values of SDI<sub>T</sub> would correspond to 100/T. SDI<sub>15</sub> is popular and is specified by many membrane manufacturers and SDI<sub>5</sub> is occasionally used.

Spiral-wound modules used in reverse osmosis and nanofiltration generally require an SDI < 5 while hollow fine fibre elements require an  $SDI_{15} < 3$ . Based on experience, for spiral elements, SDI15 values less than 3 would give a long service without significant fouling. SDI<sub>15</sub> of less than 3 would ensure smooth operation with occasional cleaning. SDI<sub>15</sub> values between 3 and 5 would require more frequent cleaning and anything above 5 is not conducive for reverse osmosis desalination. With ultrafiltration in the pretreatment system, one can achieve much lower values, close to 2 allowing the freedom to operate at higher specific recoveries and long intervals between the cleaning cycles. The SDI values vary with temperature as the flow rate changes in the range of 25-35 °C due to changes in viscosity of water and hence are not comparable. The measurements must be made at a constant temperature within a variation of about a degree Celsius.

SDI values can also be converted to plugging factor (PF) by dividing by the value obtained by the maximum value possible. For  $SDI_{15} = 2$  the plugging factor would be  $2/6.7 \times 100 = 29.8\%$ .

For  $SDI_5 = 6$  the plugging factor would be  $6/20 \times 100 = 30\%$ .

Thus, plugging factor values enable us to compare the SDI measurements made for different durations. Largest SDI values in each of the methods would correspond to 100% plugging factor.

#### 5.4.2 Oxidation Reduction Potential (ORP)

It refers to the environment under which the membrane process is in operation. As the membranes used in desalination are organic membranes based on polyamides they degrade in the presence of chlorine (attributable to N chlorination) (Al-Abri et al., 2019) and are also susceptible to oxidation. Therefore, it is necessary to keep a reducing environment by ensuring a positive reduction potential. Monitoring the same using ORP metre online before the feed enters RO section.

# 6 High-Pressure Pump and Energy Recovery Devices

#### 6.1 High-Pressure Pumps

Treated water with requisite specifications is sent to the suction of the high-pressure pump. Pumps can be either positive displacement type or centrifugal type. The choice of the pump requires consideration of energy recovery system to be adopted. In reverse osmosis desalination, the pretreated feed is pressurised significantly above the osmotic pressure of the feed to produce relatively pure water. The extent of pressure required depends on the fraction of feed recovered as product water and its purity. As the concentration of feed water increases with progressive removal of relatively pure water, the osmotic pressure also increases leading to less productivity and scaling threat, making it impossible to recover 100% water. Under the current scenario, seawater desalination plants operate, recovering anywhere between 35 and about 50% water, discharging a significant amount of concentrated feed at high pressure representing a major portion of unutilised energy. With a view to reducing the specific energy consumption (power required to produce one unit of water), it has become imperative to recover energy subject to economic feasibility. Hence, the choice of a high-pressure pump must be in tune with the energy recovery system to be deployed. In the initial years, large capacity RO desalination plants were utilising multi-stage centrifugal pumps and the corresponding energy recovery devices, whose efficiencies were less. At present positive displacement pumps with aligned energy recovery systems are deployed leading to low specific energy consumption for the reverse osmosis desalination process.

Both types of pumps have their own merits and challenges (Schunke et al., 2020). Centrifugal pump delivers the feed at designed pressure and flowrate continuously. These pumps run at a very narrow range of pressure head and flow rate, without much flexibility to change the flow rate or pressure head in a significant manner when contingencies arise. Variable frequency drives can be used to alter the characteristics to some extent (Nagaraj et al., 2010) to save energy instead of using throttle valves. Multi-stage pumps have been used for large capacity reverse osmosis desalination plants along with energy recovery devices that absorb the energy available in the reject stream either mechanically or hydraulically to discount the energy required for the feed pump. No doubt, they are compact and require less footprint. However, for small capacity seawater desalination plants, centrifugal pumps are not economically viable as the number of stages for raising the pressure becomes high. The efficiencies of these pumps are normally around 0.7. However,

in brackish water reverse osmosis desalination plants, where the operational pressures are much lower centrifugal pumps are used.

Positive displacement type of pumps can deliver desired flow at any pressure nearly at constant efficiency. Minor variations in volume with time have been overcome with buffers. The maintenance requirements are relatively high compared to the centrifugal pumps and require a relatively large footprint area. However, the efficiency is high (0.95 or more) compared to centrifugal pumps. In fact, efficiency increases marginally at higher pressures. Positive displacement pumps can handle variations in pressure and flow with almost constant efficiency. In practice, a greater number of positive displacement pumps are installed for easy handling and maintenance along with energy recovery devices operating on the same positive displacement principle.

#### 6.2 Energy Recovery Devices

Energy recovery devices play an important role in reducing the specific energy consumption of seawater RO desalination plant, as it recovers the unutilised hydraulic energy available with the reject stream, directly or indirectly and transferring to the incoming feed. Consequently, the power rating for the high-pressure pump becomes less, saving energy and space (Guirguis, 2011). Without energy recovery, the specific energy consumption was around 9–10 kWh/m<sup>3</sup> in the initial phase of technology. The specific energy consumption for large capacity seawater RO plants has been progressively brought down over three decades from about 7 to about 3 kWh/m<sup>3</sup> with various developments in membranes, energy recovery devices and high-pressure pumps.

Energy recovery devices used in the initial stages was of centrifugal type like 'Francis turbine', Pelton wheel or hydraulic turbo chargers (Arenas Urrea et al., 2019). In the last decade, 'isobaric chamber energy recovery' devices have become popular and most of the newly installed reverse osmosis desalination plants use them where the hydraulic energy of concentrated stream is directly transferred to the incoming feed stream. Compared to centrifugal energy recovery devices, isobaric energy recovery devices are highly efficient as it involves single energy conversion step, i.e. direct transfer of energy from concentrated stream to feed steam. Centrifugal devices have two step energy conversion from hydraulic energy of the concentrated stream to mechanical energy (of the drive) and thence to the hydraulic energy of the feed stream thus resulting in reduced efficiency.

Isobaric energy recovery devices can be classified as rotary or piston driven. In rotary type, low-pressure feed is filled in a central rotor sealed and then the high-pressure feed is introduced, which in turn pressurises the low-pressure feed pushing it to the reverse osmosis membrane modules. The chamber is again filled with low-pressure feed emptying the depressurized concentrated stream and again the high-pressure concentrate is introduced thus allowing the cycle to repeat. This device, commercially known as 'PX (Pressure Exchange)' is highly efficient. The other characteristics include compactness, modularity and sturdiness making it a preferred option for seawater reverse osmosis desalination plants (Arenas Urrea et al., 2019; Farooque et al., 2004; Kadaj & Bosleman, 2018). 'Piston-driven' device follows the same mechanism of pressurisation-depressurization cycle inside cylindrical chambers and is controlled by 'switcher valves'. However, piston- driven isobaric energy recovery devices are less compact requiring more capital investment and frequent maintenance compared to PX devices (Guirguis, 2011).

The efficiency of the energy recovery device, defined as a ratio of 'increase in feed pressure' to the 'decrease in concentrate pressure' is about 0.95–0.97 for isobaric devices where for the centrifugal devices the efficiencies are in the range of about 0.75–0.85 (Arenas Urrea et al., 2019). For small capacity seawater reverse osmosis desalination plants, energy recovery devices are not economically viable and hence is not part of the desalination scheme. Energy recovery devices are not useful as brackish water plants normally operate at high recoveries, low pressures and small capacities.

# 7 Membrane Module Dispensation

Spiral membrane element is used in a majority of the reverse osmosis desalination plants whether small or big. The discussion henceforth considers spiral configuration alone. The following terminologies shall be used while dealing with the design of the reverse osmosis membrane section. Figure 10 represents the schematic view of reverse osmosis desalination.

# 7.1 Terminologies Normally Used in Reverse Osmosis Desalination Plants

*Raw feed water*: water drawn directly from the source. *Salinity*: expressed as parts per million (ppm) equivalent mg/L.

*Feed water*: pretreated and quality monitored water entering the suction of high-pressure pump.



Fig. 10 Schematic of single-stage RO desalination

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*Permeate*: purified water emerging out of the membrane module.

*Product water*: post treated permeate water suitable for supply to the user.

*Feed water salinity*: total dissolved salts content in parts per million in feed water.

*Product water salinity*: total dissolved salts content in parts per million in product water.

*Permeate water salinity*: total dissolved salts content in parts per million in permeate water.

*Reject / Concentrate water*: feed water remaining after recovery of permeate.

Solute rejection (%): (feed water salinity – permeate water salinity) /feed water salinity  $\times$  100.

*Recovery*: permeate water/feed water  $\times$  100 (can be ratio of rates or quantity in fixed time).

*Pretreated water*: raw feed water after pretreatment for use in reverse osmosis membranes.

*Membrane*: membrane polymer coated on a support exhibiting the properties of preferential separation of water. *Membrane element*: the membrane made into a spiral configuration through which feed water can be sent and permeate is drawn.

*Membrane module*: Membrane element or elements accommodated into a pressure vessel and ready for use in the field.

*Module bank*: consists of a set of modules with parallel feed flow.

*Staging*: Reject water of the first bank used as a feed for the subsequent bank.

*Multi-stage*: When two are more stages are involved in the plant.

*Pass*: Permeate of first module bank used as feed for the succeeding module bank.

*Array*: Arrangement of module banks either in series or parallel configuration.

Specific Energy consumption: energy required for producing one unit of water  $(kWh/m^3)$ .

*Specific production rate*: amount of permeate produced per module.

*Specific recovery*: fraction of permeate to feed per membrane element.

# 7.2 Performance Characteristics of Reverse Osmosis Membrane Element

# 7.2.1 Effect of Pressure and Temperature

Separation through the membrane as encountered in reverse osmosis is a surface phenomenon. Operating pressure should be greater than the boundary layer osmotic pressure. The driving force is equal to the pressure difference between the operating pressure and the osmotic pressure of the boundary layer. Since the solute permeability is normally less than 1%, the osmotic pressure of the permeate is neglected. If it is significant, the net driving force would be more to that extent. Permeate flux is directly proportional to net differential pressure normally referred to as transmembrane pressure (TMP). Solute flux is directly proportional to the concentration gradient across the membrane and is independent of pressure. Increasing operating pressure increases water flux but at the same time increases the boundary layer concentration as well. Therefore, there is a limiting pressure beyond which water flux cannot increase and solute rejection also would pass through a maximum. Similar would be the behaviour with temperature. Increase in temperature results in a reduction in viscosity and an increase in osmotic pressure. For the same productivity, the plant can be operated at reduced pressures which in turn results in savings of energy (Al-mutaz & Al-ghunaimi, 2001).

# 7.2.2 Membrane Element, Module, and Bank

For use in desalination, the prepared polymeric membrane is configured into spiral configurations called membrane elements. Spiral membrane elements in cylindrical shapes are available from the membrane manufacturers with diameters ranging from 2" up to 16". All the membrane manufacturers use FPS units for specifying the membrane element dimensions. First two digits correspond to the diameter of the element and the next two digits as the length of the membrane element. For example, 8040 refers to 8" diameter 40" length. For large capacity plants normally 8" or 16" modules are used, while 4" elements are used for small to medium capacities. When the diameter is increased by a factor (say 2), the membrane surface area roughly increases by the square of the factor (i.e. 4 times) of 4. Normally an 8" element has about 35 m<sup>2</sup> of membrane area compared to about 140 m<sup>2</sup> for 16" element. These figures may vary for different manufacturers depending on the dimensions of the accessories such as spacer. The performance of each model of the membrane element with respect to specific water flux, specific recovery and solute rejection under a standard condition of seawater or brackish water is provided by the manufacturers. Based on the information, it is possible to estimate the approximate performance of the membrane element such as permeate rate and solute rejection, knowing the feed salinity and operating pressure.

A module consists of a single membrane element or multiple elements connected in series, assembled in a single pressure vessel, ready for use. Depending on the process design, each module may contain anywhere up to 8 elements connected in series. The high-pressure pump delivers the pressurised feed into membrane modules. In such an arrangement the feed flows from the first element in series to the last element successively. The following can be envisaged in this context.

- The concentrate of the first element becomes the feed to the second, and so on. The product water tubes (central pipe) of all elements are coupled and connected to the module permeate-port. Consequently, the initial pressure at which the feed enters gets reduced by a few psi (about 0.1 bar) per element at the initial period of operation and may increase with time or mal-operation (excess pressure drop would indicate accumulation of foulants or scale formation on the membrane surface). This is normally termed as 'system pressure drop'
- 2. As the permeate water flows out of the membrane element, the volumetric flow of feed will reduce in each successive membrane element.
- 3. The feed concentration will keep increasing in every successive element.
- 4. The permeate rate will keep reducing for every succeeding element in series due to reducing driving force consequent to the gradual increase in the osmotic pressure of the feed.
- 5. With increasing recovery, the permeate quality also marginally decreases, as more solutes diffuse towards the permeate due to progressively increasing concentration gradient.
- 6. Higher operating pressures lead to higher permeate output but in the long run probability of scale formation increases due to higher boundary layer concentration.

Accordingly, the design of the membrane section requires optimization amongst the operating parameters, flow rate, operating pressure, and recovery in accordance with the post pretreatment feed water parameters, particularly SDI values. Since the membranes and the feed spacers used by different manufacturers have minor variations, the final products have characteristics particularly different from each other albeit marginally. Based on the internal experiments, each manufacturer has set their own specifications with reference to the feed rate, concentrate to permeate ratio related to SDI values or source of water. Hence, it is necessary that the design aspects of the membrane section shall be in accordance with the membrane element chosen for use in RO modules. Each of the manufacturers has their design software and is available such as 'Winflows' of Suez Water Technologies, ROSA of Filmtec and IMS Design of Hydranautics of Nitto Group to name a few. The range of feed flowrates, the maximum recovery per single element and the permeate to reject ratio of the membrane elements are specified by manufacturers. The number of elements in a module is accordingly decided. In most of the designs using 8" elements around six to eight elements are assembled in a module for achieving around 40-50% recovery. Since the flow rate is limited for each module, the desired capacity of the plant can be achieved by having a greater number of modules in parallel. In the membrane section, the recovery dictates the number of elements in a module connected in series, while the capacity determines the number of modules in parallel.

#### 7.2.3 Stages and Passes

Sustainable performance of the modules is reasonably ensured by following the restrictions on feed flow rate and specific productivity of membrane elements but limits the recovery and permeate quality. In a single-stage system, two or more modules are arranged in parallel. Feed, concentrate and permeate lines are connected to manifolds. Single-stage system is typically used in seawater desalination plants where the recovery rate does not exceed 50%. For achieving high permeate recoveries, more membrane elements cannot be added in series due to scaling threats. In such an event, the concentrate from all the modules are collected in a common header and redistributed to a smaller number of modules complying with the flowrate constraints. Sometimes a booster pump is also used for increasing the pressure. Depending on the design recovery number of elements in each of these second stage modules can be limited to a lesser number. This arrangement by which the recovery is increased is known as 'staging'. Two stages are used to achieve recovery rates up to 75%. The number of modules per stage decreases in the flow direction.

To achieve better permeate quality, the permeate from the modules can be processed through another reverse osmosis module (Fig. 11). The additional cost would be marginal as the process would require neither pretreatment nor high pressures (due to low osmotic pressure of the permeate stream). Since the feed (permeate of the first stage) is free from scaling and fouling species, high recovery (>90%) and high productivity of the membrane modules can be achieved. This concept by which the permeate is treated by one more reverse osmosis unit is known as 'Pass'. However, an additional pump is necessary to pressurise the permeate before introducing it to the modules.

In general, staging leads to an increase in recovery but deterioration in permeate quality and passing improves the purity of permeate but decreases recovery. Multiple stages can be used for achieving high recoveries while passing is not done more than once. Recycling of a fraction of concentrate can be done to maintain constant permeate quality in brackish water desalination plants where the raw water salinity changes with seasons. To improve recovery in the event of passing, the concentrate of the second pass is mixed with the original feed. This has twin advantages: dilution of original feed resulting in higher permeate rate and lower permeate salinity.

# 7.3 Post Treatment

Water permeating out of membrane modules in seawater reverse osmosis desalination is slightly acidic, as dissolved carbon-dioxide is not efficiently removed and contain some



Fig. 11 Concept of stage and pass in RO desalination

dissolved solutes, mostly sodium and chloride with small amounts of calcium, magnesium, and traces of boron. The concentration depends on the membrane used and its age, apart from the design parameters including operating pressure and recovery. Boron concentration in seawater is in the range of 0.5-9.6 mg/L (Tang et al., 2017). The permeate is corrosive as it consists mostly of sodium and chloride ions with insignificant levels of multivalent species such as calcium, carbonate, and magnesium. The permeate contains between 70 and 450 ppm of dissolved solutes and is corrosive with a negative Langelier Saturation Index (LSI). As per WHO drinking water standards, drinking water shall contain about 100 ppm CaCO<sub>3</sub> hardness (at least 10 mg/L of magnesium ions and 30 mg/L calcium ions as a precaution against heart diseases) with overall salinity less than 500 ppm and boron concentration limited to 0.5 mg/L. Boron content of seawater varies with the geographical location and higher concentrations of boron were found in Arabian Gulf seawater. Several methods were tried for boron removal in the pretreatment using mineral clusters (Cho et al., 2015), post treatment stage using specific ion exchangers (Jacob, 2007), and use of two pass RO with new generation reverse osmosis (RO) membranes without pH adjustment (Farhat et al., 2013).

The permeate can be passed through a lime column to adjust the pH and hardness content or blended with pretreated ground water (Nada et al., 1987). For industrial use, the requirements may vary from ultra-pure water to soft water and post treatment methods would vary involving mixed bed exchangers, electro-deionisation (EDI) or capacitative deionization (Čuda et al., 2006).

# 8 Design Methodology for Seawater Reverse Osmosis Desalination Plants

A typical flowsheet of seawater reverse osmosis desalination plant is shown Fig. 12.

The basic steps involved in the design of reverse osmosis desalination plants are listed below.

- 1. Fix the capacity of the plant and the quality of product water to be delivered.
- Assess raw water quality in terms of foulants, suspended matter, bio-load, total dissolved solids.
- 3. Identify the pretreatment system requirements and expected quality of pretreated water.
- 4. Use the software program of the selected membrane element manufacturer with inputs of water quality and capacity and estimate the operating pressure, number of elements required and configuration of modules that yield the required quality and the percent recovery.
- 5. Choose the energy recovery device and work out the net power consumption.
- 6. Identify the dosing chemicals suggested and the requisite approximate dose rates.
- 7. Design the post treatment system and the cleaning system.
- 8. Design the distribution system.
- Prepare the specifications for all the equipment including pumps, energy recovery system, pre and post treatment, instrumentation, and control systems.

# 9 Brackish Water Desalination

The steps are similar for brackish water or industrial wastewater. Since the operating pressure is less and recovery high, the energy recovery device may not be viable. The challenge in designing brackish water desalination plants is the change in salinity with seasons and the continuous withdrawal of ground water. Unlike seawater desalination, brackish water design objectives must balance economics and conservation of groundwater to ensure sustainability. Reject recirculation is suggested to maintain constant



permeate quality and conserve water when the salinity is less (Sarkar et al., 2008). Sensitivity analysis for at least a 10% change in water quality is desirable and the pumps and pretreatment system are designed carefully to accommodate the fluctuations.

10 Operation and Maintenance

Operator efficiency plays a critical role in the sustainable performance of reverse osmosis desalination plants. Maintenance of pretreatment parameters particularly ensuring the absence of chlorine and maintaining a reducing environment is important for the chemical integrity of the membranes. Dosing of anti-scalants and operating the plant within the design range of pressure and flow rates are necessary to minimise fouling and scaling on the membrane surface. Whenever the plant is stopped for any reason temporarily, it is essential to ensure that the product channel is full so that the boundary layer of the membrane gets diluted by osmosis. Suck-back tanks are normally provided for this purpose. The total dissolved contents of seawater may be less particularly when it is drawn from shallow depth during the rainy season and care must be taken to operate the plant at reduced pressure lest specific recovery may increase leading to heavy scaling and fouling and consequent deterioration of membrane performance.

Process control and instrumentation are relatively simple in reverse osmosis-based desalination plants. Most of the instruments like flow metres, pressure gauges, oxidation reduction potential (ORP) and salinity (conductivity) metres are indicative in nature and based on the values seen the operator may have to control the operations. Control may be needed if variable frequency drive is used for the high-pressure pump (Alatiqi et al., 1999).

#### 10.1 Membrane Cleaning

Despite best operating practices, scaling does occur on the membrane surface and cleaning becomes mandatory. Frequent cleaning may lead to deterioration of the membrane. Normally cleaning becomes necessary when the fouling and scaling build on the membrane surface to such an extent that the membrane flux (normalised with respect to operating pressure) reduces by about 10-15% or the solute passage increases by more than 10% or the feed flow pressure drop increases more than the expected values. The accumulated species on the membrane surface may be a mixture of loose particulates, bio-organisms, compounds of calcium, strontium, aluminium, silica, iron, etc. The cleaning processes include flushing, soaking and rinsing using a variety of chemical solutions including acids both mild and strong, bases, complexing agents, detergents, surfactants, etc. (Ang et al., 2011). Several proprietary chemicals are available in the market for cleaning. The best practice, however, is to follow the procedures and chemicals prescribed by the concerned membrane manufacturers.

The membrane elements used in reverse osmosis deteriorates slowly with time reflected by falling solute rejection and reduced water flux. When the permeate concentration is beyond the acceptable limits the membrane elements are 96

considered defunct and replaced. Earlier an increase in the solute passage of more than 15% was considered as the indicator for replacement. With a second pass reverse osmosis in place, the limit is relaxed.

In practice, all the membrane elements in the series do not deteriorate to the same extent. It is observed that the first element which has the maximum net pressure differential as driving force and high specific recovery, deteriorates faster due to deposition and compaction of foulants including bio-materials. On the other hand, the last element in the series encounters maximum feed concentration and least fluid velocity and hence is prone to more scaling. In view of these considerations, the present practice is to discard the first element after few months or a year of operation and introduce a new element at the end of the module and push all the elements one step ahead. This procedure not only allows the membrane replacement to spread over a long period but also has a possibility of having a nearly uniform permeate quality.

#### 11 Environmental Aspects

Reverse osmosis desalination process is an ambient temperature process and does not have any thermal discharges. Seawater intake and discharge of the concentrated brine discharges from the pretreatment system, periodic discharge of wastewater during chemical cleaning and ultimate disposal of used membrane elements are of concern to the environment. Besides, there are minor concerns on noise pollution due to the running of high-pressure pumps (Tularam & Ilahee, 2007). During intake particularly from open sea aquatic organisms may be trapped by screens and may cause either resistance to flow or get carried into the plant and affect the fishing activity. The discharges are the major concerns as they contain up to double the initial concentration of solutes and small amounts of chemicals dosed during the operation. Normally, the discharges are dispersed along the coast into the sea through long pipelines, so that the salinity of seawater is not significantly changed, and marine organisms are not unduly affected. The dosing chemicals are present in trace levels and apparently, no effect is observed of these chemicals. There is a possibility that some of these chemicals may get accumulated at seabed or aquatic organisms. However, the information in this regard is yet to emerge. Technically, there is not much of an environmental hazard as such for RO desalination plants. Disposal of the used membranes could be a real challenge as they cannot be bio-degraded, nor can be easily shredded. As more reverse osmosis desalination plants are installed, this aspect requires attention. Because of the inherent strength of the module element probably they can find use as a construction material.

# 12 Economics of Reverse Osmosis Desalination Plants

Compared to all other desalination processes reverse osmosis desalination is found to be economical because of its low specific energy consumption, modularity, and ease of operation (Baron, 1966; Seawater Desalination Costs, 2012). The capital expenditure (CAPEX) has two cost components; one related to the technology components of the reverse osmosis including pretreatment equipment, high-pressure pump and energy recovery device, support structure for membrane elements (excluding membrane elements) as per design configuration, post treatment, cleaning equipment and other comprising of seawater intake system, distribution logistics, financial and project management and the other corresponds to the distribution of the product water. For plants with 50 MLD and above capacities, 50% of the costs relate to technology components while intake accounts for about 20-25%. For smaller capacities, the proportion of intake cost would be high if direct intake from the sea is to be considered. The operational expenditure also has two components: one technology related variable costs including energy, consumables such as membrane elements, chemicals used for dosing, cleaning and post treatment chemicals and maintenance (preventive and curative) requirements and the other relates to fixed operating costs comprising of labour, amortisation, administration, insurance, and other statutory levies. Nearly 75% of the operating costs are accounted by energy, consumables (chemicals, cartridges and membrane elements) and labour. Based on the study conducted by 'Water Reuse Association' in 2012, the cost of water by reverse osmosis desalination process has stabilised in the range of \$0.79 to \$2.38 per m<sup>3</sup> depending on the location since 2005. The reasons for variation are attributed to the country dependent variation of costs with respect to intake systems, energy, construction, finance, labour and environmental regulations. For small capacity plants, the cost of water would be higher particularly with intakes.

#### 13 Future Direction and Expectations

Any developmental efforts should satisfy the criteria of sustainability, affordability, and acceptability. The strategic components of RO desalination technology are energy, membrane, and environmental considerations. The thermodynamic minimum energy at 25 °C is about 0.7 kWh/m<sup>3</sup>, i.e. for drawing one cubic metre of water from the infinite source of seawater (at almost zero recovery). However, in practice the recoveries are significant and hence the minimum energy is somewhere between 1 and 1.5 kWh per cubic metre. Considering the irreversible energy losses in pumps, fittings, etc. the minimum energy attainable maybe around 2–2.5 kWh. On this front, the developments have almost reached the ultimate stage as the efficiencies of energy recovery systems and high-pressure pumps are more than 95% based on the positive displacement concept. Deploying a greater number of pumps and energy recovery systems rather than one large unit ensures flexibility in operation and ensuring sustained productivity considering the maintenance requirements of positive displacement pumps and the demand variations. Suppliers of RO plants are guaranteeing 3 kWh/m<sup>3</sup>. Therefore, any further effort in bringing down the energy consumption may not lead to any significant improvement or benefit.

Brackish water reverse osmosis plants do not use energy recovery systems because of low capacities and high recoveries. They are mostly used for providing safe drinking water in remote villages and isolated areas using saline groundwater whose quality and availability change with time. To conserve the water resources, the reject water should also have moderate salinity which can be used for non-potable uses. Reducing recovery would increase the specific energy cost. In this context development of cost-effective energy recovery systems would help in reducing the energy cost.

The heart of the reverse osmosis desalination process is the membrane. It is considered a consumable requiring replacement periodically during the mechanical life of the plant. Improvements in membrane productivity is a challenging concern. Membrane development with less propensity for fouling and sustainable performance is one step in this direction. Efforts to incorporate benign nanoparticles on the surface has yielded membranes with high flux and sustained high salt rejection and has been commercialised recently. The other development corresponds to the use of CTA based hollow fine fibre membrane elements, which are compact and are resistant to chlorine, an Achilles' heel for commercially popular TFC based membrane elements. The continuing operation and establishment of the CTA-hollow fibre membrane elements production unit at Saudi Arabia is a pointer to the successful run of these membrane elements even though not much operational experience is in the public domain yet. These developments contribute to sustainability as well as cost reduction towards affordability if we consider the life cycle costing.

Increasing recovery for seawater RO desalination plants beyond 50% without booster pumps and without the scaling threat would probably be the next step. A forward osmosis reverse osmosis system looks like a possibility. Effluent water can be used as feed for forward osmosis with raw seawater as draw solute. In this process, the seawater gets diluted requiring less operating pressure and one can get high recoveries without scaling threat. The strategy required is to dilute the incoming seawater so that reverse osmosis plant can operate at lower pressures and higher recoveries.

A few studies have been reported in this regard (Li et al., 2020; Volpin et al., 2018) have indicated the potential of this concept for cost reduction. With the development of aquaporin membrane (Li et al., 2017) it appears to be a good possibility. Further possible developments include simultaneous recovery of energy in FO dilution of seawater following the principle of pressure retarded osmosis (Cheng et al., 2018). A hybrid nano-filtration- reverse osmosis (NF-RO) system for seawater desalination is reported in literature operating at low pressures (Wafi et al., 2019) to be cost effective. The scheme involves the use of nanofiltration as the first stage and reverse osmosis as a second pass by pressurising the permeate of nanofiltration. Since the nano-filtration permeate is free from foulants and relatively free from sparingly soluble salts of calcium and other multivalent species, high recoveries can be achieved without scaling a threat to reverse osmosis membranes. As of now, energy recovery may not be viable because of the low-pressure operation coupled with high recoveries in reverse osmosis section. Since pressurisation is involved in both the steps, specific energy consumption increases even if both steps run with recoveries of 60 and 70% respectively. NF-RO hybrid system does not offer any advantage with reference to energy consumption but enables value recovery. The concentrate of the RO step is rich in sodium chloride compared to normal seawater without much multivalent species and is a more suitable feed stock for caustic production using membrane electrolysers. Alternately the stream can be used in crystallisers for producing pure sodium chloride crystals. Similarly, the concentrate of nanofiltration being relatively rich in magnesium compared to seawater can be used for magnesium production. Of late there is a renewed interest in the recovery of trace heavy metals such as uranium (Abu-safiah, 1991; Pal et al., 2006) caesium, rubidium, etc. (Nur et al., 2016). Complexation/ precipitation ultrafiltration has already been demonstrated to remove trace heavy metals (Kavitha et al., 2018) and its application can be extended for the removal of trace metals from concentrated seawater.

With the proliferation of RO desalination plants across the world, disposal of the membranes after their service life is still a challenge. Presently the spent membranes are disposed of as landfills. Some studies indicate reuse of membrane after the destruction of the TFC layer by oxidising agents (Paula et al., 2017). The elements are strong as they are designed to withstand about 80 bar pressure. Hence, the opening of the sealed membranes or shredding them is an energy and labour-intensive process. Based on the strength, the use of these membrane elements as construction materials requires serious consideration.

In addition to the technological aspects, the cost to the consumer is highly site specific as it includes pre and post technology logistics including intake and distribution. The optimization objective of minimum cost with reference to consumer point is more desirable rather than battery limit cost. In this context, the choice between high capacity plants at a single location versus distributed capacity at different locations is worth considering for savings on the distribution cost, particularly in congested urban cities along the coast. Small capacity plants can use beach-well intake which could be cost effective. The pretreatment cost becomes less because the source and the membrane elements have no scale up advantage. As the current trend is to use multiple pairs of positive displacement high-pressure pumps and energy recovery systems, the basic technology cost also would be proportional. The other strategy could be to co-locate the desalination plants along with power generating stations or industrial complexes which draw cooling water from the sea. This helps in savings of investment as well as operating costs as facilities and skilled manpower

Apart from cost savings through technological aspects of the desalination process, the cost of water from a desalination plant can be reduced by value addition as well. Some of the values that can be recovered from the concentrated stream include sodium chloride salt, caustic by electrolysis process, rare metals, etc. (Al-mutaz & Box, 1987; Pal et al., 2010; Sodaye et al., 2009). This calls for integrating the RO desalination plants with systems to recover value which will make the whole system eco-friendly and cost effective.

#### 14 Conclusions

can be shared.

Reverse osmosis technology is a mature technology capable of supplying relatively pure water in a cost-effective manner both as a point of use device as well as in multi million litres per day capacity. It is less energy intensive, easy to start and stop, amenable to flexible operation. However, there are a few challenges like disposal of reject brine and spent membranes which require serious consideration as the use of reverse osmosis has been proliferating. The technological cost has stabilised, but to overcome the logistics component of the cost, new strategies are necessary based on site specific environment.

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# **Desalination by Membrane Distillation**

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# Abstract

Despite covering approximately 70% of the earth's surface, the limited supply of clean water remains one of the major issues affecting the survival of humanity and the entire living beings and ecosystem. High salt content, high turbidity and salinity make water unsuitable for drinking and general use. Many technologies have been developed to attain clean and pure water appropriate for consumption. Desalination, a technique to remove salt and mineral components present in water, is an effective method in water treatment. Recently, membrane distillation that involves the utilization of hydrophobic membrane to accomplish filtration and purification across the membrane has gained huge attention. Divided into four major categories-direct contact, sweep gas, air gap and vacuum, membrane distillations are now commonly employed and effective for water desalination. Many innovative membrane hybrid systems have been developed to further improve the efficiencies and resolve the challenges especially to meet the needs of clean water-scarce region.

#### Keywords

Desalination • Membrane distillation • Configurations • Modules • Brackish groundwater desalination • Seawater desalination • Produced water desalination • Membrane hybrid systems

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#### 1 Introduction

"Water is the nature's driving force", without which there will be no life. Although water makes up much of the surface of the earth, there is still a severe scarcity of drinkable water in many countries around the globe (Thimmaraju et al., 2018). Many concerted efforts have been made to explore sustainable water resources and to develop membrane-based technologies for water desalination and purification especially from brackish, ground and seawater (AlhathalAlanezi & Abdallah, 2016; AlhathalAlanezi & Sharif, 2012; Ismail et al., 2019; Pal, 2017; Yasukawa et al., 2018; Zheng, 2017). The key objective is to produce freshwater for drinking, cultivation or industrial use. The source of reusable water can be from the processing of brine or condensed feed (such as from mineral power plants) and freshwater from evaporation systems. The productivity and processes must be analyzed and optimized for direct utility and cost competitiveness (Aguirre et al., 2014; Al-Obaidani & Curcio, 2008; Rastogi & Nayak, 2011; Xu et al., 2006), and new materials and strategies need to be developed to improve the efficiency (Elmarghany et al., 2020; Kebria & Rahimpour, 2020; Kim & Kwon, 2017; Morciano & Fasano, 2020; Pangarkar et al., 2011a). More important is to address the supply of clean water issues urgently as more than 2 billion people around the world reside in areas facing extreme water distress, and almost 4 billion people endure water shortages for at minimum one month per year (Morciano & Fasano, 2020). In such circumstances and dire needs, the availability of concentrates may be more useful than freshwater, if the desalination technologies are available and can be implemented (Rahimi & Chua, 2017).

Desalination based on membrane system like reverse osmosis, or minimal-temperature thermal desalination utilizing ocean temperature gradients, is an effective strategy owing to minimal effort of deployment and reduced energy costs. The two key desalination techniques are thermal

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**Fig. 1** Techniques involved in desalination of water. (Modified from Thimmaraju et al. (2018))



desalination and membrane desalination technology, as shown in Fig. 1. Both require different specific methods, some of which are complementary such as freezing as well as ion transfer, but conventional or green energy is commonly used (Thimmaraju et al., 2018). As energy is an essential part of the desalination system, the utilization of green resources should be adopted as the way forward (Jalihal et al., 2019). (MD) Membrane distillation is an isolation mechanism, wherein vapors can move across pores-containing hydrophobic membrane. The distinction is in the variations in the vapor pressure between the membrane layers. Since the membranes are highly porous, the pores could not be flushed with a feed solution. During the process, half part of the membrane interacts with the hot salt water to create the hot end, and another end is in contact directly or indirectly with the cool, clear water to create the cool layer. The vapor going across the membrane is concentrated at the cold part to create the distillate. The standard distillation module of the membrane is shown in Fig. 2. There are primarily four categories of membrane distillation techniques: direct contact membrane distillation, sweep gas distillation, air gap membrane distillation and vacuum membrane distillation. In these methods, the hot inlet mixture is in close interaction with the membrane surface. The membranes employed in the membrane distillation system should be strongly hydrophobic, with high porosity, but poor thermal conductivity. Operating parameters, for instance, water temperature, air gap width, flow rate, membrane width, membrane thermal conductivity, tortuosity, porosity and deep-term activity, have impacts on the distillate output (Pangarkar et al., 2011a; Siyal et al., 2019; Zheng, 2017). Different membrane distillation configurations are being utilized to draw aqueous feed by employing microporous



**Fig. 2** General illustration of membrane distillation employed in water desalination. (Modified from Zheng et al. (2017))

membranes which are generally hydrophobic as shown in Table 1.

The aims of this chapter are to highlight the different membrane distillation configurations and modules. The applications in water desalination and the recent advances are discussed and elaborated.

# 2 Membrane Distillation Configurations

## 2.1 Direct Contact Membrane Distillation

Direct contact membrane distillation (DCMD) is a system, wherein the permeate and feed solutions have direct interaction with a hydrophobic porous membrane as shown in

MD Configuration	Benefits	Drawbacks	Uses
Direct Contact Membrane Distillation (DCMD)	<ul><li>✓ Superior permeate flux</li><li>✓ Reliable at commercial level</li></ul>	Conductive heat loss	<ul> <li>Desalination</li> <li>Crystallization</li> <li>Arsenic removal</li> <li>Dye removal</li> </ul>
Sweeping Gas Membrane Distillation (SGMD)	✓ Lowering mass transport barrier via forced flow	☐ Complex procedure ☐ More threat of temperature polarization	<ul> <li>Desalination</li> <li>VOC removal</li> <li>Azeotropic mixture separation</li> <li>Wastewater treatment</li> </ul>
Air Gap Membrane Distillation (AGMD)	<ul> <li>✓ Simple procedure</li> <li>✓ Minimum conductive heat loss</li> <li>✓ Low risk of temperature polarization</li> </ul>	□ Lower flux	<ul> <li>Desalination</li> <li>VOC removal</li> <li>Azeotropic mixture separation</li> <li>Concentration of fruit juices</li> </ul>
Vacuum Membrane Distillation (VMD)	<ul> <li>✓ Superior permeate flux</li> <li>✓ Reliable at commercial level</li> </ul>	☐ Complex procedure ☐ More threat of membrane pore wetting	Desalination     Wastewater treatment     Aromic compounds     retention

Table 1 Major configurations of membrane distillation employed in water desalination

Data from Kebria and Rahimpour (2020)

Fig. 3. The water vapor passes via the membrane and is concentrated directly in cold permeate within the membrane container. The design is more widely studied because of its basic mode of operation relative to other MD configurations. The DCMD is known to be the safest option for systems where water is the key factor of the supply solution and applicable in chemical, food, medical and environmental sectors (Tomaszewska, 2015a). When the main feed is salt, DCMD is ideal for desalination and extraction of aqueous solutions and is effective for treatment at low temperatures to preserve product quality (Laganà & Barbieri, 2000).

#### 2.2 Sweeping Gas Membrane Distillation

Sweeping gas membrane distillation (SGMD) is a system wherein the sweeping gas is employed to make the hot aqueous feed stream flows toward the porous hydrophobic membrane, as represented in Fig. 3. The membrane present inhibits the liquid from saturating the pores, but allowing instead the vapor/liquid contacts to be established at each pore. Evaporation of the volatile compounds occurs at the pores, and the cool sterile gas running in the other part drives the permeate to separate from the reservoir. Vapor condensation takes place in the active condenser. The driving force in the SGMD is the partial pressure difference of the volatile compounds on each side of the membrane that causes the flow across the membrane. The heat in the MD is transferred from the feed inlet via the membrane readily, and the temperature of the sweeping gas rises substantially throughout the tube (Tomaszewska, 2015b). SGMD has shown great potential for desalination (Perfilov & Fila, 2018).

## 2.3 Air Gap Membrane Distillation

Air gap membrane distillation (AGMD) is a system wherein an air surface is weaved amongst the porous hydrophobic membrane and the condensation layer, as shown in Fig. 3. The volatile compounds and water, which exist in the hot feed, evaporate on the vapor/liquid layer on the surface of the membrane. The vapor passes through the pores, as well as the gas gap, and eventually collected on the cool surface within the membrane container. The mass movement of the feed in the AGMD is driven by the vapor density difference on each side of the membrane (Tomaszewska, 2016). In comparison with other MD designs, the AGMD has no permeate interaction with the membrane and has reasonably high flux, reduced heat loss and low tendency to fouling. The membrane system maintains a static air distance along the membrane surface and the condensation layer. The vaporization of the volatile components at the liquid-vapor interface within the pores is due to the difference in temperature between the feed fluid and the cool surface (Ajdar & Azdarpour, 2019). AGMD has also shown its potential application in the desalination method (Elhenawy & Elminshawy, 2020).

## 2.4 Vacuum Membrane Distillation

Vacuum membrane distillation (VMD) is a recent membrane-based isolation technique developed especially for the processing of "ultrapure" water from the salt solutions, the isolation of dissolved gases and the volatile solvent removal from the aqueous system (Bandini et al., 1997). It involves the use of vacuum in its operation, created by the



Fig. 3 Four major configurations of membrane distillation.  $\mathbf{a}$  direct contact membrane distillation;  $\mathbf{b}$  air gap membrane distillation;  $\mathbf{c}$  sweep gas membrane distillation; and  $\mathbf{d}$  vacuum membrane distillation. (Modified from AlhathalAlanezi and Abdallah (2016))

vacuum pump as shown in Fig. 3. The VMD method has attracted considerable interest in many engineering applications (Hassan & Darwish, 2015).

## 3 Membrane Distillation Modules

## 3.1 Tubular

Tubular membranes as shown in Fig. 4, employed in the VMD, AGMD and DCMD modes, have been utilized to desalinate seawater, brackish water and wastewater. The membrane components are primarily ceramic, polypropylene (PP), polyvinylidene fluoride (PVDF) and polytetrafluoroethylene (PTFE). The membrane assembly is composed of a hydrophobic membrane and a core. In the AGMD tubular design, the feed stream is added externally, and the cold stream is circulated upstream via the hollow copper tubing. The condenser is in the middle of the hollow copper pipe and the insulation shell. The diameter of the tubular membrane assembly is ranging between 10 and 25 mm, with a variation of packaging capacity. The tubular module can have a large flow rate, to reduce the membrane fouling and polarization anomalies, allowing simple cleaning procedure. Unlike hollow fiber membrane, the tubular modules have higher strength and tubular cover, and no support sheet is required. The tubular membrane modules also have higher surface area and can process higher volumetric flow rate than the flat sheet systems and therefore are more preferable (Kebria & Rahimpour, 2020).

## 3.2 Plate and Frame

Figure 5 shows the representation of the plate-and-frame membrane assembly. The membranes and the spacers are alternatively packed like a sandwich and placed in a framework. It is the easiest way of packaging the flat sheet membranes and can be built in a range of ways and sizes, from pilot plant-scale carrying a single, narrow membrane to the structures capable of accommodating higher number of membranes. The major drawback is inadequate membrane reinforcement and smaller packing capacity or effective membrane area per module. The lack of support limits the operation to a minimum hydraulic pressure or at an almost equal pressure on each end of the membrane sufficient for a fairly good process control. Small packing density also leads to a greater footprint, higher capital and running costs such as for membrane auxiliary work. There are also issues related to direct and indirect sealing, challenges in controlling the stability of the membrane and more limited operating conditions such as the fluid velocity and pressure (Rastogi & Nayak, 2011). The plate and frame systems are applied in water management especially in cases of waste



sources producing large amount of fouling or high viscosities (Yasukawa et al., 2018).

## 3.3 Hollow Fiber

The hollow fiber membrane consists of 0.1–1 mm diameter fiber, as shown in Fig. 6. The feed is introduced which moves through the asymmetric membrane placed in the container, and the open exteriors of the U-shaped fibers are positioned on the top plate. Hollow fiber membrane systems have been utilized for gas extraction, waste heat recycling, water treatment, material purification, liquid distillation and pervaporization. The module has small packaging capacity but with vast area of friction between the shell side and the tube side. The tube side can handle heavy pressure although the driving force can be small. Epoxy resins can be sealed at both edges of the pipe. The fibers are bundled together to create a hollow fiber bundle, placed in a wider tube container to create a container and tube heat exchanger system (Zhang, 2013). The hollow fiber membrane exhibits higher surface to volume ratio than the spiral-wound design, to permit higher output and higher desalinated water quality, with minimum footprints (Ismail, et al., 2019).

## 3.4 Spiral-Wound

The spiral-wound modules contain flat sheet membranes, spacers and channels mounted and placed along the drain, as shown in Fig. 7, to attain a wide range of packing capacity while minimizing the volume of storage occupied (Aguirre et al,



**Spiral Wound Module** 

2014). The membrane, a mesh spacer, a permeate protector and a protective sheet for the membrane create a shield that is twisted and curved about a pierced collection channel. The feed flow passes through the exterior of the membrane in an axial course. Following the initial condensation process, the permeate flow is carried across the central tube and deposited in a perforated seep collecting tube. The spiral-wound membrane materials include PP, PTFE, polyvinyl chloride (PVC), polyethylene (PE) and synthetic resins that are used to desalinate brackish and seawater (Kebria & Rahimpour, 2020).

## 4 Applications in Water Desalination

Power-efficient desalination and water management techniques are pertinent to produce clean water with less energy burden. MD for the desalination of high-salinity

waters could utilize smaller-grid or energy generated from wastes, to maximize the renewable supply of electricity, a core component of the water-energy nexus (Deshmukh & Boo, 2018). MD is more economical by using reduced ambient temperatures, relative to those used in the conventional processes, and the product which is primarily water, does not require heating to a boiling point. Furthermore, the hydrostatic pressure in the MD is smaller than the membrane technology which is pressure-driven for instance the reverse osmosis (RO). Hence, the less costly polymeric materials can be used. A complete product separation is also possible based upon the vapor-liquid equilibrium of the MD. However, the number of pores for the MD is comparatively more than the other membrane separation including the RO (Alkhudhiri & Darwish, 2011).

## 4.1 Brackish Groundwater

The deteriorating groundwater conditions from overexploitation and contamination have become a major concern. Treatment of groundwater is necessary to meet the requirements of standards for human consumption. Desalination technique provides avenue for freshwater production from brackish water. MD, with its lesser energy demands and lower operating temperature and pressure, is a cost-effective alternative method, to RO and distillation (Pangarkar et al., 2011b). Brackish waters containing sodium chloride (at 10 g/L) and arsenic (III) (at 300-2000 µg/L) have been processed by the VMD to reach the acceptable permeate standard, with more than 99.5% of salt, and 98.5% of As(III) removal. For VMD, a priori oxidation stage is not required to transform As(III) into As(V), as normally used in other methods, and no effects of fouling from humic acid and calcium, and any scaling of membranes, are observed (Dao & Laborie, 2016). The brackish groundwater desalination has also been carried out using the direct contact membrane distillation (DCMD) with PVDF membranes. The PVDF membrane displays a strong rejection of non-volatile inorganic dissolved salts, with a gross permeate flux of 24.5 kg m<sup>-2</sup> h<sup>-1</sup>, with the feed at 70 °C. The feed content does not have a major influence on the permeate flux and the solute rejection. As the normal groundwater is fed into the DCMD, the CaCO<sub>3</sub> accumulation could clog the membrane to reduce the clean water output. Acidification of the feed could reduce the negative effect of scaling and prevent any deposits on the membrane surface. The DCMD is effective in producing clean drinking water from brackish groundwater, with a water retention of up to 75% (Hou & Wang, 2010a). The PVDF membrane has demonstrated a strong degree of inorganic salt and solute rejection, achieving permeate flux of 35.6 kg m<sup>-2</sup> h<sup>-1</sup> with a feed at 80 °C and a cooling water at 20 °C. The feed composition, however, does not affect the permeate flux and the removal of the fluoride. With time, the output starts to decrease primarily due to the CaF<sub>2</sub> particle depositions on the membrane surface (Hou & Wang, 2010b).

Microporous, hydrophobic hollow fiber membranes (HFMs), the PVDF and PTFE, have been tested in the DCMD. Water flow and salt discharge are calculated as a feature of unit packaging density and duration in the laboratory-scale setup. The PVDF HFMs typically display increased water movement as compared to the PTFE HFMs, due to the thin membrane walls and reduced porosity. When the density or duration of the unit is enhanced, the water flow decreases. Nevertheless, the amount of water output per module is improved owing to the greater membrane surface area. In the United States, a pilot DCMD project has been carried out at the second largest geothermal-heated

greenhouse as a field trial, over the period of twenty two days. The findings suggest the applicability and robustness of the DCMD module operated under varied conditions in the plant (Huang & Arning, 2019). The DCMD at a low feed temperature (55 °C) makes it possible to concentrate a synthetic solution of fluorine polluted groundwater. The average initial flow of 13.3 L/m<sup>2</sup>/h results in 75% recovery of water, with a mere 15-17% reduction in flow, yielding a permeate with 96-99% of F removal. The presence of S, Mg, Pa and Ca has been detected on the membrane surface. In comparison, the DCMD vacuum operation on the permeate side has been successful in enhancing the permeate flow by 42%. The continuous operation of the vacuum-enhanced DCMD with membrane cleaning in between produces 67% permeate, which is promising in the treatment of F-contained groundwater (Plattner & Naidu, 2017).

A system comprising a fluidized bed crystallizer (FBC) and an AGMD cell has provided new insights into the characterization of the fluidized surface, the development of the CaCO<sub>3</sub> seed particle size and the AGMD operational parameters. The technological viability has been demonstrated on the experimental scale (Bouguecha & Dhahbi, 2003). The effectiveness of two separate scale management strategies has been evaluated: a higher pH preconditioning system; and with an anti-scalant (AS) application, for RO concentration reduction in the laboratory-level AGMD. As compared to the earlier studies utilizing a DCMD model, the efficiency of the AGMD batch setup has been evaluated with higher salinity, alkalinity, SiO<sub>2</sub> and magnesium hardness in the RO concentrates, both in the presence or absence of the method to lessen the scale-forming components. The high-pH pre-treatment utilizing cool lime-soda ash cycle is shown to be more efficient than AS, leading to greater salt precipitation where the concentration factor (CF) in the AGMD configuration increases beyond 1.3. The greater precipitation under increased pH has resulted in substantial reduction of magnesium (96%), SiO<sub>2</sub> (96%) and calcium (86%). As shown in Fig. 8, based on this chemical demineralization technique, the AGMD system could withstand high temperature, with enhanced permeate flow (Rioyo & Aravinthan, 2019).

Modified pure PVDF nanofibre membranes with silica nanoparticles (SiO<sub>2</sub> NPs) and with organically functionalized (salinized) silica nanoparticles (f-SiO<sub>2</sub> NPs) have been tested for brackish water desalination. Highly hydrophobic f-SiO<sub>2</sub> NPs-functionalized PVDF membranes (with contact angle of  $156 \pm 6^{\circ}$ ) have demonstrated an improved water flow and smaller flux degradation than that of the pristine and SiO<sub>2</sub> NPs-functionalized PVDF membranes, under the conditions of lessened salinity (LS) and elevated salinity (HS) in the feed fluid. However, the larger organic **Fig. 8** Illustration of air gap membrane distillation for brackish groundwater desalination. (Modified from Rastogi and Nayak (2011))



component of the LS in the feed increases the wetting and fouling in both pure and improved membranes (with flux deterioration of close to 73.6% in the neat membrane samples). A hydrophilic coating, composed of silver nanoparticles (Ag-NPs) and functionalized multi-walled carbon nanotubes (f-MWCNTs), has been deposited on the pure and improved membranes to confer anti-fouling properties. The layer reduces considerably the flux loss and improves the tolerance to losses in the salt refusal of pure, SiO<sub>2</sub> NPs-functionalized and f-SiO2 NPs-functionalized PVDF membranes, in LS and HS feed mixture. The hydrophilic coating layer promotes fouling resistance characteristics, strong brine-refusal and lower flow degradation in the hydrophobic PVDF nanofibre membranes, suggesting the ability to obtain drinking water from brackish water using the MD systems (Nthunya & Gutierrez, 2019). The multiple fouling prevention methods during the processing of brackish groundwater demonstrate the effectiveness of concentrating by four-fold the concentration level to significantly minimize the amount of wastewater. Careful use of fouling management strategies has been proven successful to minimize the subsequent fouling. Intense agitation, like stirring, may cause premature membrane degradation especially when dealing with brackish groundwater which is susceptible to inorganic fouling. The thermal crystallization system has been successful in enhancing the membrane output and facilitating the processing of calcium carbonates. By means of transverse vibration, the concentration is increased by 8.7-fold in the processing of the brackish

groundwater with humic content, at 70  $^{\circ}$ C (Zhong & Ji, 2018).

Different design configurations of a hybrid system have been incorporated including saline recycling and cascading by simulations. The improvement in the efficiency is attributable to the system integration, which extends the shelf-life and reduces the energy usage. A multi-stage hybrid device connected in series could have the shelf-life extended by 90% and reduce the actual energy usage to 0.9 MJ/kg. The actual energy costs may escalate significantly from the use of external inter-stage heating, and the use of a free energy supply (including a geothermal or waste energy system) could offset the extra cost and offers an optimal setup (Ali & Orfi, 2018a). An exergy analysis on the standard MD module has been carried out to find the sources of inefficiency in the operation of an MD for brackish water desalination. The use of concentrated brine causes the depletion of resources where the geothermal feed capacity is not efficiently utilized. The output volume and energy consumption can be optimized by placing the output line to consecutive MD units attached in sequence. With this, the exergy risks show the reduction from 70 to 30%. To address the hot permeate which results in further heat loss to the atmosphere, the repeating outlet could recycle the permeates in consecutive MD reservoirs, connected in parallel, to enhance process efficiency. The desalination cycle in cascade configuration utilizes more of the usable exergy from the geothermal supply to generate sufficient freshwater and also removes the saline brine and hot permeate, at reasonably low levels of exergy (Ali, 2017). A conceptual model of the MD system has evaluated the weight and energy ratios which are balanced against the collected distillate output rate as well as the temperature of the discharge permeate. The established model assesses the efficiency of the cycle utilizing the energy and exergy analysis to assist in the selection of process design and operating conditions that conserve resources and increase efficiency. The exiting warm permeate is found to be responsible for a significant volume of energy losses when released to the atmosphere (Ali & Orfi, 2018b). There is a tremendous potential for the use of MD for groundwater or brackish water desalination. It offers a practical technical solutions for accessibility to clean water in rural and urban areas, with optimal energy consumptions.

## 4.2 Seawater

Despite the growing regional water shortage and the reality that about 50% of the global population resides within the 100 km perimeter from the coast, seawater has not been an unlimited water resource (Amy & Ghaffour, 2017). Seawater makes up 97% of the total earth's surface that its desalination utilizing the MD technology is seen as one of the options to produce freshwater (González & Amigo, 2017; Hubadillah & Othman, 2018). A detailed study of DCMD output based on mass flow and heat capacity has been performed, taking into account the transport anomalies, structural membrane characteristics as well as the process variables, to provide the baseline for the optimization of methods and materials. The exergy and sensitivity analysis with economic evaluation determine the viability of the DCMD approach. An improvement in the temperature gradient is found to result in a rise in the transmembrane flux as well as thermal efficiency. The improved DCMD output is achieved by utilizing polymeric membranes with high porosity, having the flux and thermal efficiency reduced by 26 and 50%, respectively, and the thermal conductivity increases between 0.1 and 0.5 W/m K. The optimum width level of around 0.7 mm could operate at a lower temperature level (less than 5 °C) (Al-Obaidani & Curcio, 2008). The experimental DCMD setup, as shown in Fig. 9, utilizes commercially available hydrophobic microporous membranes. The effects of the operating parameters on the permeation flux are investigated in a plate and frame assembly for the seawater desalination. The PTFE membrane exhibits 99.9% salt refusal at flow rate of 800 ml/min and the feed at 80 °C (Shirazi & Kargari, 2012). A robust hypothetical framework has been developed to estimate the flux of a hydrophobic (flat sheet) hybrid membrane, encompassing the active PTFE film and the scrim-backed PP support film, in the DCMD. The humic compounds interact with the organic matter and the biofouling throughout the processing of seawater by the DCMD. The humic compounds and the low molecular weight (LMW) organic matter are the primary biological matter of the seawater. The humic compounds are thermally disintegrated into the LMW-humic organics in the DCMD. The disintegration is much improved with the presence of NaCl, whereas the inorganic scalant (CaSO<sub>4</sub>) lowers the dissociation of the humic compounds due to the strong binding. The assimilable organic carbon (AOC) trial is employed in this analysis to quantify the biological fouling potential throughout the MD operation. The AOC accumulation is clearly associated with the biofouling development in water and membrane. The production of AOC decreases as the production of LMW-humic organics improves, suggesting the potential of reducing biofouling in the membrane and feed (Naidu & Jeong, 2015). Eco-friendly silica-templated ceramic hollow fiber membranes obtained from the rice husk ash waste have been developed utilizing sintering and phase inversion techniques. The DCMD graft membrane output is operated for 200 h utilizing synthetic seawater at different amounts of NaCl at the specific feed temperatures. Furthermore, the permeate flux is reduced with the increased feed quantity, feed temperature, producing a higher water flow rate of 38.2 kg/m<sup>2</sup> h, and a salt discharge of almost 99.9% (Hubadillah & Othman, 2018).

A computer system has been established to predict the output of the DCMD integrated solar thermal device for seawater desalination utilizing historical weather conditions. The findings suggest the significance of simulating the DCMD cycle in combination with the power basis. The water flow velocity in the feed and distillation networks have improved the water stream and thermal capacity of the DCMD unit. If the DCMD section is joined to the solar thermal reservoir, increase in the water flow rate could reduce both the system water flow and thermal performance. The output of distillates relies on the supply of solar energy throughout the day. A smaller scheme of a 7.2 m<sup>2</sup> of DCMD spiral-wound panel and a 22.6 m<sup>2</sup> flat sun thermal accumulator generate more than 140 kg/day of distillate in real weather conditions. This leads to an average distillate output rate of or  $6.3 \text{ kg/m}^2$  of the sun thermal capacitor or 19.7 kg/m<sup>2</sup> of the membrane (Duong & Xia, 2017). A theoretical model for DCMD has been built to forecast the transmembrane flux of hydrophobic flat hybrid membrane, having PTFE dynamic layer and PP scrim-back supportive layer. The model involves momentum, energy, species and mass balances for permeate and retentate flows, in combination with the mass transfer of the water vapor via hybrid membrane and the heat transfer across the membrane and the boundary layers, to the surface of the membrane. The surface porosity is the key parameter affecting the permeate flux more than the performance ratio (Lee et al., 2015). The design of real industrial-scale application for seawater Fig. 9 Illustration of direct contact membrane distillation for seawater desalination. (Modified from Al-Obaidani and Curcio (2008))



desalination by DCMD could be greatly assisted by the application of simulation. The open-source simulators (hollow fiber membranes and flat sheet membranes) built on the MATLAB GUI framework could complement the DCMD scale-up module. A combined "tanks-in-series" and "black box" statistical method established is to generate reliable simulation and to develop profiling of all the main variables in relation to the membrane length. Based on the pilot-scale test findings in one system as model inputs, the simulation built could predict the DCMD system output in the co-current and counter-current operations, at the larger scale. Such predictions have been demonstrated to have a high predictability in both the laboratory- and the large-scale setup. The simulators could identify the development requirements to guide the correct scaling up framework for the DCMD phase. The main technical requirements for the construction of the industrial modules are defined and evaluated to provide general and realistic guidelines for appropriate scale-up systems to ensure maximum freshwater efficiency using the DCMD technique. The built simulators are open-source, accessible to build optimal DCMD system scale-up techniques with the selected membranes (Dong & Kim, 2017).

A system of submerged membrane distillation (SMD) for processing "Red Sea" water utilizing hollow fiber membranes is developed similar to the conventional DCMD system. In the SMD method, the hollow fiber membranes are incorporated at each side to ensure that the simple open membrane package assembly is immersed in a coolant reservoir, fitted with a stirrer. The heated feed stream is transported through the lumen portion of the membrane at the feed inlet. Complete mixing and contact with the coolant allows more uniform concentration and temperature. The use of co-current and counter-current flow for feed-coolant streams is evaluated, and the comparison is made with the conventional DCMD. In the SMD cycle, the 10.2 kg/m<sup>2</sup>/h water flux is accomplished by keeping the temperature of the inlet feed at 80 °C and the coolant at 20 °C. At the same conditions, a feed of 10.1 and 11.6 kg/m<sup>2</sup>/h of water vapor flux, during the co-current and counter-current streams, are observed with the conventional DCMD. The water generation in the SMD cycle is similar to the DCMD cycle, although the feed-cooling streams are in co-current mode (Francis & Ghaffour, 2015). An experimental vacuum membrane distillation (VMD) system, utilizing PP hollow fiber, was modeled and mounted on a reservoir as illustrated in Fig. 10. The seawater inlet is warmed by the excess heat produced by the vessel generator, achieving the desalination rate of 99.99% and 5.4 kg m<sup>-2</sup> h<sup>-1</sup> flux at 55 °C (Xu et al., 2006). A VMD has been utilized to process brines from reverse osmosis, the simulations are carried out for optimization of the conditions, and validation is performed in the small-scale unit, utilizing real RO saline to produce 300 g/L of salt content. The influencing factors are a fully permeable membrane, higher feed temperature, a turbulent fluid system and low permeate pressure, to allow large permeate fluxes even at a highly concentrated salt content. Calcium sulfate and calcium carbonate immediately precipitate out owing to their poor solubility, and crystals are deposited on the surfaces of the membrane, but not covering all the pores. The crystals can be efficiently removed by cleaning the membrane. A universal retention rate of 89% could be accomplished by combining the VMD and RO (Mericq & Laborie, 2010). Thermoplastic effects have been demonstrated





through photothermal excitations of silver nanoparticles (AgNPs) integrated into the microporous PVDF membranes. The hot feed on the membrane surface is subjected to the UV irradiation to achieve high desalination efficiency using the VMD. The flat sheet microporous membranes embedding the AgNPs are fabricated by utilizing a non-solvent-induced phase inversion technique. The nanoscale thermal hotspots of the AgNPs-embedded PVDF membrane, under the UV enhance the surface temperature. irradiation. The laboratory-scale validation suggests an 11-fold increase in the transmembrane flux at the highest NP composition (Politano & Argurio, 2017). A groundbreaking method based on "vacuum multi-effect membrane distillation modules" (V-MEMD) has been developed for pilot-scale desalination of seawater. The integrated system uses the seawater feed current in the condenser, where the heated feed increases the thermal performance achieving the production ratio for seawater of 3.2. The total distillate shows  $8.5 \text{ L} \text{ h}^{-1} \text{ m}^{-2}$  flux at 75 °C for hot feed temperature and 150  $L h^{-1}$  for the feed flow rate. However, the scaling lessens the distillate supply by 50%. The scaling can be effectively eliminated by acid washing, and the output is restored, without having to resort to an anti-scalant as a pre-treatment (Andrés-Mañas & Ruiz-Aguirre, 2018).

The VMD template has been designed both on the membrane scale (membrane pore system) and on the module scale (feed bulk longitudinal system) and then connected to a comprehensive solar energy system. Impacts of different solar and VMD variables on the regular water generation, energy usage and "gained output ratio (GOR)" are evaluated via a sequence of simulations. The findings suggest that the reduced condensation heat recovery with smaller running temperatures is optimal to attain the regular water generation

of 8 kg  $m^{-2}$  of the reservoir and may reach for the drinking for residential area over a 12-h period. The GOR of above 0.7 is similar to the single-stage MD systems powered by indirect heat supply (Ma & Ahmadi, 2018). The viability of an efficient hybrid device comprising freezing desalination and VMD, powered by surplus energy and sunlight energy for seawater desalination, has been reported. Freezing stage is the primary phase in the cycle where fresh ice is collected via freezing the seawater. To guarantee the ice purity, the fresh ice is melted by portion of the VMD distillate. The PVDF, tri-bore, hollow fiber films are used in the VMD. The resulting membrane possessed an average  $51.7 \text{ kg/m}^2$ (permeation flux) at 70 °C (feeding temperature). With every one kilogram of the seawater supplied to the system every hour, the gross water retention is 0.74 kg/h. The boiler function may be run either by 13.8  $m^2$  solar panels or by 3.10 kg (regasification) of LNG, if the device is completely integrated (Chang & Zuo, 2019). The solar powered VMD energy efficiency has been examined on four standard days during four different seasons in a year, including equinoxes and solstices, using commonly used energy metrics like specific energy usage, production ratio and heat retention factor. The energy efficiency is closely related to the solar radiation, and provided that the VMD is run on solar, it is able to compete with the industrial techniques based on RO, MSF and MED (Miladi & Frikha, 2019). A new solar seawater distillation module has been designed to improve its thermal efficiency by utilizing seawater to condense the steam and preheat the feed. Trials are performed under various environmental parameters to determine the function of thermal storage module in reducing the impact of disruptions under sun irradiation. The thermal energy can be supplied to the distillation system at a constant temperature,

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even with varied sun irradiation. A simulation using a quasi-dynamic system is carried out to determine the profile of the distillate output where the running period is throughout the typical year, taking into account the various heat settings at the membrane unit inlet (60, 70 and 80 °C). The quantity of the distilled water produced yearly, from the simulated data, is in the range of 41.7–70.5 m<sup>3</sup>, based on the set-point. The membrane distilling system provides water almost evenly throughout the year, with a typical flux of  $5.5 \pm 1 \text{ L h}^{-1} \text{ m}^{-2}$  at the highest set-point (Andrés-Mañas & Roca, 2020).

The AGMD method aimed at seawater desalination has been optimized to achieve high distillate output with lower electrical and thermal power input. The pilot study shows the major effect of water flow stages and inlet temperature of the evaporator on the system efficiency. Improvement in the distillate output and the energy performance can be accomplished by increasing the evaporator air intake temperature. There exists a trade-off between the power usage and the distillate output rate, as the amount of water input differs. The increase in the rate of water input has contributed to a change in the rate of distillate output and also in the electrical energy usage. For low energy consumption in the experimental AGMD, the feed salinity can affect the plant output, while the influence of temperature and saturation variation are minimal. The seawater desalination based on the AGMD could be accomplished at different thermal and electric power usage, respectively, at 90 and 0.13 kW h  $m^{-3}$ . This suggests the economic feasibility of the AGMD for smaller units, as well as for off-grid seawater desalination, where solar thermal or minimum-grade power sources are freely accessible (Duong & Cooper, 2016). The two "multi-channel spiral-wound air-gap membrane distillation" systems have been evaluated for seawater desalination on a commercial scale. The key difference between the systems is the membrane surface area of 24 and 7.2  $m^2$ , with the channel lengths of 5 and 1.5 m. The effects of inlet evaporator temperature, inlet condenser heat and feed velocity are determined. The profitability and energy efficiency are found to be mainly linked to the heat retention within the system (Ruiz-Aguirre & Andrés-Mañas, 2018). A techno-economic evaluation of the water gap MD (WGMD) and AGMD for seawater desalination has also been explored. The simplified cost of water (SCOW) approach is used to measure the expenditure expenses, fixed and variable expenses, amortization effects and the market impact over the duration of implementation. The trial results based on a small-scale MD desalination system are also employed. The most realistic scenario for the WGMD is with the capability of  $1000 \text{ m}^3$ per day utilizing waste and solar radiation, where the prices of the MD are close to the traditional desalination technology at the same rate (Amaya-Vías & López-Ramírez, 2019). Two separate industrial PVDF membranes with specific pore sizes (0.45 and 0.22  $\mu$ m) are tested and evaluated for various operating parameters in a single-stage AGMD. The trial findings suggest that the wider pore membrane exhibits improved mass transfer efficiency which contributes toward a small decrease in water generation quality. Nevertheless, the AGMD method would yield water with a high salt discharge of greater than 99% and a gross permeate flux of 9.06 kg m<sup>-2</sup> h<sup>-1</sup> at 77 °C (feed temperature) (El-Mokhtar & Boubakri, 2019).

Although the Seawater Reverse Osmosis (SWRO) currently dominates the demand in desalination, significant technical problems persist and have not been satisfactorily addressed. In particular, the comparatively small water retention factor (of around 50%) achieved, which has resulted in massive volumes of brine released. Furthermore, the energy usage of 3-5 kWh/m<sup>3</sup> is far higher than the required thermodynamic level of around 1 kWh/m<sup>3</sup>. The efficiency of novel devices incorporating SWRO, reverse electro-dialysis (RED) and MD for combined processing of energy and water has been examined. The recovery of the hypersaline waste brine via salinity gradient power generation through RED and the attainment of higher retention factors suggest the possibility of zero liquid release and less energy desalination, in real applications. The investigation is accompanied by the laboratory-scale experiments performed on the RED and MD under different operating conditions. The exergetic performance reaches 49% for the best situation at MD 60 °C feed temperature, MD saline amount of 5 M NaCl and RED power density of 2.2 W/m<sup>2</sup> per pair of the membrane. A decline of up to 23% in electricity usage and a 16.6% in real energy usage is accomplished by using the RED model. The efficiency of thermal power supply for the MD is important, while small-grade surplus energy or solar thermal sustainable sources may theoretically be accomplished. The proposed interconnected framework is compatible with the new circular economy model and the process intensification principle (Tufa & Noviello, 2019).

The nanofiber membranes comprising poly(vinylidene fluoride-co-hexafluoropropene) have been fabricated via electrospinning, followed by coating with TiO<sub>2</sub> nanoparticles for membrane renewal during actual seawater desalination with MD. Compared to industrial PVDF membrane, electrospinned-TiO<sub>2</sub> the membrane has improved anti-fouling and anti-wetting features that can be easily restored by plain physical water cleaning. Fouled membranes display renewed hydrophobicity after 30 min of plain water cleaning, with the contact angle restored from  $131.4 \pm 2.2^{\circ}$  to  $151.3 \pm 1.2^{\circ}$  (Guo & Deka, 2019). An easy and practical approach to enhance the desalination efficiency has been devised by integrating the 2D boron nitride nanosheets (BNNSs) into polyvinylidene fluoride-cohexafluoropropene (PVDF-co-HFP) to form the BNs-PH electrospun nanofiber membrane. The BNs-PH membrane displays robust water vapor influx (18 LMH) and excellent salt discharge (99.99%) after 28 h (the commercial PVDF: flat PH with wetting in 4 h and fast deterioration in 28 h). The method implemented is able to be extended to other desalination schemes to improve the different water treatment programs (Lee & Woo, 2020). A novel superhydrophobic electrospun PVDF membrane has also been fabricated and added to the MD-operated desalination technique. The casting solution configurations is designed to create an irregular surface of transparent microspheres. The membranes display a reasonable permeability at 152.4° contact angle, which is suitable for the MD-operated desalination. Following the real seawater desalination for 120 h, the developed membrane has exhibited more robust permeate flux as compared to the normal PVDF film. The desalinated water freshly produced from the membrane exhibits low conductivity attributable to the membrane fouling resistance, induced by the air gap between the film and the feed. The fabricated membrane offers a potential technique to enhance the short-term performance of the marine water-type MD desalination (Su & Zhang, 2020).

Photovoltaic (PV)-MD system has been reported capable of reliable production of clean water (greater than 1.64 kg  $m^{-2} h^{-1}$ ) from seawater while co-generating the power production (greater than 11%) under the sun. Its high output rate of clean water is accomplished by a mega-stage membrane distillation (MSMD) system with the solar cell power backing to recover the latent energy of the water vapor condensation at each distillation point. This hybrid design could substantially minimize the cost of capital expenditure by using the same land and the same mounting structure, thereby offering a possible opportunity to convert an electrical power station to a source of freshwater generation (Wang & Shi, 2019). Being the second generation separation technique, MD is described as a critical industrial method with exceptional performance for seawater desalination. The advent of nanotechnology technologies could further improve the MD performance especially by the modification of nanomaterials and the fabrication of advanced polymers (Alsebaeai & Ahmad, 2020). Membrane fouling and scaling are the key problems restricting the implementation of MD on a larger scale. Various MD designs, driven by geothermal, solar and waste energy, have been developed and tested to achieve efficient desalination of seawater, especially for hybrid projects requiring zero liquid drainage. Novel membranes, membrane assemblies and MD designs have been introduced in recent years to achieve better desalination of seawater. To ensure resilience and prolonged utilization of the MD, there is a need for life-cycle analyses of MD projects to measure their environmental impacts.

#### 4.3 Produced Water

Produced water, which is the wastewater produced by the oil and gas industry, is a mixture of a variety of organic and inorganic components. The physicochemical characteristics of the extracted water differ considerably based on the geographical position of the region, the geological composition and the types of hydrocarbons produced. The characteristics and quantity of the extracted water differ depending on the reservoir, and the content of the water derived from various sources differs by the orders of magnitude. The key components of the produced water include soluble mineral formations, dispersed and dissolved oily components, produced chemical compounds, dissolved gases and produced solids (which involve scale and corrosion products, waxes, formed solids, asphaltenes and bacteria) (Ali & Quist-Jensen, 2018). Proper handling and storage of the produced water is a big problem in the gas and oil sector. Membrane techniques are attractive due to the large separation capacities, high performance and low operating costs. The key factors in the selection of membrane operations are the lower energy demands, a broad range of workable separations, compact size, lightweight, lesser degradation concerns and lower scaling (Ali & Quist-Jensen, 2018). The membrane crystallization (MCr) and membrane distillation (MD) attract considerable attention for water processing owing to the wide range of concentrations and reduced fouling capacity (Ali & Quist-Jensen, 2018; Siyal et al, 2019). The DCMD has been investigated for hot brines and aqueous treatments below 100 °C. The DCMD provides additional benefit for feed solutions that need extra energy to chill the solution, as compared to the RO which could not exploit the heat present in the supply solution. The produced water collected from the steam-assisted gravity drainage (SAGD) is one of the models in which DCMD is a valuable method for desalination. The DCMD with porous flat layer PTFE film has been investigated in the region 80-130 °C for brines comprising 10,000 ppm NaCl. The efficiency of prepared membrane is also examined with saline feed with NaCl (3000 ppm), phenol (45 ppm), cresol (45 ppm) and naphthenic acid (10 ppm). There is no loss of salt in any conditions investigated. The maximum water vapor flow of 195 kg/m<sup>2</sup> h obtained is higher than that obtained from the seawater based on the RO cycle. The water produced by DCMD method can be employed for steam production in the SAGD cycle (Singh & Sirkar, 2012). The DCMD method utilizing polymeric membrane has been evaluated for the treatment of oil derived water. The permeates analyzed show that the MD mechanism is effective for the desalination of the processed water, with a total salt removal efficiency of above 99% and a total carbon

removal of above 90% (Macedonio & Ali, 2014). The efficiency of hydrophobic microfiltration membranes in a DCMD unit is investigated for the treatment of heavy salinity produced water (total dissolved solids of approximately 300,000 mg/L). PTFE and PP membranes produce the largest permeate flux at 5.6 Ll/m<sup>2</sup>/h/kPa (distillation coefficient). Examination of the membranes after prolonged experimentation with real processed water exhibits unevenly dispersed inorganic concentrations of substantial iron content. The findings suggest the prospect of membrane distillation to handle heavy salinity wastewater from natural gas production (Lokare & Tavakkoli, 2017).

The MD has been studied for the treatment of hydraulic fracking produced water (HFPW) having high salinity. The raw and electrocoagulation (EC) pretested HFPW models are evaluated with the DCMD under a range of operating conditions. Higher pollutant elimination throughout EC process leads to lower surface fouling and a greater flow reduction with the DCMD. The extended period of EC-DCMD tests has been performed desalinating the feed water equal to 265 g  $L^{-1}$ , producing 135 g  $L^{-1}$  total dissolved solids (TDS). Sustainable water flow with reduced fouling is recorded over an extended period of 434 h (Sardari & Fyfe, 2018). Four monomers, namely acrylic acid, hydroxyethylmethacrylate, 1-vinyl-3-hexylimidazole bromide and 1-vinyl-3-allimidazole bromide, are grafted on the interface of the PVDF membrane. The base and modified membranes are investigated in a DCMD system to treat actual produced water. The neat membranes and the membrane with 1-vinyl-3-allimidazole bromide are previously treated with electrocoagulation. These membranes are evaluated to desalinate artificial wastewater made by introducing large inorganic compounds found in the processed water into deionized water. For membrane fused the with 1-vinyl-3-allylimidazolium bromide, the maximum fluxes are produced. The stability and efficiency of the membrane can be enhanced by modifying the characteristics of the monomeric components in the polymer chains (Anari & Sengupta, 2019). A new Janus hybrid hollow fiber membrane in DCMD mode is employed for the desalination of real processed water containing 154,220 mg/L of TDS. The prepared hybrid (Janus-HFM) is characterized by a slim and hydrophobic PVDF and superhydrophobic silica nanoparticle (Si-NP) on the outer surface with high porosity, and the hydrophilic PVDF contains a polyethylene glycol (PEG) internal coating. In comparison with the clean PVDF hollow fiber membrane, the hybrid Janus-HFM demonstrates both enhanced water permeate and better energy performance, with 99.99% salt refusal (Zou & Gusnawan, 2020).

The AGMD has been used to handle produced water where the permeate fluxes, denial parameter and energy usage for three separate membranes, TF1000, TF450 and TF200 with pore dimensions of 1, 0.45 and 0.2  $\mu$ m, are S. Ahmad et al.

evaluated at various operating conditions. The flow is enhanced with the increase in the feeding temperature and the flow rate and reduced as the coolant temperature increases. The energy usage calculated at various pore sizes suggests that it is independent of the membrane pore sizes (Alkhudhiri & Darwish, 2013). Microporous membranes constructed from hydrophobic polymers like PVDF are commonly employed for MD. Nevertheless, the hydrophobic MD membranes are susceptible to the moistening small surface tension liquids, thus restricting their use in the treatment industrial wastewater including the shale gas produced water, with more complex composition. A simple fabrication method of omniphobic PVDF films that remove both oil and water has been developed. A surface positive charge is provided to the alkaline-preserved PVDF membrane through the functionalization of aminosilane that permits the permanent attachment of the negative charged silica nanoparticles (SiNPs) to the film via electrostatic interactions. The layer with attached SiNPs is later layered with fluoroalkylsilane to minimize the film surface variation. The covering of the SiNPs with ultralow external energy greatly improves the membrane's wetting resistance toward the lower surface tension fluids. The desalination efficiency of the changed membrane is tested in the DCMD with industrial wastewater comprising sodium dodecyl sulfate surfactant including the mineral fuel and the shale gas generated water. The prepared omniphobic membrane exhibits consistent MD efficiency, suggesting its capacity for the desalination of industrialized wastewater comprising numerous low-surface pollutants (Boo & Lee, 2016). The membrane distillation combined with crystallization (MDC) is suitable to process the "shale gas produced water" (SGPW) due to its resilience against high salinity and the intrinsic geothermal power. In combination with crystallization, the scalant loading is minimized adequately, and hence, the membrane wetting is successfully prevented, as shown in Fig. 11. The implementation of such MDC mechanism improves cumulative retention to 62.5% (Kim & Kwon, 2017). So as to determine the effectiveness of the MDC, the efficiency metrics including the rate of water recovery, solid production rate (SPR) and basic energy usage are evaluated utilizing the actual SGPW. The findings indicate that the water and minerals are efficiently recovered with an 84% retention rate and an average of 2.72 kg/m<sup>2</sup> each day at the optimum operational conditions. The scale function is established as a limitation factor for the reduction of the MDC ideal operating conditions. The MDC can sustainably and efficiently recover minerals and water using lower power usage from the SGPW by improving the operating conditions (Kim & Kim, 2018).

The viability of an optimized forward osmosis membrane distillation (FO-MD) method has been determined for the production of water generated from the shale gas extraction



**Fig. 11** Illustration of membrane distillation accompanied by crystallizer for shale gas produced water desalination. (Modified from Kim and Kwon (2017))

plants, with high salinity. In the developed hybrid method, the FO draws water from high-salinity stream, whereas the MD restores the diluted FO draw solution. The EC pre-treatment of the FO-MD system demonstrates good efficiency with limited fouling. The EC reduces gross organic content and gross suspended solids by up to 78% and 96%, respectively (Sardari & Fyfe, 2019). Analysis shows that the thermal energy of the MD activity adds much to the overall expenditure for the production of the processed water in the MD facility. The sensitivity analysis indicates that the amount of TDS for feed and radiant energy rates have substantial effects on the overall cost of production of processed water. The techno-economic assessment (TEA) is important to determine the fiscal viability of MD for water management generated relative to additional methods for water resources provided by the shale gas (Tavakkoli & Lokare, 2017). Despite extensive efforts to customize the membrane surface wettability to lessen the wetting and fouling in MD, the developed membranes are highly affected by the complexity of the actual industrialized wastewater. A thorough consideration should be given to the issues of wetting/fouling resistance, water quality, and recyclability, in the design and manufacturing of suitable films for MD application. Effective monitoring system should be put in place to determine the MD efficiency for continuous use and repeated application over prolonged period (Du & Zhang, 2018).

# 5 Recent Advances

MD is a viable alternative to the thermal-based membrane technology to provide pure freshwater from multiple water resources. Nevertheless, as a stand-alone system, the efficiency of the MD remains a stumbling-block to its wider application. The hybrid MD, by integrating MD with additional systems, could provide a realistic solution to improve efficiency and to accomplish a significant resource recovery. The performance and associated issues of the different MD configurations on resource recovery must be addressed. The recovery of useful salt/component from the polluted water bodies can be achieved, but the hybrid MD-crystallizer is restricted to the removal of large salts. The MD-adsorbent also shows capacity for selective recovery of useful elements that can reduce treatment costs. The combined membrane distillation forward osmosis (MDFO) and membrane distillation bioreactor (MDBR) are attractive for water recovery from the wastewater and for the recovery of resources and biogas that minimize the environmental pollution. The regeneration of water and power can be accomplished through dual membrane distillation pressure retarded osmosis (MD-PRO) and membrane distillation reverse electro-dialysis (MD-RED). The benefits of hybrid MD for product recovery in specialized fields have been well reported (Naidu & Tijing, 2020). A novel technique has been devised to build a stable and superhydrophobic membrane through electrospinning, accompanied by electrospraying, to improve the membrane anti-wetting effects induced by fouling and scaling. The durable PDMS-3 superhydrophobic membrane consists of a superhydrophobic surface formed by electrospraying a PVDF/PDMS/silica smoke blend mixture and electrospun nanofibrous PVDF reinforcement. A better MD flow of 28 kg m<sup>-2</sup> h<sup>-1</sup> and a salt discharge of more than 99.99% could be obtained during the DCMD process in 160 h continuous mode, at the feed and permeation concentrations of 3.5% (w/v) NaCl solution at 333 K and the purified water at 293 K. These findings emphasize the significance of the production of nonabrasive.



Triple-layer Nanocomposite Membrane Model

superhydrophobic membranes to treat wastewater composed of varied organic and inorganic material, in the MD (Liao & Zheng, 2020).

A two-tier superhydrophobic intertwined hybrid film built on a spatially ordered polypropylene (iPP) layer and electrospun PVDF nanofibrous coating has been prepared. The top layer has a disfigured nano/microstructured microsphere exterior with a strong super-hydrophobicity and additional anti-fouling properties owing to the synergistic impact of small surface free energy content and surface roughness. In assessment, for the simulated, the engineered superhydrophobic hybrid membrane demonstrates a reasonable permeate flux (53.9 kg/m<sup>2</sup> h) of high-salinity sunset vellow freshwater and a full discharge over 50 h of treatment at 40  $^\circ$ C, owing to its porous nanofibrous support and strong liquid repulsion (Deng & Liu, 2020). Hybridization of MD and FO has the ability to meet the increasing demand for clean drinking water. However, effective evaluation of systems operating parameters, improving the water transfer rates, with an acceptable drawing solution, is needed to achieve high permeate output. A study on the FO-MD method has been carried out to determine the drawn solutes, based on poly(diallyldimethylammonium) chloride, tetraethylammonium bromide and sodium chloride to represent the different FO and MD permeate fluxes. The fouling is found to be more prevalent in the FO, but the differences in the transfer rates of water are found in both systems. The analytical findings and the template are utilized to forecast the MD stream temperature and cross-flow velocity changes to attain the water equilibrium for each drawn solution. The optimization of the parameters has resulted in the output of the permeates enhanced, suggesting that the FO-MD method is excellent for water distillation when the efficiency is optimized (Zohrabian & Hankins, 2020).

Effective coating of industrial porous polytetrafluoroethylene membranes with poly(vinyl alcohol) (PVA) is accomplished by solvent dipping, accompanied by cross-linking. The fabricated membranes are tested for their success in DCMD configuration in improving water flow and salt removal. The flux is improved by a factor of 2.7 relative to the sterile membrane, suggesting that the addition of hydrophilic properties to some amount of hydrophobic membrane could greatly increase the efficiency of MD, without reducing the capacity for salt removal (Floros & Kouvelos, 2020). A new triple-layer nanohybrid membrane has been fabricated utilizing polyethersulfone (PES) and carbon nanotubes (CNTs) as the main base content and poly (vinylidene fluoride-cohexafluoropropylene) as the top and bottom surfaces of the layer, as shown in Fig. 12. The customized PES with CNTs is used as the main base content of the triple-layer membrane to achieve a highly porous membrane in a DCMD configuration, and the efficiency is evaluated by varying the concentrations of salt water, feed velocity and feed water temperatures. The triple-layer nanocomposite membrane is cost competitive and a viable alternative to other MD membranes (Elmarghany et al., 2020). A hydrophobic nanofibrous materials for VMD have been developed, composed of a hydrophobic PVDF nanofibrous surface and a hydrophobic PP nonwoven framework (NWF). The nanofibrous PVDF coating is processed straight on the exterior of the PP NWF substrate utilizing an electrospinning method. 1H, 1H, 2H, 2H-(FTES) perfluorooctyltriethoxysilane modified grapheme oxide (GO) nanosheets are inserted into the PVDF nanofiber film throughout the electrospinning phase to improve the VMD hydrophobic and permeation water flow. The permeation water distribution achieves an average of 36.4 kg m<sup>-2</sup> h<sup>-1</sup>, and the water velocity increases twofold with the salt discharge of over 99.9% at 50 °C, 3.5% sodium chloride aqueous medium and 31.3 kPa permeation pressure (Li & Shi, 2020). MD is an innovative membrane system that provides the opportunity to handle extremely salty water such as seawater, brine water and chemical wastewater and petroleum-gas field derived water. The normal hydrophobic membranes are facing rapid wetting and extreme fouling, particularly when the compounds having small surface tension are present in the salty water, which would affect the efficiency of the MD. The developments in materials engineering and nanomaterial research have contributed to the application of unique wetting effects on the membrane layers. The membranes with unique wettability could maintain a high tolerance to fouling and wetting of the membrane, with the trade-off between the permeability and selectivity of the membrane (Yao & Tijing, 2020).

# 6 Conclusion

Membrane distillation, a desalination process that uses hydrophobic membranes, has been proven effective for water desalination and for the elimination of toxins including heavy metals, radionuclides and organic matter from the brackish wastewater generated by factories. This system is based upon the pressure difference in the transmembrane, as a consequence of the change in temperature across the hydrophobic membrane. There are four specific designs of the membrane which are commonly used-the flat plate, hollow fiber, spiral-wound and tubular membranes, each with its own advantages and drawbacks. The MD is being primarily employed for the extraction of salt from seawater and brackish water. Although it is capable of handling different types of water, its potential can be expanded with the current technologies, including reverse osmosis, electro-dialysis, multi-effect distillation and multi-stage flush distillation. However, these are all high energy-consuming systems. The MD is useful to be incorporated with cost-effective power sources, for instance, the solar panels, geothermal and waste electricity. The facilities in sectors with large small-grade waste heat bases, such as the chemical plants and power stations, may therefore be good for MD facilities. The potential uses of the MD include the development of high water purity, the extraction of ionic, colloid or other non-volatile aqueous substances and the elimination of volatile organic components from wastewater. These are applicable for seawater and brackish groundwater desalination, environmental sanitation, conservation of energy, and food and medical sectors. The MD method could be conducted at lower operational pressure and temperatures as compared to the feed's boiling point, needs lesser vapor interphase changes and could withstand higher osmotic stress, with higher separation factor of the non-volatile solution. High fluxes and wetting are the major drawbacks of the MD. Superior permeable membranes and appropriate units with enhanced hydrodynamics would increase the permeate flow and the efficiency. The hydrophobic membrane and pore structure are important criteria to reduce membrane wetting. The MD has big potentials for large-scale market adoption, for the supply of clean water usage at the much reduced desalination unit costs.

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# Check for updates

# **Nuclear Desalination**

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# Abstract

Thermal purification is an energy-intensive method that meets the demand from traditional sources with fossil fuels. Existing work seeks to identify solutions to thermal desalination for fossil fuels. Nuclear technology is a feasible choice for electricity cogeneration and freshwater development owing to a large amount of usable power that has been retrieved. Heat is used for steam production and on-spot power storage in thermal and membrane waste treatment plants. It is necessary to use massive or SMR. This chapter discusses the numerous facets of nuclear purification, nuclear containers related to processes of desalination, and integrated desalination schemes coupled with nuclear containers. We address the health of nuclear desalination activities and general awareness, as well as the new economic analyses and evaluations of nuclear desalination projects on-site. Ten large facilities worldwide are run mainly as nuclear desalination plants. MSF, MED, and RO are the most

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significant desalination processes associated with nuclear SMRs. It was calculated that the rate of water construction through purification was between  $0.4/m^3$  and  $1.8 \$ m<sup>3</sup>, dependent on the container form and method of purification. Chapter findings suggest that proper preparation and control are required for the potential for marine impacts. In contrast with other co-locations, they often show detrimental maritime, atmospheric, and socio-economic consequences. Public safety problems are addressed, and perspectives are described. Higher environmental efficiency than other co-location alternatives is anticipated in nuclear desalination facilities. Environmental preparation and tracking operations are also much easier and less comprehensive and the key surveillance criteria are mentioned. Ultimately, it is proposed that nuclear desalination be introduced as a less polluting method.

## Keywords

Water Treatment • Desalination • Nuclear • Distillation • Energy

# 1 Introduction

The introduction of desalination technology in several areas of the world is becoming a realistic option for fulfilling the rising need for freshwater. Since the 1950s there has been a significant growth of the desalination industry. Capacity expansion in the Atlantic, Caribbean, and southern California has been reported (Jouhara & Khamis, 2013; Khamis & Kavvadias, 2013). Conventional waste management technology depends heavily on fossil-fuel electricity that eventually contributes to deforestation and global warming. Purification processes are in theory classified into big categories: thermal and non-thermal (membrane procedures) (Alsaman et al., 2016). Multi-stage flash, multi-effect

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distillation, vapor compression, reverse osmosis, forward osmosis, and electrodialysis are amongst the membrane purification treatments are the major thermal procedures. Which include MSF and RO are the two commonly employed systems. RO accounts for 63% of global power, while MSF accounts for 23% (Abdelkareem et al., 2018; Qasim et al., 2015; Walker et al., 2014).

Over recent decades there has been a substantial decline in the expense of desalinated water. However, several variables remain essential to the expense of desalinated water. Therefore, the equipment type, plant scale, position, plant ability, pretreatment specifications, feed water quality, and power cost are considered. Such considerations are considered. Studying the subsequent components: (1) energy expense (2) efficiency of traditional energy source (3) the environmental effects of fossil fuel and (4) volatility in fossil fuel price, the discovery in alternate sources of oil to desalination processes may seem to be a benefit. In a certain purification process, thermal energy is required as for Mass StageFlash and Mean Effect Distillation, whereas for example Reverse Osmosis and Forward Osmosisfilm technology require electricity. As a result, a big initiative is being made to investigate renewable energy options, such as wind, nuclear plants, and geothermal, in the form of desalination (Abdelkareem et al., 2018).

Nuclear desalination seems a feasible and promising alternative for low-cost desalination plants (Khamis, 2009). It is the product in a nuclear power plant of fresh/drinking water from coastal water. The technology produced can be used to produce electricity by thermal desalination processes and through a cogeneration system (Faibish et al., 2002; Khamis, 2013; Megahed, 2001). Since the 1960s, the IAEA has extensively researched the usage of nuclear electricity in desalination (Hammond, 1964; Starmer & Lowes, 1973). In the past, studies of the viability of using nuclear technology in desalination have been carried out by the IAEA effectively. The IAEA has released several studies (Ashoor et al., 2016; Megahed, 2001). Tests revealed some desirable features for nuclear desalination, including environmental security by reducing the production of greenhouse gases, potential replacement of existing sources of energy (fossil fuel), and economic viability in rural places where fossil fuel does not occur. The findings also presented this system as a common awareness and generated more technological trust in its application.

# 2 Nuclear Energy

In both developing and emerging countries, fossil fuel has been the main form of energy over the last 100 years, with a share of 81% (Edenhofer et al., 2014). Over the last 15 years, though, the electricity use trend has improved with

substantial expenditure over clean and alternative energy sources. The safety of energy sources has been the primary interest for many countries, especially oil importers (Khetarpal et al., 2016). It has contributed to many work projects to identify renewable energy sources that are inexpensive, reliable, and safe (Mushtaq et al., 2014). A significant focus has been given to nuclear power. In nuclear plant construction in the 70s and 1980s, the prospect of lower-cost nuclear fuel was the key motive, especially after the oil crisis in the 1970s. Nuclear power stations operate in several counties worldwide. E.g. in Japan, the United Kingdom, and Middle Eastern countries (Im et al., 2012). By 2016, there were 441 reactor types powered by [(gigawatt (electric))] in more than 30 provinces, a total of 382.9 GW(e). 68 reactors, 45 of which are under development in Asia only and have a total ability of 67.4 GW(e), are still under development. The latest estimates have shown that the global potential of nuclear electricity is going to be 511 GW(e) by 2030. This comes about by the need for expanded supplies of electricity, the extension of resources, the minimization of sustainability on non-renewables and oil import dependency (Sagir et al., 2014a, 2014b, 2016). Such considerations correlate with numerous environmental issues resulting from the overuse of fossil fuels as prime sources of resources, like warming, the greenhouse effects, and environmental damage (Adamantiades & Kessides, 2009). The IAEA's 2012 study suggested that global energy demand will rise by around a third by 2035 (Kavvadias & Khamis, 2014).

# **3 Nuclear Reactors**

The nuclear fission cycle was used in 2016 for producing electricity from both of the running nuclear power plants. The break of the strong atomic nuclei that are broken apart to create light atomist nuclei releases immense energy through nuclear fusion. Such Atomic Nuclei are considered by mass quantities, atomic statistics, the sum of the released neutron, and the µ-rays related to prime nuclei excitation. The task of the nuclear reactor is to turn thermal energy resulting from it into fuel. Various variations of the nuclear plant are widely used worldwide. They are divided into generations based on their historical growth. The main types are the following: Pressurized Water Reactor, Boiling Water Reactor, Pressurized Heavy Water Reactor, Gas-Cooled Reactor, Advanced Gas-cooled Reactor, Light Water (cooled) Graphite (moderated) reactor. Fast Breeder Reactor. High-Temperature Gas-cooled Reactorand Liquid Metal Cooled Fast Reactor) (Alemberti et al., 2014). There are approximately 441 nuclear reactors around the world today. PWR reactors are approximately 68%, BWR reactors are approximately 20%, PHWR reactors are approximately 6% and the rest are GCR, LWGR, and FBR (Khamis, 2009).

## 4 Technologies for Nuclear Desalination

The desalination of seawater may be defined by environmental, hydraulic, chemical, and electrical energy sources (Alkaisi et al., 2017). This segment provides a summary of emerging desalination technology in operation, with an emphasis on energy sources. The following sections are illustrated according to the new literature accessible.

# 4.1 Membrane Technologies

Desalination dependent on a membrane is one of the most critical processes of freshwater processing. It is because of a variety of factors: performance, working easiness, and strong productivity (Amy et al., 2017; Kochkodan& Hilal, 2015). This is installed on a semi-permeable membrane that helps desalinated water to disperse (permeate) while leaving behind (retentate), under a guiding force (Wang et al., 2017a), a concentrated salt solution. Presently in usage, the major desalination technologies based on membranes are RO, FO, NF, ED, and ultrasound filtration. The primary source of energy is power, which is the cause of membrane desalination (Shahzad et al., 2018; Azam et al., 2014; Mushtaq et al., 2014; Talebian et al., 2015). The membrane is characterized as a thin, permeable film that enables water molecules to move through and prevents the transmission of certain large molecules, such as salt, bacteria, metal, and virus, which lead to biofouling. The membranes are typically made with polymeric materials. For example, acetate, cellulose, and nylon are part of the polymeric materials. Within this segment, the major membrane processes are discussed, comprising (reverse osmosis), ED (electrodialysis), and MD (membrane distillation) (Mushtaq, 2014; Sagir et al., 2014c).

## 4.1.1 Electrodialysis

The movement of ions by a semipermeable membrane, below the guiding power of potential differences, requires electrodialysis. ED was used in various areas around the world for brackish desalination (California Energy Commission, 2005; Mushtaq et al., 2015; Sagir et al., 2014d, 2014e; Ullah et al., 2015). Within the ED cell, alternative patterns are set up for cation and anion exchange membranes. Cation exchange membranes permit only cation movement, whereas anion exchange membranes require anions to move through. With a direct electric current (DC), the sodium ion is absorbed by heading through electrodes that are coated in the electrolyte. This mechanism is done to isolate the saltwater comprising salts. Several scientific studies on solar-powered ED were performed (Ayotamuno et al., 2006; Gonzalez et al., 2017; Herrero-Gonzalez et al., 2018; Luo et al., 2017; Mekhilef et al., 2012). There were positive outcomes in these trials. The risks were minimized.

#### 4.1.2 Reverse Osmosis (RO)

RO is an osmotic pressure-influencing mechanism that operates the semi-permeable membrane to isolate pollutants from feedwater. The osmotic pressure is normally strong (50-80 bar), and the water can be transited through a membrane unit. With much more than 45% of the overall desalination power, RO is classed as the most energy-effective method used in freshwater construction(). Recent experiments have shown that RO systems are appropriate for salty purification purposes as they can provide a range of water types; both drinking and irrigation water are fairly inexpensive (Kim et al., 2017; Malek et al., 1996). Depending on energy source the price of water derived from the membrane process is typically approximately 1 USD/m<sup>3</sup>. The recorded cost is, for example, from 1 to 5 USD/m<sup>3</sup> if the membrane method is solar aided (Al-Othman et al., 2018). Research exertions are underway to test the cycle combination of various renewable energy sources with RO (Eltawil et al., 2009). Photovoltaic panels analyze solar energy as a feasible way to power pumps and/or produce electricity (Sagir et al., 2018; Ei-Ghonemy, 2012). Wind energy may also be used as a supply of energy in conjunction with the RO-systems with PVT collectors. RO systems powered by PV and thermal solar are currently sold for several hundred cubic meters a day in several locations with a range of capacities. For a Solar Rankin Cycle (RC) driven RO device, Shalaby sent a general design recommendation. The RO desalination membranes with a thermal water pump have been tested in the thermodynamic process. Photovoltaics/diesel/battery/reverse osmosis hybrid energy system output was improved. hybrid energy system. Wind as a potential source of renewable energy for RO desalination systems was examined. The tests revealed that the quality of the hybrid renews can be popular with increased efficiency (Ullah et al., 2019a, 2019b).

## 4.1.3 Membrane Distillation

MD is a thermal mechanism in which a molecule of water migrates under a vapor pressure gap across the microporous hydrophobic membrane (Liu et al., 2011). Recent experiments have shown that MD has a range of benefits such as waste heat extraction and water quality efficiency (). Several literature experiments were performed on improving MD thermal performance (Sagir et al., 2020; Duong et al., 2017; Guillén-Burrieza et al., 2012; Mahmoudi et al., 2017; Sagir & Talebian, 2020; Wang & Chung, 2015). The usage of thermal sustainable sources including solar energy and geothermal resources was discussed in recent studies (Banat et al., 2002; Mericq et al., 2011). Banat et al. (2002) described solar now incorporated solar technological feasibility program for the generation of drinking water. In a simulation analysis for various conformations of a VMD device lunar panel and solar collector systems Mericq et al.

(2011) introduced. The findings revealed that the usage of the solar collector may be successful as long as the movement of water stays 142 L/m<sup>2</sup> h. Many types of research centered on geothermal energy extraction of drinking water. This suggested that the use of geothermal vapor compression would result in substantial water costs (around 59%).

#### 4.2 Thermal Technologies

Freshwater is created by a phase shift cycle during the thermal desalination, i.e. by evaporations and condensations to remove the salt from the water. These processes are thus categorized by an enormous amount of heat energy. In this sense, the traditional thermal purification technologies addressed are MED and MSF desalination (Koschikowski et al., 2009; Youssef et al., 2014).

#### 4.2.1 Multi-Stage Flash (MSF)

In the early 1950s, the multi-stage desalination of Flash (MSF) was introduced (Donkelaar et al., 2016). It is founded on the purification theory in the multi-stage chamber, which unexpectedly decreases the pressure at every succeeding stage. MSF is a cycle of energy intensity (Failed, 2004; Younos, 2005). Over recent decades, the company's usage has grown dramatically, with about 60 and 80% of worldwide purification and Middle East purification respectively (Failed, 2005). Throughout the Middle East, it has risen considerably. The high efficiency, well-proven equipment, ease of use, and low output loss of MSF are characteristic across the years of usage. MSF plant has historically been used primarily in the Middle East where fuel is available and RO encountered difficulties (Compain, 2012). Most of the profitable MSF facilities available are built with stages of 10-30, with a drop in temperature of 2 °C per stage (Buhre et al., 2005; Ei-Ghonemy, 2012). The traditional MSF system contains a saltwater heater that lets and reheats feed water, flickering stages that minimize strain, thus fast evaporation or flushing, vacuum ejector (vacuum), chemical attachment pump (for scaling and corrosion control) and feed displays (Ettouney et al., 2002). The latest research on MSF systems based on 2 factors: (1) the elimination of MSF system costs and (2) the introduction of renewables. It appears that the expense of water desalinated using MSF technologies has been greatly decreased. Statistics indicate that since 1960 the cost of purified water by MSF has dropped by a factor of 10 (Reddy & Ghaffour, 2007). In MSF schemes, sustainable forms of energy including solar collectors and geothermal resources were incorporated (Gude, 2018).

#### 4.2.2 Multiple-Effect Desalination (MED)

MED is one of the oldest purification techniques. The MED system comprises of preheaters, condensers and distillation units. The evaporation of seawater usually takes place on the top of a steam-heated tube bundle. The vapor is diluted within the tube and then vapor produced in increasing consequence is used. The vessel's pressure and temperature are substantially decreased (Al-Shammiri & Safar, 1999). MED plants typically act as an unparalleled method without recirculating significant amounts of savory brine throughout the farm, thus minimizing plumbing needs and the creation of scales. The bulk of the MED plants are commercially related thermal steam compressors identified as desalination from MED-TVC. The evaporation of the first effect is carried out in this method through the compression of a vapor component from a solar collector or standard boiler at the required temperature at the last effect. Many MED devices are combined with devices classified as (MED/MVC) mechanical vapor compression which are not commonly available in the industry (Wang et al., 2017b). Throughout the past couple of decades, the existence of the MED was minimal relative to the MSF because of numerous management challenges, such as balancing up and extraordinary investments/operating costs, but several studies have shown that the MED cycle may soon overtake the MSF cycle, owing to lower energy requirements. In the last 10 years, MED development has experienced a range of changes. Such changes involve a major rise in tube scaling efficiency to 22,700 m<sup>3</sup>/day through proper design and increased aluminum surface heat transfer (Aniceto et al., 2013; Fath et al., 2013; Mezher et al., 2011; Zhou et al., 2015).

## 5 Nuclear Desalination

The method of constructing freshwater using a nuclear container onsite is nuclear desalination (Schwarzenegger, 2005). Plant capability and water quality are amongst the many variables that may expressively influence the claim for energy in the cycle of purification. As already mentioned in this context, the energy needed for desalination power may be thermal or electrical. To order to power MSF, RO and MED electricity, natural options such as geothermal and solar energy may be utilized, although they are installed in with smaller plants (Im et al., 2012). Compared to other traditional and alternative technologies, nuclear technology offers greater energy densities. As fossil fuel continues to deplete, the population continues to rise and freshwater demand increase, the construction of nuclear reactors is currently a critical need for developed developing countries. Because of the wide-area covered by the facility, the construction of large power plants in developed countries will present greater health and economic danger. Yet the construction of smaller plantations, development of hydrogen, energy generation, and development of freshwater through local desalination will address some of the problems of emerging technologies (Sovacool & Ramana, 2015). Over the past two decades, the IAEA has regarded nuclear desalination as one of the most effective and viable solutions for the manufacturing and production of freshwater. Since the 1990s, IAEA has begun a variety of study activities with nine leaders of state (Gowin & Konishi, 1999). They aim to study, support, and improve nuclear desalination (Grechko et al., 1998). They are referred to as organized research projects (CRP). Further goals shall be to examine nuclear desalination effectiveness, performance, cost analysis, and health. The studies presented sufficient evidence for potential nuclear purification system and outlined the significant benefits for nuclear purification: (1) the capacity to harness usable heat in thermal procedures like MED and MSF,; (2) build a multi generation network that is environmentally friendly; and (3) minimizing the overall costs for pre-massive thermal processes Globally, nuclear purification system fell into two primary groups: nuclear purification with production of electricity or autonomous nuclear purification.

## 5.1 Nuclear Reactors Types for Desalination

The first worldwide nuclear purification plants were built in the 1970s in Kazakhstan and Japan (W. H. Organization, 2009). Before the 1970s, nuclear desalination was tested by study, and its feasibility and reliability in comparison to other traditional energy sources were demonstrated (Faibish & Konishi, 2003; Methnani & Faibish, 2002). Two kinds of nuclear containers are commonly used in purification, according to the coolant type: LWR and HWR. BWR and water pressurized reactors (PWR) are both listed in the LWR grouping. By comparison, the HWR type is composed of HPWRs. Certain types of reactors such as the LMFR and high-temperature HTGR reactors are usable (Stacey, 2018). Well-established technology makes the water-cooled reactors preferred. Throughout the paper, the most collective nuclear reactor joined with desalinating processes is the PWR, the PHWR, and the LMFR (Nisan & Dardour, 2007). The IAEA rated nuclear reactor in the three groups based on power production: small reactors with an electric production less than 300 MWe; newspapers, with a power output of between three and seven hundred MWe and broad reactors with a capacity greater than seventy hundred MWe (Ingersoll,

2009). Current nuclear reactor technology for electricity production is focused on 1100–1700 MWe reactors. Large-scale desalination reactors are technically feasible, but different considerations, such as health and reliability, must be assessed before service (Alonso et al., 2012).

# 5.2 Nuclear Reactors Coupled Desalination Processes

The goal is to generate power and use waste heat to manufacture steam, which is supplied through the MED equipment. The following procedures will be taken for the construction of nuclear desalination: (1) careful simulation of reactor desalination systems; (2) detailed safety assessments, and (3) scientific findings of the desalination cycle itself.

## 5.2.1 Nuclear Reactor Coupling with MSF and MED

In this case, the MSF theory is already clarified. Cogeneration network and energy storage will be developed for the on-site nuclear power plant. However, the excess heat is used to heat water in the bay. This design will boost the economics of the network and thereby rising costs. In this case, the concept of the method of desalination is clarified beforehand. The MED structure is a collection of adjoining rooms, surrounded on one side by heat sources, and on the opposite side by a heat sink. As steam flows from one impact to another, heat and sweater are exchanged. By the conclusion of the show, more sodium is gradually evaporated, and the salt may become more condensed. The nuclear power plant provides an energy cogeneration network. However, both MSF and MED are costlier than membrane technologies as RO and more energy-intensive. They are used as a consequence of their development with the best water content (Wu & Zhang, 2003).

## 5.2.2 Nuclear Reactor Coupling with RO

Today the Reverse Osmosis deemed as the most cost-effective desalination process. The strain is placed on one side of the membrane to relieve the osmotic resistance of sweaters and to allow them to bring fresh water into the membrane. RO may be used if adequately handled to handle wastewater, sea, brackish water, and oily water. Coupling RO to every power plant and producing the electricity needed to run RO units is feasible. Energy is used for the process of pumps and power plants. The nuclear reactor is used for steam production in which the power used to run the pumps is transferred via the expander (turbine). From the 126

review and the previous paper, it is apparent that the key processes of desalination combined with nuclear reactor for the processing of freshwater consist of (1) MSF, (2) MED and (3) RO. A definite nuclear container design dependent on the form of energy needed is necessary for the desalination phase.

## 5.2.3 Nuclear Desalination via Small Modular Reactors (SMRs)

According to the IAEA study, SMR expenditure in desalination is increasing significantly. The IAEA plans 96 projects across the world to take shape by 2030. SMR has many benefits including simpler, faster, and healthier projects, fewer building time, and thereby decreased costs. They are still in service globally at various places. The most significant forms are: (1) SMRs (integral or iPWRs) that are harder to water-cool. For example, the KLT-40s in Russia, Korea SMART, USA IRIS, and Argentina CAREMs, (2) the heavy-duty SMRs such as India's PHWR 220s, and (3) gas-cooled reactor high temperatures such as China's HTR-10, Japan's GTHTR 300s.

# 6 Commercialization of Nuclear Desalination

To order to test and refine nuclear reactors together with desalination, ten primary programs around the world have been launched. These ventures include INVAP, Argentina, CANDESAL, Canada, China, Egypt's NPPA, India's BARC, KAERI, Moroccan CNESTEN, Morocco's OPPE, JSC, OKBM, Malaysia's Russia's Strong, and Tunisian CNSTN. In comparison, the EURODESAL initiative in Southern Europe tests the technological, protection, and economic viability of MED-RO nuclear power with a reactor of 600 MWe of PWR (AP600) (Alessandroni et al., 2002; Nisan et al., 2003). Other countries include Vietnam, Albania, Chile, Croatia, DR Congo, Thailand, Peru, Thailand, Uganda, Uruguay, and Zambia (Budnitz et al., 2018). At present, the Commission also involves nuclear facilities in the area. For starters, for economic reasons, Vietnam currently had to cancel its plans. Such programs should usually be used in tandem with potential efforts to pair such processes of desalination with nuclear reactors. Several factors, including environmental health, must be taken into consideration to deter pollution from freshwater with hazardous contaminants and plant economics. Blockades between container and purification must be included in the process design. Throughout the Middle East, many nations, in the UAE and Saudi Arabia in particular are worried about nuclear desalination for their water needs. Desalination has become a major concern in these countries, and many studies have been undertaken to determine the viability of this A. Mukhtar et al.

alternative utilizing statistical methods (Darwish & Al-Najem, 2008). Such countries could be perfect options to proceed with nuclear desalination, according to the reports. A further range of nuclear power stations is being installed worldwide (Wolff et al., 2015).

## 7 Trends in Hybrid Nuclear Desalination

All Multi-Stage Flash and Multi-Effect Distillation are more energy-efficient than reverse osmosis (RO) which provide a higher volume of water. As such, a range of combination patterns for investigating a nuclear container with one or more such purification processes was published in the literature. To order for the production and reliability of the desalination cycle to increase, Wu et al. researched a mixture network linking the nuclear heating reactors NHR-200 PWR reactor to the MED and RO. Two methods have been evaluated: (1) pressurized water reactors PWR NHR-200, MED + RO low temperature, and (2) NHR-200, PWR with MED + MED + Vapor compression low temperature. The study decided that much of the power supplied by the NHR reactor will come from freshwater processing with an additional few megawatts at a lower cost than MED at MSF separate methods.

Many reports also discussed facets and analysis of the thermos-economics of nuclear purification hybrid systems such as nuclear-RO-MED and nuclear RO-MSF. The studies suggested that hybrid nuclear desalination is a practical possibility as a feasible option to reduce the expense and to achieve better water quality. The NDDP facility in Kalpakkam, India that uses MSF-RO PHWR, 6300 m<sup>3</sup>/day, the Shevchenko facility in Aktau, Kazakhstan, up to 145,000 m<sup>3</sup>/day and the LMFR, MED-MSF and Karachi Nuclear Power Project (KANUPP) RO-MED project are recognized examples of hybrid wastewater treatment facilities around the world. Near to nuclear reactors, MSF-RO hybrid, and MED-RO electric power stations. A scheme for traditional MED-RO hybrid operation.

The synthesis of RO-MSF was explored and tested in a nuclear power station. The findings revealed that the RO-MSF hybrid device gave the following benefits: (1) optimal production in the two processes; (2) decreased energy demand; (3) decreased hybrid process costs; (4) increased water quality, and (5) increased operational capacity. MED was researched and a theoretical model built in tandem with a thermal vapor compression method and the RO. The findings revealed that the MED-RO method is the right workout tool. The literature indicates that integrating two purification technologies with a nuclear power plant has the greatest benefits over two methods, resulting in improved water quality and capacity and cost reductions.

# 8 Environmental Impacts of Nuclear Desalination

It does not exclude its ability to inflict significant adverse impacts, as a nuclear purification plant is a co-situated facility. Biological, technological, political, and economic influences can influence the nature and extent of these impacts. The following parts describe the possible effects of the reference plant.

## 8.1 Coastal Impacts

Nuclear desalination projects have environmental effects by building, land usage, and vibration as well as visual disabilities, much like any other coastal manufacturing scheme. However, all such effects are smaller or fewer than other similar co-located plants concerning electricity and desalination.

Construction: The design or extension to the current nuclear plant of a desalination plant involves a construction project characteristic of most coastal industrial production. This will include the use of large grading equipment, crop digging, drilling, de-watering, material distribution, sand and gravel filling, pipeline building, electricity, and road network development. In contrast to other desalination methods, the scale of the building site, and the decreased particular usage of materials for the power plant, the resultant results are of benefit to nuclear desalination. For example, wind facilities require 5–10 times as much steel and concrete per  $MW_e$  as a nuclear power. Nuclear desalination, though, has the potential to have a significant effect due to its extended development period, and a management strategy is thus required to offer this problem particular consideration (Ausubel, 2007; Damitz et al., 2006). This program involves site-specific operating controls, seasonal limits on other operations, stringent replenishment protocols, planting vegetation, systems on water management, usage of biodegradable materials, etc.

*Land Utilization*: The usage of land for nuclear desalination is also one of its advantages, particularly, water and energy needs are to be tackled at the same time or when geographical restrictions demand the construction of the plant in an already established environment. The co-location of nuclear power with desalination, owing to resource sharing, has a marginal dimension of land usage. The land used for piping and pumping plants must also be considered. The position of the desalination plant concerning the customers is highly affected by this. Usually, 200,000 m<sup>3</sup>/d desalination marine plants, the total area needed is approximately 0.4 km<sup>2</sup>. In our comparison case, for the auxiliary desalination systems, the average 40 TJ heat is an extra 10%, which is 44 TJ. The 3 Km<sup>2</sup> of total region for the comparison

atomic purification plant is less than coal plants in WEC compared with the area needed for wind and desalination plant supplies (Khamis, 2009; Newmark et al., 2010; Trieb & Elnokraschy, 2007).

*Noise*: The referenced facility is supposed to be moderate because the noise level of the distilling plants is smaller than that of the RO, and OTC systems are higher than that of the cooling towers. Thermal power stations, like nuclear power plants, will also provide a source of the steam ejector and turbine noise. As reported, the OTC system has a benefit over low noise chilling towers. Besides, in contrast to cooling once carried out, the California Energy Commission has identified noise as a "significant drawback" for Air Cooled Condenser systems. This is worth remembering that windmills have had a disruptive noise impact relative to alternative forms of electricity. Nevertheless, if the activity of the nuclear desalination plant results in noise that disturbs the environment and the residential areas, appropriate acoustic controls will adequately reduce the noise level (Ausubel, 2007; Plan, 2019; Schwarzenegger, 2005).

Visual Effect: In the event of a renovation of a nuclear plant with a purification system, visual effects would not be anticipated. Newer nuclear purification plants will, however, is placed in coastal areas which are also deemed a strong landmark. Mitigation steps refer, as affects every other coastal growth, to primarily use of the topography of sites. Of note, although the environmental effect is limited owing to the minimal land use criteria for nuclear purification, the development in a populated environment means a lower visual impact. In this situation, refrigeration systems are perhaps the better choice. In contrast, energy effective RO desalination will be the most appropriate option in command/purification facility with non-disposable resources and does not generate a by-product of waste oil. The scenario, though having a main part in the manufacture and performance of renovated plants and hence their scale, will entail covering an equally wide area with 80  $m^2$  with a wind turbine or 1.2 Km<sup>2</sup> with solar panel, according to conventional WEC numerical data for land use only by power sources for water generation, without the need for additional power required for other purposes (Newmark et al., 2010).

#### 8.2 Marine Impacts

In addition to being a radioactive waste site and waste drain, seas are also abundant in biodiversity. The particular ecological influences that on the aquatic system, particularly with focus dumping, have been devoted to coastal waters, host-sensitive marine environments, and not unexpectedly to substantial sections of science work on marine desalination. Nonetheless, recent experiments have shifted the emphasis by proposing a greater effect on the aquatic climate, even when it comes to autonomous desalination, by preparation and impinging of animals, rather than salt discharges. Many of the detrimental impacts of the co-locations of fish embryos, larvae, and plankton are triggered by large trappings. The impingement of larger (adult) species, believed to lead to these adversative effects, may also trigger container shut-down by hindering consumptions for nuclear desalination (California Coastal Commission, 2004).

It is necessary to remember that several variables related to the diversity of aquatic animals, and their associations with existing information on the subject are challenging to determine. Many fish species with a population loss in southern California have already been identified as the most entangled larvae, 6, and are often known to have a major environmental effect intake system. We may take the example of Diablo Canyon NPP from the period when the detrimental environmental consequences of any power generating project were generally unknown and ignored to place the marine impacts in context (McLean, 2003). The 2.2  $GW_e$  plant is in seashore with a 9.5 million m<sup>3</sup>/d coastal irrigation network from OTC. Studies have calculated that the aforementioned training/impingement intensity is, over nearly two decades, 10-30% for five chosen nearshore fish embryos, with the subsequent drastic effects on the local coastal climate. The study of the effects of intakes of the Diablo Canyon by the energy commission in California reveals that the depletion of larvae through instruction is  $1.2-2.4 \text{ km}^2$  of the coral environment (Davis et al., 2020).

Some of the factors for this tremendous impact are the enhanced exact need for refrigerating water in nuclear power plants, apart from the place of intake selected in the field of strong biological activity. Such criteria are primarily attributed to the decreased usable waste oil, which is smaller than that of fossil fuels (5–10%), because of the thermal capacity of the nuclear plants. Understanding that water withdrawal volume and pace are two main marine effects for the nonstop consumption used in once finished chilling, advanced training, and intrusion levels are required. Nuclear OTCs will have specialized monitoring equipment and intake sites in the most bioactive aquatic areas to prevent significant negative effects on the coastal ecosystem (Commission, 2005).

We may measure the potential of  $4,800,000 \text{ m}^3/\text{d}$  for the reference facility, an average conservative 200 m<sup>3</sup> per MWh. The further intake of 500,000 m<sup>3</sup>/d of seawater will consider the specifications of the desalination plant based on the real average cycle regeneration proportion of 40% for thermal purification, instead of the current "apparent" ratio of 20% that reflects cooling water, too. As the reactor has the function of the heater it is not necessary to refill the desalination water. The intake impacts of OTC systems would rely on their position, water intake pace, screening

technologies, etc., using a total withdrawal rate of  $5,300,000 \text{ m}^3/\text{d}$ . But, compared with other (fossil fuels) choices for co-location, they still have a proportionate capacity (Schwarzenegger, 2005).

Concerning aquatic impacts from the disposal, the nuclear purification plant connection has a smaller influence than stand-alone installations. That can be told. The profit, based upon the average intake estimates, is not evident relative to the other colocation alternatives, while higher intake of cooling water mitigates the harmful effects of the brine. Our comparison desalination marine of approximately 300,000 m<sup>3</sup>/day will be roughly 58,000 ppm TDS, considering the average value of maritime water for TDS (Total Dissolved Solids). Water with 36.000 ppm TDS will be mixed with the residual 4.800.000 m<sup>3</sup>/d of chilling water before being discharged (N. R. Council, 2008).

The impacts of insufficient oxygen and other chemical elements, e.g. chlorine or acids, in saltwater are generally mitigated as well. In the case of the example, the concentration of chemicals should be lowered approximately 15 times before publication, although additional procedures would possibly be required to decrease concentration again to No Effect Concentration (NEC). Dilution might not be the remedy for corrosion goods provided by the MED plant because the load is more critical than the concentration. Corrosion materials including copper and iron will bio-accumulate and inflict serious adverse effects on higher trophic rates. Specific steps can of course be implemented in this situation, although they are not unique to nuclear purification. However, the elevated temperature of the OTC discharge is typically also a problem for power plants. Nevertheless, in the particular case of the co-located facility, the desalination system is used as a heat sink which greatly decreases the effect of OTC discharge. The regular intermediary loop, dividing the nuclear loop and the purification loop, is a particular function in the comparison case. This is a protection prerequisite, but still an unwelcome thermal shield. The figures suggest that about 23% of excess heat for desalination should be used rather than distributed in the atmosphere for lower power reactors. If we should conclude that in the comparison case there will be at least as much surplus heat available, the MED plant should have more than enough resources for this phase(W. H. Organization, 2009).

The output of 200.000 m<sup>3</sup>/d would result in 40 TJ/d being extracted from the heat spread in the aquatic climate, presuming that the energy concentration is 200 MJ/m<sup>3</sup> for the MED cycle, which confers to the U.S. National Academy of Science, is a low-cost element. The requisite temperature range can be provided in the nuclear system by the temperature of the steam. Besides, an NPP of 1 GWe mounted, which is equivalent to 175 TJ per day, can spread 2 GW<sub>t</sub> per hour. Not all of the heat stops in coastal waters but in this situation, we should assume that authorizing the temperature rise in OTC discharge to be measured at a modest 10 °C. Compared to 100% of the waste heat generated by the OTC, the discharge of the nuclear desalination plant would raise the temperature by  $8 \in C$  before disposal, thus growing the impact region. Based on a basic fluid mixing method, it is feasible to predict that in impacted areas with temperatures over 1 °C there is a decrease of around 20%, without an ecological value.

Similar standardized findings may be obtained in other desalination solutions. There are, however, a substantial benefit of a standalone nuclear power plant when attached to a desalination system: the following steps to lower the OTC's discharge temperature from 8 to C would be cheaper and easier compared with the standalone nuclear plant in case of a nuclear desalination facility. There must be no disregard for the rise of the temperature difference among the real discharge and the discharge temperature boundary: a colder seawater intake may result in a colder discharge, with a decrease in power and water production due to thermal discharge limits. In the cycle of evaluating radioactive waste with water, scaling up the expenses by out-put, or determining cooling needs, this 40 TJ of heat used in purification often improve the capacity of the NPP by 33-50% approximately.

#### 8.3 Atmospheric Impacts

Compared to every other co-location method, nuclear purification has a lower effect on the atmosphere in that it has indirect negative effects of purification on the atmosphere, which is induced by the process power supply. As a direct consequence of the desalination operations, air pollution comprises mainly of deaeration oxygen and nitrogen releases with minimal environmental effects. While the energy intensity of desalination has decreased considerably in the last decades, the thermodynamic minimum value of energy is reaching which does nothing to further the effect on the environment. Many energy solutions are required. Compared with other energy sources, the ambient consequences of nuclear power are well known and are deemed lower, providing a mitigation option for one of the most important impediments to desalinization-its air influence.

If we consider electricity waste heat as an energy supply that does not have any external effects on the atmosphere, the releases of 200.000 m<sup>3</sup>/d of MED co-situated desalinated water per m<sup>3</sup> are only attributed to the auxiliary network that uses 4 TJ every day. We should then measure that, per m<sup>3</sup> of CO<sub>2</sub> for carbon, or in general, 480 tons of CO<sub>2</sub>eq each day would add 2.5 kg of CO<sub>2</sub> to the atmosphere. One m<sup>3</sup>/d of purified water adds around 5 kg of CO<sub>2</sub> to the environment for coal in the co-location of MED desalination, which corresponds to 1000 tons of CO<sub>2</sub>eQ each day. This CO<sub>2</sub> production per day is equal to 225.000 new traveler cars (150 g/km) driving an average of 30 km/d per day. Because of the 4 TJ/d desired for RO purification, the windmills, which are free from usable waste heat and RO, may generate  $CO_2eq$  emissions of 0.08 kg per m<sup>3</sup>/d, with a 16 tons of total  $CO_2eq$  per day. This may also be a natural option. In this particular situation, it should be remembered that throughout the activity of the facility, the wind is substantially varied and needs backup power for continuous water source (O. A. Plan, 2019).

As this backup power typically comes from a source of fossil fuels, the carbon footprint of purified water with renewable energy may be higher than the conventional estimates seen above. Considering the 0.7 wind energy supply factor, a quick estimate reveals 1 m<sup>3</sup> of purified water to have a CO<sub>2</sub>-equivalent carbon footprint of from 0.8-1.6 kg, dependent on whether the back-up electricity comes from a nuclear or gas power plant. This is the case for the desalination project Perth RO, which utilizes wind power to cover for pollution from other electricity sources the facility has used. In comparison, emissions of approx. 0.035 kg or 7 tons of  $CO_2$ , which may be due to the desalination ability, could be approx. for the comparative nuclear purification plant per m<sup>3</sup> per day. That is below 5% of the second finest alternative: the wind-driven, gas-based RO purification. In comparison, nuclear air emissions from NPPs are 100 times smaller than in a similar-size (1000 MW) coal power plant. A nuclear purification plant may thus be deemed environmentally sustainable from the perspective of air quality and an acceptable cause of electricity for desalination, thus allowing ambient emissions to be mitigated. The American National Academy of Science also advocates the usage for desalination of nuclear and alternative forms of electricity (Failed, 1993).

#### 8.4 Socio-Economic Impacts

The socio-economic effects have a significant impact on certain environmental consequences. The assessment and tracking efforts will consider them. The value of nuclear desalination as an economic and social aid is determined by many factors. The growth opportunity attributable to the supply of resources and water will be the most significant. This will not only alter the habits of customer behavior but can allow individuals, wealth, and money dramatically redistribute. Water and energy supply may have significant, restricted, or enhanced impacts on local growth. At the emergence and growth of Aktau (Kazakhstan), where nuclear purification was used to maintain the increasing town and industry of the region rich with mineral, the ability of nuclear desalination for production is better illustrated.

Yet the spatial delivery of water is constrained relative to electricity and therefore the effects that not be as common as coal. Therefore, the consequences of water-led growth may be more serious, and construction above and beyond local or national planning. In particular, it has been proposed by the California Coastal Commission that growth-inducing desalination impacts would possibly be "the most important indirect possible adverse effects". Growth because of water supply will fuel new residential and commercial construction, creating extra (unsustainably large) strains on local infrastructure. The supply of resources will, of course, boost this impact. We should continue with agriculture as a water-intensive operation by analyzing the socio-economic effect of the comparison facilities. FAO assumes 1000 m<sup>3</sup>/y of which 15% are normally irrigated for a total dietary intake of 2800 kcal/d per capita. This ensures that the comparison power will be used to deliver suitable irrigation for up to half a million citizens depending on the region. To sustain a town of 400.000 to 500.000 people and the normal water requirement, the Sidney-style nuclear desalination project also suffices (Rivera & Oamar, 2003).

This technology has tremendous consequences and must be complied with before the desalinated waters are rendered available; not just water conservation strategies but all the required facilities will be prepared for a growing population. Uncontrolled growth will influence the social equilibrium, local organization, and structure. Population increases outside municipal or national planning programs may contribute to environmental deterioration, when public resources and the facilities required for help (e.g. utilities) outgrow, add to social and economic disturbances. No matter what, it is not a propensity to reach natural limits with water-based growth that causes coastal habitat harm, irrespective of the desalination alternative. Desalination, though, has beneficial benefits, such as source flexibility, and decreases the impact of excessive water tension. This will boost drinking water safety, mitigate water scarcity, and do reduce environmental harm, as suggested in the purification study by the US National Academy of Sciences (Anastasov & Khamis, 2010).

Water accessible will reduce, reduce, and build wealth through tourism, manufacturing, and farming development and provide jobs. Water access will reduce tension over limited waters. For example, the case of Malta demonstrates that desalination and water-prevention initiatives may be helpful for the lucrative tourism industry. Bezdek and Wendling's nuclear power plant work has, however, concluded that the results have been overwhelmingly positive on local land, economy growth, revenue from taxation, public utilities, the creation of the city, jobs, and welfare, and schooling. This suggests that, if properly articulated within the context of a state, regional or local development strategy, nuclear desalination facilities of the reference potential would most likely have a broad and mostly beneficial social impact. Relative to certain electricity costs, the economic value of nuclear power is well established. As for the expense of desalinated water, a variety of factors depend on it. The IAEA DEEP algorithm analysis shows price levels from 0.5 to 0.96 US\$/m<sup>3</sup>, a favorable premium for desalination. The strong power supply efficiency of the NPP with an average of 83% in 2006 makes the unbroken electricity source for the comparison MED facility is an important factor in economic productivity. Half of the world's reactors are 86% available and above, 91% or more of the world's reactors are operational. Data are currently being modified on the fossil-fuel plant to provide a more detailed image of the current supply.

# 9 Public Health Implications for Nuclear Desalination

At first, it is well-known both in Aktau and Kalpakkam cases involving radioactivity were never identified as cross-contaminated in the liquid water. This is critical as the most significant public safety considerations and related threats in a nuclear desalination plant project are likely to affect the public's view. The key source of the issue is the toxic hydrogen isotope tritium, because it may pass through the liquid water through various physical barriers. This is no need to give public attention to the potential radiation pollution of the liquid water and the methods for avoiding it. This is critical for the execution of the project. A variety of health devices and working procedures are included in nuclear desalination. The most critical aspect is the above-described isolation loop, which is greater than the nuclear circuit and preferably smaller than the desalination loop. This further decreases the risk of tritium leakage through the heat exchanger shielding, as well as eliminating any potential outages of polluted coolant from the nuclear to the purification system. Tritium is constantly extracted from the nuclear coolant and even coolant tritium amounts per liter are held below WHO requirements (Muralev, 2003).

Finally, the product water is stored in retention chambers, in which it is necessary to calculate the degree of tritium content before discharge into the water stream, guaranteeing that the product water is healthy to use. The practice so far indicates that tritium in desalinated water is not induced by pollution or safety issues. In the desalination source of MAEC's 6 Bq/l Muralev records the highest tritium concentration. The Kalpakkam Reverse Osmosis plant has received desalination water with tritium content below the measurable level outside the factory. Likewise, there has been no evidence that the usage of desalination water for on-site use is a safety concern in the United States or Japan. Nuclear purification interactions with tritium indicate that the liquid water is secure for consumption by the community from the viewpoint of the legal guidelines for tritium in drinking water. We will use a central region heating loop in the Bohunice NPP, Slovakia to display waste heat being used from the nuclear system in non-electrical purposes. The loop is smaller than 1 Bq/l of tritium; the separation loop also has tritium rates far below the limits of the WHO (Muralev, 1998).

Besides, the radiological study carried out by the Aktau facility based on close-by soil and chilling pond sediment found that the plant has an insignificant relation to the radiological condition in the territories examined. The salt, therefore, does not present a possible danger to public safety as a hazard to radioactivity. The experience thus far has proven that the community will consider nuclear purification as an alternative for water supply. The factory in Kalpakkam, for example, has a growing appetite for desalinated water, in an area where water is scarce. The other related knowledge in nuclear purification is much more considerable: established in the desert, Aktau is the owner of the water generated by the nuclear desalination project and its population development. Nonetheless, the public's tolerance for nuclear desalination is not consistent with satisfactory assurance. Therefore, as the project is created, effective contact centered on data with the public must be provided (Paley, 1998).

## 10 Environmental Monitoring and Planning:

We will define the environmental mitigation targets based on the predictable environmental effects of nuclear desalination. Steps to minimize unmeasured effects ought to be part of the environmental strategy. Following the monitoring exercise, the function of an atomic water treatment plant should be verified inside the expected impact limits and the scheduled mitigate action. Environmental preparation will commence with the selection of an acceptable facility venue. Of course, it would be better to provide an environment that needs water and electricity. This will require seawater sources of consistent consistency (in terms of solid contented and temperature), low rates of contamination, and solid in the seawater and low potential for biofouling. For the reason of nuclear purification, it looks like the intake would be put further inland from the coast and lower than in the center.

Moreover, various filter styles and screens will be required to deter high intrusion and impingement levels in some nuclear power plants. As seen in some nuclear plants. A reporting scheme, with input on the effect of the intake and future changes, would have to be developed. It is not expected that spill impacts related to nuclear desalination would vary as of additional multiple network installations. Conservational strategies shall focus on heat and contamination of waste effluents in particular related to desalination operations and on their thermal effects on the aquatic ecosystem. The control approach will possibly be focused on heat remediation devices (e.g. heat pipe and heat exchanger), chilling facilities (tower or pond), or a mixture of the diffusers from the shore.

The location of the discharge in regions with heavy mixing streams will greatly decrease the discharge effect. The IAEA Health Guide No needs a control system for waste effluents. The implementation of NS-G-3.2 is suggested. In the case of a nuclear desalination plant comparison, primarily the temperature and copper amount of the discharge should be considered. The above-mentioned coastal impacts are projected to be lower than for alternate co-location solutions. The visual effects and the dimension of land usage, in particular, offer an incentive for even simpler mitigation steps in the event of nuclear desalination. Nevertheless, the design strategy for such a project would be somewhat different than how the other collocated solutions were designed. Since noise rates can be managed effectively by the correct acoustic preparations, the surveillance system, which reduces the risk for seawater pollution, will be focused mostly on a problem of pipeline leakage (Ns-G, 2002).

Besides, a monitoring program will be needed in the construction process to ensure that operation will induce increased sedimentation of the settling content that would, therefore, impact turbidity and, thus, photosynthetic in the coastal environment or increase the toxicity of this environment if major runoffs are occurring. The air consequences of a nuclear desalination plant are supposed to be negligible and this form of monitoring is also intended to only be focused on accordance with local regulations. Environmental planning will consider many aspects as far as socio-economic effects are concerned, including demographic growth, efficiency transition, or improvements, as well as infrastructure construction that would be required to endure these shifts and their environmental stress. The monitoring system, for example, is focused on population development, dynamism and liquid volume, surplus and emissions production, ecological impartiality, etc. The potential significant impacts of a nuclear purification plant such as a comparison capability could, without any negative effects on its equilibrium, be used to improve the socio-economic regional situation.

The related environmental effects would, therefore, be avoided. However, to achieve public acceptance, public health is perhaps the supreme-significant phase of nuclear purification. Chemical water health preparation must focus on the components, procedures, and tritium separation activities as well as food storage containers to confirm that the water supplies are not generated from polluted water. Consequently, the control scheme, of these three, the most important are: isolation loop, salinity loop and containers of water will be organized around multiple stages. Dependent on the detection apparatus and permissible assurance criteria, the water tanks needed to be checked for tritium activities numerous hours earlier discharge into the water supply network (Anastasov & Khamis, 2010).

# 11 Conclusion

A detailed analysis was done in this chapter on the different facets of nuclear desalination systems, including the numerous nuclear plants, the hybrid patterns, the protection, and environmental analyzes and on-site economic evaluations. It became evident that there was a substantial rise in the production of various nuclear reactors though compact reactors of limited scale (MSR) were granted considerable publicity. It is attributed to the many advantages they provide compared with big reactors, including modest assembly area, shorter construction period, cost-effective construction, and healthy service. Therefore, particularly for newcomer countries, it seems to be more appealing. The analysis also highlighted the role of the IAEA in leading work, advising, and funding initiatives for nuclear desalination worldwide particularly in developing countries. There are also the following conclusions:

- Until linking nuclear reactors to any desalination plant the water quality, health, potential pollution, and method of desalination plant should be carefully examined.
- In contrast to traditional fossil-fuel methods, the techno-economic experiments carried out in the literature have identified the viability and profitability of nuclear desalination.
- Work into nuclear hybrid desalination facilities has demonstrated many advantages to hybrid systems where the usage to waste heat in nuclear reactors may produce low-pressure steam. The thermal cycle (MSF or MED) may be aimed at this. In RO or membrane processes, electricity can be produced to power the required pumping mechanism. Optimal characteristics may be achieved from active desalination technologies.
- The most stable choice for "new coming" nations, who also lack experience in nuclear power plants, is the use of SMRs compared to those with larger reactors. Another danger factor is the confusion surrounding the environment of massive reactors. In construction (e.g. offshore architecture, which calls for a path of heat to pass or the use of heat pipes), however, the health protocols should be strictly enforced in service and quality control.

- Environmental studies of nuclear desalination plants have found that they are equal to the wind and hydroelectric capacity, having the least effects on the environment relative to other desalination plants.
- Tests revealed some desirable features for nuclear desalination, including environmental security by reducing the production of greenhouse gases, potential replacement of existing sources of energy (fossil fuel), and economic viability in rural places where fossil fuel does not occur. The findings also providing this system an overall considerate and generated more practical trust in its application. Future research activities, therefore, concentrate on comprehensive design studies that answer key engineering questions including multiple intermediate circuits for the safety of treated water.

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# **Desalination Battery**

# Zahra Pezeshki and Ahmed Kettab

## Abstract

The existence of the human race and all creatures are dependent on water and today the water contamination and scarcity are important problems that treat human life. There are many reasons for them such as industrial activities, waste discharge, oil spill, climate change, pollution, global warming, and high utilization. As the population, trade, and economy are increasing day by day, we must track the ways to overcome these problems and desalination is an efficient process that can help to eliminate them. This process can be defined as a system or batteries. Desalination batteries are utilized more to provide drinking water from seawater, but these batteries can be used for producing freshwater from recycled water or wastewater too. This chapter has focused on the desalination field from 1960 to 2020. It introduces the desalination batteries and their properties to fill the future water needs because all organisms on our planet need water for survival and this technology can do that.

## Keywords

Desalination • Salination • Battery • Energy Efficiency • Coulombic efficiency • Drinking Water • Fresh Water

### Abbreviations

AC	Alternative Current	
AEM	Anion Exchange Membrane	
BDI	Battery Electrode Deionization	
CDI	Capacitive Deionization	
CEM	Cation Exchange Membrane	
DB	Desalination Battery	

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DC Direct Current ED Electrodialysis EV Electric Vehicle HEV Hybrid Electric Vehicle **ICP-MS** Inductively Coupled Plasma Mass Spectrometry IEM Ion Exchange Membrane IOMDC Individually connected Quadruple Microbial **Desalination Cell** LED Light-Emitting Diode MA Metal-Air MDC Microbial Desalination Cell MEDC Microbial Electrolysis Desalination Cell MF Microfiltration NF Nanofiltration OER Oxygen Evolution Reaction ORR Oxygen Reduction Reaction POMDC Parallel Connected Quadruple Microbial Desalination ΡV Photovoltaic QMDC Quadruple Microbial Desalination Cell RES Renewable Energy Source RO **Reverse Osmosis** SD-CC Seawater Desalination-Carbon Capture SQMDC Serially Connected Quadruple Microbial **Desalination Cell** UF Ultrafiltration

# 1 Introduction

Today, traditional underground resources or rainwater collectors, as shown in Fig. 1 (Fang et al., 2019), are no longer sufficient to meet the demand for drinking water as a short and long-term needs for household demands and human use, such as irrigation and washing, etc., due to unbalanced division of drought and rainwater, extreme misuse of

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**Fig. 1** Schema of traditional underground resources or rainwater collectors (Fang et al., 2019)



groundwater resources as well as its insufficient recharge, and degradation of water quality (Pezeshki & Bielefeldt, 2002; Shaltami et al., 2020) related to the clearance of industrial and domestic wastes without adequate treatment (Thimmaraju et al., 2018). So, it is proved necessarily, if not essentially, to use the surface water such as seawater which is naturally provided by evaporation, condensation, precipitation, and collection in our environment (Hani et al., 2012; Kettab, 2020; Kettab & Bessenasse, 2005; Kettab & Janah, 2020; Tu et al., 2020). Figure 2 shows the water cycle in nature.

Meanwhile, the surface water contains impurities which have origins as follows (Kettab, 2020; Mandi & Kettab, 2014):

- (a) The dissolution of the surrounding bodies (rocks, transverse soils, etc.) They are responsible for pH and conductivity.
- (b) Mineral colloids (clays, silica gels, etc.) Which are responsible for turbidity.
- (c) Original materials which can have two origins:





- Natural origin: degradation products of plants, metabolites of algae, and micro-organisms.
- Artificial origin: due to urban, industrial, and agricultural pollution (pesticides, fungicides, herbicides, etc.).

Thus, surface water should undergo physical, chemical, and biological changes (Gafsi & Kettab, 2016) that make it drinkable. For this, it is, therefore, necessary to treat surface water from organic/mineral materials, soluble/insoluble materials, toxic/non-toxic materials, and inert/living materials by appropriate means, which are very knife-edge and require skilled work. To treat the surface water, we have two types of treatment processes, including (1) biological treatment which is called biodegradation (Pezeshki & Bielefeldt, 2002), and (2) physicochemical treatment. In modern treatment chains, we use the physical–chemical process which consists of three phases (Kettab, 2017, 2020; Oliveira et al., 2019; Pezeshki & Bielefeldt, 2002) (Fig. 3):

- (a) Pretreatment
- (b) Clarification processing
- (c) Bactericidal and virucidal treatment.

Also, water heavily loaded with dissolved mineral salts can have drawbacks in certain uses. Consequently the certain annoying ions, e.g.  $Ca^{2+}$  and  $Mg^{2+}$  (Hardness);  $CO_3^{2-}$  and  $HCO_3^-$ (Alkalinity) or  $CO_4^{2-}$  (Aggressiveness) must be totally or partially eliminated by a method which is called "desalination" (Saji et al., 2020; Tang et al., 2019). Desalination is divided into two processes including membrane (Dasgupta et al., 2020) and thermal processes (Kress, 2019; Yusufov et al., 2019). In this context, the elimination of the mineral salts could be done using several methods (Amin Abdel-Fatah & Al Bazedi, 2020). The most used are: (a) ion exchange, (b) Electrodialysis (ED) (Dasgupta et al., 2020; Qasem et al., 2020; Tang et al., 2019; Xu et al., 2020), which (a) and (b) commonly are used together with Microfiltration (MF), Ultrafiltration (UF), Nanofiltration (NF) (Das et al., 2018, 2019; Dasgupta et al., 2020; Oliveira et al., 2019), and

nanomaterials such as MXenes (Fig. 4) (Ihsanullah, 2020), (c) Reverse Osmosis (RO) (Fig. 5) which is known as the best method due to long-lasting membranes, such as graphene (Mehrdad & Moosavi, 2019) or calcite ooids (Bakr, 2020) due to having better performance (Bdour et al., 2020; Dasgupta et al., 2020; Hani et al., 2012; Kettab & Bessenasse, 2005; Khiari et al., 2019; Mandi & Kettab, 2014; Metaiche & Kettab, 2003, 2004, 2005a, 2005b; Metaiche et al., 2003; Oliveira et al., 2019; Saji et al., 2020), and (d) distillation, for example, with cool steam or solar distiller (Arnau et al., 2019; Benyahia, 2019; Kettab, 2020; Kress, 2019; Wazwaz & Khan, 2020) (see Fig. 6; Kress, 2019; Amin Abdel-Fatah & Al Bazedi, 2020)). These methods have many applications for the various fields of industry such as the food industry, pharmaceutical industry, electronics industry, electro-technical industry, etc. (Metaiche & Kettab, 2005a).

In Fig. 5, the components of a reverse osmosis system are divided into production series by a set of stages. The production of each stage supplies the next stage and so on so that the supply of each stage has characteristics identical to those of the production of the previous stage. The decline of all the stages is collected to give the total rejection of the system at the end. In the case of energy recovery, and for achieving the maximum rejection, this system is connected to a turbine (Al Zubaidi, 1989; Malek et al., 1996; Maurel, 1996; Metaiche & Kettab, 2005a; Metaiche et al., 2002).

In particular, desalination can be a straightforward, effective, and economical solution for using fresh/drinking water instead of the pipe network connection which creates transportation cost, labour cost, and extra fuel (Abozaid et al., 2020). It can be integrated with Renewable Energy Sources (RESs) (Caldera & Breyer, 2020; Goosen et al., 2011; Gude & Fthenakis, 2020), such as solar energy and wind (Al Jibouri & Buckley, 2020; Alsehli, 2019; Bdour et al., 2020; Charrouf et al., 2020; Chen et al., 2019a, 2019b; Ebrahimpour et al., 2020; Fang et al., 2019; Kandathil et al., 2019; Khiari et al., 2019; Pezeshki & Zekry, 2020; Rezk et al., 2020; Roslan et al., 2020; driven by the grid and

**Fig. 3** Example of the physicochemical process (Fang et al., 2019)


**Fig. 4** Applications of MXenes (Ihsanullah, 2020)







micro-grid networks, Electric Vehicles (EVs), and Hybrid Electric Vehicles (HEVs) called desalination system (Hani et al., 2012; Kettab, 2008; Pasta et al., 2012; Tu et al., 2020) (Fig. 7). The aggregated demand for this integration in kWh can be computed as follows (Eq. 1):

$$\forall t \cdot GE_t + dg_t + PV_t + WT_t + EV_{v2gt} + \Delta SoC_t \ge D_t + DS_t + ql_t + EV_{g2v,t} (1)$$

where GE<sub>t</sub> is the grid electricity import,  $dg_t$  is the diesel generator electricity, PV<sub>t</sub> is the electricity generated from the photovoltaic (PV) panels, WT<sub>t</sub> is the electricity generated from wind turbines,  $EV_{v2gt}$  is the discharge of the vehicle-to-grid electricity,  $\Delta SoC_t$  is the stationary battery change in charge condition,  $D_t$  is the base electricity demand, DS<sub>t</sub> is the consumption of the desalination plant,  $ql_t$  is the electricity losses from AC–DC conversion and battery operations, and EV<sub>g2v.t</sub> is the electricity consumed in charging the electric vehicles (Tu et al., 2020).

Desalination systems can be defined as a battery in which desalination batteries utilize an electrical energy input for extraction of e.g. Chloride, Cl, and sodium, Na, ions from seawater instead of generating electricity from salinity differences to produce drinking water. This battery can also be utilized to produce fresh water from the used water, such as wastewater or recycled water (Etale et al. 2020; Hani et al., 2012). It can be utilized as a low-cost rechargeable battery too (Pezeshki, 2020; Senthilkumar et al., 2019).

## **Fig. 6** Desalination methods (Kress, 2019)



**Fig. 7** Schema of desalination integration (Tu et al., 2020)



#### 2 Definition and Classification of Desalination Batteries

The desalination batteries have a simple structure and provide favourable energy efficiency. While they work at room temperature, they have lesser corrosion problems than the current desalination technologies. So, as shown in Fig. 8, the path integral of the energy consumption,  $\Delta E$ , versus q curve is used to offer the energy consumption and desalination cycle in real seawater for NaCl elimination in these batteries.

The  $\Delta E$  is computed by dividing the electrical energy to the whole of the ion removal during the desalination process as Eq. (2) (Lee et al., 2019a):

$$\Delta E = \frac{\int V \times I \times dt}{\int (C_i - C_e) \times \Phi \times dt (C_i - C_e) \times \Phi \times dt}$$
(2)

where  $c_e$  and  $c_i$  are the ion concentration of the treated and initial water, respectively. The V and I are the applied voltage and respective current during the desalination step, respectively. Also, t and  $\Phi$  are the periods of the experiment, and flow rate, respectively. So the charge efficiency,  $\eta$ , is computed as Eq. (3) (Lee et al., 2019a):

$$\eta = \frac{\int (C_i - C_e) \times \Phi \times dt}{\int I \times dt/F}$$
(3)

where *F* is the Faraday constant equal to 96,485  $\text{Cmol}^{-1}$ . In this section, the types of desalination batteries are defined and described (Pasta et al., 2012).



Fig. 8 The desalination cycle of real seawater offered for NaCl elimination (Pasta et al., 2012)

#### 2.1 NMO Desalination Battery

#### 2.1.1 Ag/AgCl as an Anionic Electrode

An NMO desalination battery can be composed of a Ag/AgCl negative/anionic electrode and  $Na_{2-x}Mn_5O_{10}$  nanorod positive/cationic electrode. Silver, Ag, has some benefits such as potential, stability, corrosion resistance, and bactericidal features, but it is expensive and has a weak electronic conductivity. This desalination battery creates 0.29 Whl<sup>-1</sup> energy consumption for the 25% salt removal (Pasta et al., 2012).

In this battery, the charge constructed on the electrode surface and stored in the electrical double layer is retained in the chemical bonds, i.e. the electrode material bulk. So the battery electrodes of this battery create higher capacity as well as less self-discharge than the capacitive electrodes (Conway, 1991; Pasta et al., 2012). Then, in the 4-stage charge/discharge process, the battery electrodes split seawater up freshwater. First, the fully charged electrodes are sunk in the seawater with a fixed current,  $-500 \ \mu A \ cm^{-2}$ . A fixed current is utilized to remove the ions, i.e. chloride or sodium ions, from the seawater (Fig. 9, Step 1). Second (Fig. 9, Step 2), after extracting the drinking water, it is replaced with another seawater. So, the electrodes are recharged again, release the ions, and produce the brine (Fig. 9, Step 3). Finally, (Fig. 9, Step 4), the brine is replaced with new seawater (Pasta et al., 2012).

The overall electrochemical reaction of this battery is as follows (Pasta et al., 2012):

$$5MnO_2 + 2Ag + 2NaCl \leftrightarrow Na_2Mn_5O_{10} + 2AgCl$$

For evaluating the Coulombic efficiency,  $\eta_{C.i}$ , of the desalinated solution, this solution is usually analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) as Table 1 where  $\eta_{C.i}$  is computed as Eq. (4) (Pasta et al., 2012). This formula is employed for all desalination batteries.

$$\eta_{C.i} = \frac{z_i F(n_I - n_F)}{Q} \tag{4}$$

Where  $z_i$  is the ion valence state, Q is the total charge flown,  $n_F$  and  $n_I$  are the final and the initial number of moles in the solution, respectively.

Figure 10 depicts the desalination cycle in the actual seawater for 25% NaCl elimination. Here, the energy consumption is 0.261 J (Pasta et al., 2012).

Currently using Na<sub>4</sub>Mn<sub>9</sub>O<sub>18</sub> as a nanorod positive/cationic electrode in an NMO desalination battery



Fig. 9 Schematic diagram of the NMO desalination battery work (Ag/AgCl as an anionic electrode) (Pasta et al., 2012)

Table 1       Coulombic efficiencies         and cations/anions selectivity       (Pasta et al., 2012)	Ion	Na <sup>+</sup>	K <sup>+</sup>	$Mg^{2+}$	Ca <sup>2+</sup>	Cl-	$SO_4^{2-}$
	Sea water (mg/L)	11,250	450	1400	450	18,500	2750
	25% removal (mg/L)	9840	430	1130	280	14,470	2750
	η <sub>C.25</sub>	47%	< 1%	9%	3%	87%	Control
	50% removal (mg/L)	7860	390	860	180	11,430	2750
	$\eta_{C.50}$	57%	< 1%	9%	3%	76%	Control



Fig. 10 The desalination cycle in the actual seawater for 25% NaCl elimination (Pasta et al., 2012)

has shown a good performance at a relatively small C-rate (C/19) with temperatures differing between 30 and 70  $^{\circ}$ C, and 0.92 J maximal energy (Shapira et al., 2018).

#### 2.1.2 PPy/PPyCl as an Anionic Electrode

An NMO desalination battery can be composed of a polypyrrole (PPy)/PPyCl negative/anionic electrode and Na2-xMn5O10 nanorod positive/cationic electrode. Polypyrrole can store chloride ions in seawater. During the charge in this battery, (Fig. 11a), the  $Na^+$  and  $Cl^-$  ions are drawn from NMO and PPyCl electrodes to the mixture, respectively. Then, after discharge as depicted in Fig. 11b, the Na<sup>+</sup> and Cl<sup>-</sup> ions in the electrolyte are embedded into the NMO and PPyCl electrodes, respectively (Kong et al., 2019).

**Fig. 11** Configuration of the NMO desalination battery (PPy/PPyCl as an anionic electrode), **a** charge, and **b** discharge (Kong et al., 2019)



The overall electrochemical reaction of this battery is as follows (Kong et al., 2019):

$$x$$
PPyCl + Na<sub>0.44</sub>MnO<sub>2</sub>  $\rightarrow x$ PPyCl + Na<sub>0.44-x</sub>MnO<sub>2</sub> +  $x$ Na<sup>+</sup>

Without any reduction in both capacity of 40 mAh/g and discharge voltage, the charge and discharge of this battery can continue for 200 cycles (Kong et al., 2019).

#### 2.2 NTP Desalination Battery

#### 2.2.1 Ag/AgCl as a Cationic Electrode

An NTP desalination battery can be composed of a  $NaTi_2(PO_4)_3$  negative/anionic electrode and Ag/AgCl positive/cationic electrode with an NaCl aqueous electrolyte for electrical energy supply and desalination. Therefore, in charge operation in this kind of battery, after extracting the Na<sup>+</sup> and Cl<sup>-</sup> from the electrolyte, they are embedded into the electrodes and desalination happens. In the reverse process, the salination happens by releasing the ions and storing electricity in the electrolyte. With the help of a solar cell or any other power source, this battery can be consecutively charged and discharged for 50 cycles without any reduction of both the discharge voltage and capacity, which provides a new awareness into both efficient renewable energy and seawater desalination utilization. The energy density of this battery is 49.9 Wh kg<sup>-1</sup> at a 55.1 Wh kg<sup>-1</sup> power density, which is high. The battery configuration has been depicted in Fig. 12 (Wang et al., 2018).

The overall electrochemical reaction of this battery is as follows (Wang et al., 2018):

$$NaTi_{2}(PO_{4})_{3} + 2Ag + 2Cl^{-} + 2Na^{+} \leftrightarrow Na_{3}Ti_{2}(PO_{4})_{3} + 2AgCl$$

This kind of desalination battery has been developed by reaching an energy efficiency of 71.9% and Coulombic efficiency of 99% in 200 cycles with a capacity of 37.4

mAh/g at a current density of 1000 mA/g (Chen et al., 2018).

The elemental composition of Na, P, and Ti, charge and discharge states, and the calculated atomic ratio of Na : P have been depicted in Table 2. Table 3 shows the elemental composition of Ag and Cl in the electrode, charge and discharge states, and the calculated atomic ratio of Cl : Ag.

#### 2.2.2 Bi/BiOCl as a Cationic Electrode

Another NTP desalination battery is composed of a  $NaTi_2(PO_4)_3$  negative/anionic electrode and Bi/BiOCl positive/cationic electrode with an NaCl aqueous electrolyte for electrical energy supply and desalination. Same as before battery, in this battery, after extracting the Na<sup>+</sup> and Cl<sup>-</sup> from the electrolyte in the charging process, they are embedded into the electrodes and then desalination happens, and in the reverse process, the salination happens by releasing the ions and storing electricity in the electrolyte (Fig. 13) (Nam & Choi, 2017).

The overall electrochemical reaction of this battery is as follows (Nam & Choi, 2017):

$$3/2NaTi_2(PO_4)_3 + Bi + 3Na^+ + Cl^- + H_2O \rightarrow 3/2Na_3Ti_2(PO_4)_3 + BiOCl + 2H^+$$

This battery is a newer type of an NTP desalination battery with Bismuth (Bi) and enables to do the desalination/salination process at  $\pm 1 \text{ mA cm}^{-2}$  with the average potential input of 0.2 *V*, means that the average potential input (Nam & Choi, 2017).

Table 4 shows the atomic ratios of the Bi electrode during the charge/discharge cycles.

#### 2.3 KFCN Desalination Battery

KFCN desalination battery is composed of a  $K_4Fe(CN)_6$  negative/anionic electrode and  $ZnCl_2$  positive/cationic





Anode: NaTi<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> + 2Na<sup>+</sup> +2e<sup>-</sup> ↔ Na<sub>3</sub>Ti<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub>

(φ = -0.61 V)

Total:  $NaTi_2(PO_4)_3 + 2Ag + 2CI^- + 2Na^+ \leftrightarrow Na_3Ti_2(PO_4)_3 + 2AgCI$ 

<b>Table 2</b> The elementalcomposition of Na, P, and Ti,charge and discharge states, andthe calculated the atomic ratio ofNa : P (Chen et al., 2018)	Atomic (%)	Na	Р	Ti	Na : P
	Initial NTP	13.9	50	36.0	0.280
	Charge	21.9	46.3	31.8	0.473
	Discharge	16.4	49.5	34.1	0.332

**Table 3** The elemental composition of Ag and Cl in the electrode, charge and discharge states, and the calculated the atomic ratio of Cl : Ag (Chen et al., 2018)

Atomic (%)	Ag	Cl	Cl:Ag
Initial state	100	0	0
Charge	61.7	38.3	0.621
Discharge	73.0	27.0	0.370

electrode with an NaCl aqueous electrolyte for electrical energy supply and desalination. In the charging process of this battery, after extracting the Na<sup>+</sup> and Cl<sup>-</sup> from the electrolyte, they are embedded into the electrodes and then desalination occurs. In the reverse process, the salination occurs by releasing the ions and storing electricity in the electrolyte. This process can be done with a solar cell or any power source (see Fig. 14) (Desai et al., 2017).

The overall electrochemical reaction of this battery is as follows (Desai et al., 2017):

$$Zn + 2K_3[Fe(CN_6)] + 2NaCl \leftrightarrow ZnCl_2 + 2K_3NaFe[(CN_6)]$$

With the help of a solar cell or any other power source, this battery can be consecutively charged and discharged for 100 cycles without any reduction of both the discharge voltage and capacity which gives new knowledge into both efficient renewable energy and seawater desalination utilization (Fig. 15) (Desai et al., 2017).

This battery is very suitable for hypersaline brines desalination too. The cell potential of this battery is + 1.25 V, i.e. it is 0.5 V greater than the other desalination batteries. It has a 81 mAh/g specific capacity for cathode and 828 mAh/g capacity for the anode. Using these capacities causes energy consumption to reduce. So, it is very cost-efficient (Desai et al., 2017).

#### 2.4 NASICON Desalination Battery

#### 2.4.1 C and Pt/C as Anionic and Cationic Electrodes

A NASICON desalination battery, as shown in Fig. 16, is composed of a  $Na_3Zr_2Si_2PO_{12}$  as a separator electrolyte,

Fig. 13 The configuration of the NTP desalination battery (Bi/BiOCl as a cathodic electrode) (Nam & Choi, 2017)

2 e



**Table 4** The atomic ratios of the
 Bi electrode during the charge and discharge states (Nam & Choi, 2017)

Atomic (%)	Bi:Cl
Initial state	0.005
First charge	0.011
First discharge	0.005
Second charge	0.013
Second discharge	0.005





**Fig. 15** The charge/discharge cycling of KFCN desalination battery (Desai et al., 2017)



**Fig. 16** Schematic diagram of NASICON desalination battery (C and Pt/C as Anionic and Cationic Electrodes) (Zhang et al., 2018)

hard Carbon, C, as an anode, and Pt/C electrocatalyst as a cathode. In this battery, during the charge, the Na<sup>+</sup> ions are taken out and transferred to the anode via the NASICON separator. Afterward, they are reserved in the hard carbon. So, during the charge,  $Cl^-$  ions are taken out and transferred to the cathode via the Anion Exchange Membrane (AEM), after coupling with H<sup>+</sup> ions, yielded in the electrochemical Oxygen Evolution Reaction (OER) operation (Zhang et al., 2018):

 $Charge: 2H_2O \rightarrow O_2 + 4H^+ + 4e^- \ \ \text{to form HCl}$ 

Then, during the discharge, Na<sup>+</sup> ions reserved in the hard carbon are transferred to the cathode via the NASICON separator, after coupling with  $OH^-$  ions yielded in the electrochemical Oxygen Reduction Reaction (ORR) proceeding (Zhang et al., 2018):

 $Discharge: O_2 + 2H_2O + 4e^- \rightarrow 4OH^- \ \ to \ form \ NaOH$ 

This battery can be consecutively charged and discharged for 10 cycles without any reduction of both the discharge voltage and capacity, but after the 10th cycle, the pH value decreases because of  $H^+$  produced by the OER (Zhang et al., 2018).

The comparison of ion concentration in the first seawater and after 10th charging is shown in Table 5.

#### 2.4.2 C as a Cationic Electrode

A NASICON desalination battery can also be composed of a  $Na_3Zr_2Si_2PO_{12}$  as a separator and Carbon, C as a positive/cationic electrode with an NaCl aqueous electrolyte for desalination and electrical energy storage. In the desalination process of this kind of battery, there are two various cells: one which is utilized as a carbon-paper electrode, and

**Table 5** Comparing the ionconcentration in the first seawaterand after 10th charging (Zhanget al., 2018)

	Ion concentration (mg/L)					
Atomic (%)	First seawater	After 10th charging				
Na <sup>+</sup>	10,166	9072				
$Mg^{2+}$	1229	1171				
K <sup>+</sup>	395	369				
Ca <sup>2+</sup>	450	429				
Cl <sup>-</sup>	19,916	18,176				
$SO_{4}^{2-}$	2754	2681				
Total	34,910	31,898				

another utilized as an Ag-foil electrode to obtain the chloride ions. Then, these two cells are charged at a constant current of 0.1 mA cm<sup>-2</sup>. The charging is led to the Cl<sup>-</sup> ions oxidation in the seawater to form AgCl on Ag foil. Then, the Na<sup>+</sup> ions are transferred into the seawater via the NASI-CON separator, followed by Na deposit. Afterwards, Na<sup>+</sup> and Cl<sup>-</sup> are removed from seawater and desalination happens. During the discharging, Na is oxidized into Na<sup>+</sup> and transferred to the carbon-capture via a NASICON separator. Then, the ORR happens in the carbon electrode and provides electrical power. The created OH<sup>-</sup> ions produce the air-capture process of CO<sub>2</sub> gases from the ambient atmosphere by carbonation process because of the Ca<sup>2+</sup> ions reaction in seawater (Fig. 17) (Bae et al., 2019).

The overall electrochemical reaction of this battery is as follows (Bae et al., 2019):

 $Na^{+} + e^{-} \leftrightarrow Na$   $O_{2} + 2H_{2}O + 4e^{-} \leftrightarrow 4OH^{-}$   $2Cl^{-} \rightarrow Cl_{2} + 2e^{-}$   $Ag + Cl^{-} \leftrightarrow AgCl + e^{-}$   $4Na + O_{2} + 2H_{2}O \leftrightarrow 4NaOH$   $Ag + Na^{+} + Cl^{-} \leftrightarrow AgCl + Na$ 

This kind of battery is called Seawater Desalination– Carbon Capture (SD-CC) battery too, because carbon-capture and Na-collection, are separated by NASI-CON membrane. At a current of  $0.1 \text{ mA cm}^{-2}$ , the sequential charge and discharge of this battery result in the



Table 6       The ion concentrations         after and before the SD-CC	Ions (ppm)	Na	Mg	K	Ca	Cl	SO <sub>4</sub>
battery charge with Ag-foil	First seawater	11,000	1300	500	400	18,000	2400
electrode (Bae et al., 2019)	After charging	300	1200	300	400	2400	2400

97.3% Na<sup>+</sup> and 86.7% Cl<sup>-</sup> removal from the drinking water and CO<sub>2</sub> capture during 50 h. Figure 17 depicts this process (Bae et al., 2019).

Table 6 depicts the ion concentrations after and before the SD-CC battery charge with an Ag-foil electrode (Bae et al., 2019).

#### 2.5 **CUFCN Desalination Battery**

is composed of a A CUFCN desalination battery  $Cu_3[Fe(CN)_6]_2$ negative/anionic electrode and Bi positive/cationic electrode with an NaCl aqueous electrolyte for electrical energy storage and desalination. During the charge in this battery, after extracting the Na<sup>+</sup> and Cl<sup>-</sup> from the electrolyte, they are embedded into the separate cathode and anode electrodes and desalination happens, and in the reverse process, the salination happens by releasing the ions and storing the electricity in the electrolyte. Figure 18 depicts this process (Nam et al., 2019).

The overall electrochemical reaction of this battery is as follows (Nam et al., 2019):

$$Cu_{3}[Fe^{III}(CN)_{6}]_{2} \cdot nH_{2}O + 2xNa^{+} + 2xe^{-}$$
  
$$\leftrightarrow Na_{2x}Cu_{3}[Fe_{x}^{II}Fe_{1-x}^{III}(CN)_{6}]_{2} \cdot nH_{2}O$$

In this battery, both CUFCN and Bi are stable in acidic and neutral solutions. So, membranes are not essential for

Fig. 18 Schematic diagram of CUFCN desalination battery (Nam et al., 2019)

this battery. They can do the desalination without the use of a membrane and this characteristic makes this battery unique and simple (Nam et al., 2019).

#### 2.6 **Redox-PSQ Desalination Battery**

A redox-PSQ desalination battery is composed of 3 electrodes. During the oxidation process in this battery, the ion of Cl<sup>-</sup> is transferred from seawater into the ferrocyanide molecule immobilized polysilsesquioxane (redox-PSQ) cathode electrode, means that  $PSQ - [Fe(CN)_6]^{4-}$  is converted to  $PSQ - [Fe(CN)_6]^{3-}$ . Then, after the desalination process is done, the redox polymer electrode is taken out, cleaned with water, and then coupled with Mg anode in the electrolyte containing 100 mM KNO<sub>3</sub> mixture. The red Light-Emitting Diode (LED) can be used for this process which is turned on because of the produced high intensity and after some time it is turned off. This reaction is as follows (Silambarasan & Joseph, 2018):

$$\begin{split} \mathbf{Mg} + 2\mathbf{PSQ} &- \left[\mathbf{Fe}(\mathbf{CN})_{6}\right]^{3-} \cdot \mathbf{Cl}^{-} \\ &\rightarrow \mathbf{Mg}^{2+} + 2\mathbf{PSQ} - \left[\mathbf{Fe}(\mathbf{CN})_{6}\right]^{4-} + 2\mathbf{Cl}^{-} \end{split}$$

Overall, in this battery, in the charging process, after extracting the Na<sup>+</sup> and Cl<sup>-</sup> from the electrolyte, they are embedded into the electrodes and then desalination happens,



The overall electrochemical reaction of this battery during the desalination and salination is as follows (Silambarasan & Joseph, 2018):

$$\begin{split} \text{Desalination} &: C_1 \text{Ni}^{2+} \left[ \text{Fe}(\text{CN})_6 \right]^{3-} + \text{PSQ} - \left[ \text{Fe}(\text{CN})_6 \right]^{4-} + \text{seawater} \\ &\rightarrow C_2 \text{Ni}^{2+} \left[ \text{Fe}(\text{CN})_6 \right]^{4-} + \text{PSQ} - \left[ \text{Fe}(\text{CN})_6 \right]^{3-} \cdot \text{Cl}^- \end{split}$$

$$\begin{split} \text{Salination} &: C_1 \text{Ni}^{2+} \left[ \text{Fe}(\text{CN})_6 \right]^{4-} + \text{PSQ} - \left[ \text{Fe}(\text{CN})_6 \right]^{3-} \cdot \text{Cl}^- + 0.1 \text{ MKNO}_3 \\ & \rightarrow C_2 \text{Ni}^{2+} \left[ \text{Fe}(\text{CN})_6 \right]^{3-} + \text{PSQ} - \left[ \text{Fe}(\text{CN})_6 \right]^{4-} + \text{C}^+ + \text{Cl}^- \end{split}$$

In this process, the net input voltage of this battery is 68 mV for LED (Silambarasan & Joseph, 2018).

#### 2.7 BDI Desalination Battery

#### 2.7.1 CuHCFs as Electrodes

The Battery Electrode Deionization (BDI) is a new electrochemical cell that utilizes capacitive electrodes, such as copper hexacyanoferrate (CuHCF) (Tang et al., 2019), i.e.  $Cu[Fe^{III}(CN)_6]$ , to reduce the energy requirements (Fig. 20) for desalinating water. When this battery is charged, anions are absorbed into the anode and cations are absorbed into the cathode, but the voltage must not be higher than 1.2 V, because the irreversible oxidation and parasitic reactions happen. When it is discharged, the cations are released via the Cation Exchange Membrane (CEM) and anions are separated via the AEM. This battery with an applied voltage of 0.6 V reaches to desalination up 100 to

**Fig. 19** Schematic diagram of redox-PSQ desalination battery (Silambarasan & Joseph, 2018)



Fig. 20 Schema of effluent concentration versus energy consumption (Kim et al., 2017)

mg – NaCl/g – electrode, 50 mM NaCl effect, and less energy than Capacitive Deionization (CDI) (Singh et al., 2018). Figure 21 depicts the schematic diagram of this BDI desalination battery with the direction of Na<sup>+</sup> and Cl<sup>-</sup> in every cycle (Kim et al., 2017).

The overall electrochemical reaction of this battery is as follows (Kim et al., 2017):

$$\operatorname{NaCu}\left[Fe^{\operatorname{III}}(\operatorname{CN}_{6}) + x\operatorname{Na}^{+} + xe^{-} = \operatorname{Na}_{1+x}\operatorname{CuFe^{II}}(\operatorname{CN}_{6}]_{x}\left[\operatorname{Fe^{III}}(\operatorname{CN}_{6})\right]_{1-x}\right]_{1-x}$$

In this battery, the decrease of the density causes energy consumption to reduce, but energy recovery has less impact. For example, for the current density of 20  $\text{Am}^{-2}$ , 10% of



**Fig. 21** BDI desalination battery (CuHCFs as electrodes): **a** real schema, **b** schematic diagram (Kim et al., 2017)



energy is recovered, but for 5  $\text{Am}^{-2}$ , 51% of energy is recovered (Son et al., 2020).

## 2.7.2 NaNiHCF and NaFeHCF as Electrodes

Another BDI utilizes battery electrodes, such as sodium iron hexacyanoferrate (NaFeHCF) as the cathode, as well as sodium nickel hexacyanoferrate (NaNiHCF) as the anode to reduce the energy requirements (Fig. 22) for desalinating water. When this battery is charged, anions are absorbed into the anode, and cations are absorbed into the cathode. When it is discharged, the cations are released via the CEM, and anions are separated via the AEM. This battery has a 59.9 mg/g desalination capacity with productive energy consumption, e.g. 0.34 Wh/L for 40% Na ion removal. The ion removal is computed based on Eq. (5). Figure 22 shows the schematic diagram of this BDI desalination battery (Lee et al., 2017).

ion removal(%) = 
$$\frac{c_i - c_e}{c_i} \times 100$$
 (5)

The overall electrochemical reaction of this battery is as follows (Lee et al., 2017):

Charging:

positive electrode : 
$$Na_{x1}Ni[Fe(CN_6)]$$
  
 $\rightarrow Na_{x1-x}Ni[Fe(CN_6)] + xNa^+$ 

Discharging:

positive electrode : 
$$Na_{x3}Ni[Fe(CN)_6] + x_{sol(+)}Na^+$$
  
 $\rightarrow Na_{x3-xsol(+)}Ni[Fe(CN)_6]$ 

negative electrode : 
$$Na_{x4}Fe[(CN)_6]$$
  
 $\rightarrow Na_{x4-y}Fe[(CN)_6] + yNa^+$ 

In this battery, evaluating the Coulombic efficiency of the desalinated solution is analyzed by ICP-MS as Table 7 (Lee et al., 2017).

#### 2.7.3 FMN-Na and TEMPO as Electrodes

A newer BDI desalination battery utilizes TEMPO, i.e. 4-hydroxy-2,2,6,6-tetramethyl piperidine-1-oxyl, as catholyte and FMN-Na, i.e. Riboflavin-5'-phosphate sodium salt dihydrate, as anolyte electrodes, to reduce the energy requirements (Fig. 23) for desalinating water. When this battery is charged, anions are absorbed into the anode, and cations are absorbed into the cathode. When it is discharged, the cations are released via the CEM, and anions are separated via the AEM. This battery has stability up to 50 cycles as well as the better energy efficiency of 25% and Coulombic efficiency of  $\sim 80\%$  with maintaining the capacity of 0.2 mAh. Also, it is a cost-effective renewable energy storage battery (Liang et al., 2019).

The overall electrochemical reaction of this battery is as follows (Liang et al., 2019):

Charging:

 $Cathode: TEMPO + Cl^{-} \rightarrow TEMPO - Cl + e$ 

Anode : 
$$FMN - NA + e + Na^+ \rightarrow FMN - Na_2$$

Discharging:

Cathode : TEMPO -  $Cl + e \rightarrow TEMPO + Cl^{-}$ 

Anode :  $FMN - Na_2 \rightarrow FMN - Na + e + Na^+$ 

Figure 24 shows the charge/discharge capacity, the Coulombic, and energy efficiency for four various densities (Liang et al., 2019).

**Fig. 22** Schematic diagram of the BDI desalination battery (NaNiHCF and NaFeHCF as electrodes) (Lee et al., 2017)



**Table 7**Coulombic efficienciesand cations/anions selectivity(Lee et al., 2017)

Ion	Na <sup>+</sup>	K+	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Cl-	$SO_{4}^{2-}$
Sea water (mM)	477.5	10.8	58.9	11.1	512.1	31.5
Ion removal (average, mM)	190.2	9.7	2.7	1.2	199.7	3.5
Ion removal (average, %)	39.9	90.1	4.5	10.5	39.0	11.0
Dilute solution (charging, mM)	291.5 ± 8.6	$\begin{array}{c} 0.9\ \pm\\ 0.4 \end{array}$	56.7 ± 1.2	9.8 ± 0.3	314.8 ± 13.9	$\begin{array}{c} 27.8\ \pm\\ 0.9\end{array}$
Dilute solution (discharging, mM)	283.1 ± 15.5	1.3 ± 0.6	56.5 ± 1.8	$10.2 \pm 0.3$	310.1 ± 15.6	$\begin{array}{c} 28.2 \pm \\ 1.2 \end{array}$





Charge-desalination process

Discharge-desalination process



Fig. 24 The charge/discharge capacity, the Coulombic, and energy efficiency for various densities (Liang et al., 2019)

#### 2.7.4 NVP and CNT as Electrodes

Another BDI can utilize battery electrodes such as sodium vanadyl phosphate dehydrate,  $Na_{0.5}VOPO_4 \cdot 2(H_2O)$  (NVP), and vanadyl hydrogen phosphate hemihydrate,  $VOHPO_4 \cdot 0.5(H_2O)$  (CNT), to reduce the energy requirements for desalinating water. When this battery is charged, anions are absorbed into the anode, and cations are absorbed into the cathode. When it is discharged, the cations are released via the CEM, and anions are separated via the AEM. This battery has the stability of up to 280 cycles as well as a capacity of 24.3 mg/g and a good charge efficiency of 85%. During the 100 cycles, battery energy storage increases by 47% (Fig. 25) (Lee et al., 2019b).

Table 8 shows the atomic ratios of the BDI battery with NVP and CNT as electrodes (Lee et al., 2019b).



**Fig. 25** Schematic diagram of the BDI desalination battery (NVP and CNT as electrodes) (Lee et al., 2019b)

#### 2.7.5 Ag and NiHCF as Electrodes

The other BDI can utilize battery electrodes such as Ag and nickel hexacyanoferrate, NiHCF (Singh et al., 2018) as negative and positive electrodes to reduce the energy requirements for desalinating water. When this battery is charged, anions are absorbed into the anode, and cations are absorbed into the cathode. When it is discharged, the cations are released via the CEM, and anions are separated via the AEM. This battery has stability up to 100 cycles at a 30 Am<sup>-2</sup> current density, but the capacity has a loss of 81.3% (Fig. 26) (Lee et al., 2019a).

The overall electrochemical reaction of this battery is as follows (Lee et al., 2019a):

NaNi 
$$[\text{Fe}^{3+}(CN)_6] + x\text{Na}^+ + xe^-$$
  
 $\rightarrow \text{Na}_{1+x}\text{Ni} [\text{Fe}^{3+}(CN)_6]_{1-x} [\text{Fe}^{3+}(CN)_6]_x$   
 $x\text{Ag} + x\text{Cl}^- \rightarrow x\text{AgCl} + xe^-$ 

This battery can be a viable system due to offering an optimal environment for the electrodes (Lee et al., 2019a).

#### 2.7.6 Ag and AgCl as Electrodes

A BDI can utilize battery electrodes such as Ag and AgCl (Ahn et al., 2020) as negative and positive electrodes to reduce the energy consumption for desalinating water too. As the same as before, when this battery is charged, anions are absorbed into the anode, and cations are absorbed into the cathode. When it is discharged, the cations are released and separated via the CEM (Fig. 27). These electrodes have a great capacity of 250 mAh/g, less solubility, and fast kinetics in saltwater (Ahn et al., 2018, 2019; Bakker et al., 2012; Blair & Murphy, 1960; Fighera et al., 2017; Pasta et al., 2012; Srimuk et al., 2019).

This battery can have a great salt adsorption capacity and fast desalination under low voltage operation (Ahn et al., 2020).

#### 2.7.7 C and Pt as Electrodes

A BDI can utilize battery electrodes such as carbon C, and Pt coated with carbon as negative and positive electrodes to reduce the energy requirements for desalinating water too. When this battery is charged, anions are absorbed into the anode, and cations are absorbed into the cathode. When it is discharged, the cations are released via the CEM, and anions are separated via the AEM. This kind of BDI called Microbial Desalination Cell (MDC) is shown in Fig. 28. The removal efficiency of this battery has been shown in Table 9 (Alhimali et al., 2019).

Today, newer MDCs can solve the imbalance pH problem, due to having a desalination rate of 24.3 mg/h with a 47 mA sequential electrical current recovery during up to 3 cycles of anolyte refreshment (Jafary et al., 2020a). For **Table 8**The atomic ratios of theBDI battery with NVP and CNTelectrodes (Lee et al., 2019b)

Atomic (%)

electrodes

BDI battery with NVP and CNT

0

 $26.8 \pm$ 

12.01

Р

 $8.2 \pm$ 

2.08

Prussian Blue Analogue Prussian Blue Analogue Nat Brackish Water Fresh Water Fresh Water Ct Side Ct Side Channel Ct Side Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Channel Chan	Prussian Blue Analogue Na <sup>+</sup> CEM Brackish Water Brackish Water Concentrated Water Concentrated Water Channel Side Channel Silver (b) Regeneration Step
solution Electrode	silver Electrode

V

7.99 ±

2.03

Na

 $2.39 \pm$ 

1.21

Step 1

Step 2

enhancing the desalination rate up to  $48.12 \pm 2.16$  mg/h, i.e. up to  $98.82 \pm 0.34\%$ , and power recovery up to 17.18 mW, another technology which is called Quadruple Microbial Desalination Cell (QMDC) is employed. Figure 29 depicts the components of the QMDC battery (Jafary et al., 2020b).

There are three QMDC batteries including an Individually connected Quadruple Microbial Desalination Cell (IQMDC) battery, Parallel Connected Quadruple Microbial Desalination Cell (PQMDC) battery, and Serially Connected Quadruple Microbial Desalination Cell (SQMDC) battery. Table 10 compares these three desalination batteries (Jafary et al., 2020b).

#### 2.7.8 C and Stainless Steel as Electrodes

The BDI can also utilize battery electrodes such as carbon, C, and stainless steel as negative and positive electrodes to reduce the energy consumption for desalinating water. When this battery is charged, anions are absorbed into the anode, and cations are absorbed into the cathode. When it is discharged, the cations are released via the CEM, and anions are separated via the AEM. This battery, called Microbial Electrolysis Desalination Cell (MEDC), has been depicted in Fig. 30 (Li et al., 2020).

This battery shows a Coulombic efficiency of  $8.5 \pm 1.1\%$  as well as the removal of  $75.5 \pm 1.4\%$ . Also, the recovery efficiency of the phosphate and nitrogen reaches  $66.7 \pm 4.7\%$  and  $66 \pm 5.3\%$ , respectively (Li et al., 2020).

Table 11 depicts the MEDC performance under various voltages and Ion Exchange Membrane (IEM) pairs (Li et al., 2020).

#### 2.8 MA Desalination Battery

Metal-Air (MA) desalination battery (Fig. 31) is composed of Al as a negative/anionic electrode and air as a positive/cationic electrode with an NaCl aqueous electrolyte

**Fig. 26** Schematic diagram of the BDI desalination battery (Ag and NiHCF as electrodes), **a** desalination or discharging step, and **b** salination/regeneration or charging step (Lee et al., 2019a)



**Fig. 28** Schematic diagram of **a** the BDI desalination battery called MDC battery, and **b** experimental MDC for removal (C and Pt as electrodes) (Alhimali et al., 2019)



**Table 9** The removal efficiency(%) of another BDI battery calledMDC battery (C and Pt aselectrodes) (Alhimali et al., 2019)

Battery no	Liquid being desalinated	Remova	Removal efficiency			
		Na <sup>+</sup>	$SO_{4}^{2-}$	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Cl-
1	Pure NaCl solution	91	-	-	-	84
2	Synthetic seawater	89	-	87	96	81
3	Real seawater	85	72	79	86	84

for electrical energy storage and desalination. When this battery is charged, anions are absorbed into the anode, and cations are absorbed into the cathode. When it is discharged, the cations are released via the CEM, and anions are separated via the AEM. According to the theory, desalination can be calculated by the current, I, and F as Eq. (6) (Chen et al., 2019b).

$$mol_{NaCl} = \frac{It}{nF}$$
(6)

where n is the number of charges.

This battery reaches  $2.83 \text{ Wm}^{-2}$ , i.e. the maximum power density, at a current density of  $6.58 \text{ Am}^{-2}$  and output voltage of 0.43 V which results in the 37.8% salt removal from the drinking water with 10 mWh energy generation during 14 h. In this battery, the desalination rate and energy generation are restricted due to the dissolved oxygen concentration in the cathode (Ghahari et al., 2019).

The overall electrochemical reaction of this battery is as follows (Ghahari et al., 2019):

$$Al + 3H_2O \rightarrow Al(OH)_3 + 3H^+ + 3e^-$$

**Fig. 29** The components of the QMDC battery (Jafary et al., 2020b)



Battery name	$\eta(\%)$	NaCl(g/L)	Power (mW) <	Power density $(mW/m^2)$
IQMDC battery	97.22	35	8.75	2734
PQMDC battery	98.82	35	17.18	2684
SQMDC battery	97.67	35	8.1	1265



**Table 10**Comparing QMDCbatteries (Jafary et al., 2020b)



$$3H^+ + 3Cl^- \rightarrow 3HCl$$

$$Al + 4OH^- \rightarrow Al(OH)_4^- + 3e^-$$

 $\mathrm{Al(OH)}_3 + 3\mathrm{HCl} \rightarrow \mathrm{AlCl}_3 + 3\mathrm{H}_2\mathrm{O}$ 

Table 12 depicts the MA desalination battery performance with 0.2 mol NaCl (Ghahari et al., 2019).

Table 11The MEDCperformance under variousvoltages and IEM pairs (Li et a2020)

IEM pair	Voltage (V)	Influent		Removal effici	iency (%)	Recovery efficiency (%)	
		Ν	Р	N	Р	N	Р
1	0.8	27.5	9.7	$25.8 \pm 15.9$	$19.5\pm 6.5$	$3.4 \pm 2$	$21.5\pm3.5$
	1	28.6	10.5	$35.9\pm2.6$	35 ± 12.9	$10 \pm 0.1$	$20.2\pm5.3$
	1.5	26.7	9.4	36.1 ± 4	$30.1 \pm 11.3$	$11.3\pm5.1$	36.6 ± 3.9
2	1.5	29.4	9.6	$43.4 \pm 13.7$	34.8 ± 2.1	$21.4\pm2.5$	$29.6\pm0.1$
	2	28.8	9.2	$41.6\pm5.1$	$49.7 \pm 4$	$29\pm4.2$	37.5 ± 3.3
3	2	29.4	8.8	$79.2\pm5.8$	$79.2\pm5.8$	66 ± 5.3	66.7 ± 4.7

**Fig. 31** The MA desalination battery: **a** schematic diagram, **b** real image (Ghahari et al., 2019)

## Metal-Air Desalination Battery (MADB)





# **3** Properties of Desalination Batteries and Future Approaches

The most important properties of the desalination batteries include removal efficiency, energy consumption, specific capacity, and charge/discharge cycle. Here, these properties have shown in Table 13 for all the desalination batteries in this chapter.

Further developments must be accomplished for future desalination batteries as follows:

1. The NMO desalination battery technology must be developed via integration with other desalination

 Table 12
 The MA desalination battery performance with 0.2 mol NaCl (Ghahari et al., 2019)

Electrolyte	Desalination (g/L)	Desalination (%)	Average current density $(mA/cm^2)$
0.2 M NaCl	2.54	21	0.55

#### Table 13 The properties of the desalination batteries

Desalination Batteries (DBs)	Removal Efficiency (%)	Energy Consumption (J or Wh/L)	Specific Capacity (mAh/g)	Charge/Discharge Cycle	Reerencesfs
NMO DB	_				
Ag/AgCl as an Anionic Electrode	25	0.261 J	35	1	Pasta et al. (2012)
	-	0.92 J	23	1	Shapira et al. (2018)
PPy/PPyCl as an Anionic Electrode	-	-	40-45	200	Kong et al. (2019)
NTP DB	-				
Ag/AgCl as a Cationic	-	-	70	50	Wang et al. (2018)
Electrode	71.9	$\sim$ 50 Wh/L	37.4	200	Chen et al. (2018)
Bi/BiOCl as a Cationic Electrode	80	0.285 J	133	30	Nam and Cho (2017)
KFCN DB	85	22.8–137.2 J	81 (Cathode), 828 (Anode)	100	Desai et al. (2017)
NASICON DB	_				
C and Pt/C as Anionic and Cationic Electrodes	8.63	4.69 Wh/L	200	10	Zhang et al. (2018)
C as a Cationic Electrode	86.7–97.3	-	-	-	Bae et al. (2019)
CUFCN DB	75.6	-	~ 60	20	Nam et al. (2019)
Redox-PSQ DB	-	-	-	30	Silambarasan and Joseph (2018)
BDI DB				I	
CuHCF as Electrodes	50	-	-	50	Kim et al. (2017)
	71–3	-	29–35	51	Son et al. (2020)
NaNiHCF and NaFeHCF as Electrodes	40	0.756 J (0.34 Wh/L)	56.2	100	Lee et al. (2017)
FMN-Na and TEMPO as Electrodes	25	-	0.2–0.4	50	Liang et al. (2019)
NVP and CNT as Electrodes	87	-	50	280	Lee et al. (2019b)
Ag and NiHCF as Electrodes	-	-	~ 60	100	Lee et al. (2019a), Singh et al. (2018)
Ag and AgCl as Electrodes	80	0.19 Wh/L	250	100	Ahn et al. (2020)
C and Pt as Electrodes	85–91	-	-	4 (during 10 days)	Alhimali et al. (2019)
	$98.82 \pm 0.34$	-	-	3	Jafary et al. (2020b)
C and Stainless Steel as Electrodes	75.5 ± 1.4	-	-	30	Jafary et al. (2020a)
MA DB	37.8	-	-	-	Chen et al. (2019b)

methods and RESs (Chen et al., 2018; Pasta et al., 2012).

2. With further optimization of the electrode structure in NTP desalination batteries, the electrochemical optimization will be feasible by improving the capacity,

rate, as well as cyclability and open up novel possibilities for desalination processes (Nam & Choi, 2017).

3. Cost minimization must be considered as a paradigm for energy storage of a KFCN desalination battery (Desai et al., 2017).

- 4. Further development must be done for the cathode of the SE-CC desalination battery to improve energy efficiency (Bae et al., 2019).
- 5. The CUFCN desalination battery can be used as a proper desalination battery with further development in cyclability (Nam et al., 2019).
- Other polymers such as PPy, poly-aniline, and so on can be utilized for developing the redox-PSQ desalination battery for desalination, recycling water as well as wastewater treatment (Silambarasan & Joseph, 2018).
- As the performance of other electrochemical systems is weaker than the BDI desalination batteries, so additional improvements, e.g. enhancing the cycle performance and charge utilization for desalination, can reduce their internal resistances (Kim et al., 2017).
- Also, in BDI desalination batteries, the enhancement of water treatment is feasible with the development of the optimization of water recovery and flow-type reactor (Ahn et al., 2020; Lee et al., 2017). They can also contribute better with RESs (Liang et al., 2019).
- The BDI desalination batteries with NVP and CNT electrodes and also, Ag and NiHCF electrodes need further experiments with various ion concentrations and systems for desalination processes (Lee et al., 2019b; Singh et al., 2018).
- For real applications, further studies are required for comparing the design of MDC desalination batteries (Jafary et al., 2020b).

#### 4 Conclusion

Water is an essential need that is irreplaceable. Currently, none of the technologies of this century allows us to create water and nothing can be substituted for it because it cannot be reproduced. Therefore, it must be protected by strict legislation and it must be considered as a very precious and increasingly rare commodity.

We must promote the progress and socio-economic development in our countries, and share the fruitful results of global growth with our citizens. It all depends on our ability to find ingenious and innovative solutions. It all depends on our ability to manage this precious and capricious resource well.

So, not only knowledge, science, technology transfer, and multidisciplinary cooperation are important, but also financing, investment, and good governance are imperatives for this achievement. Likewise, their sustainable development is not feasible without the correct management of the water resource, particularly for arid and semi-arid countries. This issue is very important from an economic, social, cultural, and strategic point of view. So, desalination of water is the future of humanity, especially for countries that are already under water stress, or for countries that will reach it by 2050. Water for everyone is everyone's business (Kettab, 2014).

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## **Carbon Nanotubes Composite Membrane for Water Desalination**

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#### Abstract

The demand for freshwater has enormously risen the water stress in various parts of the globe. Accessibility of clean water is crucial for sustainable development involving socioeconomic and environmental promotion. Considering the fact that 96.5% of all Earth's water is related to seawater, desalination (producing clean potable water from sea or saline water) can be considered as a leading solution to fulfill water scarcity problem. Among various advanced and conventional techniques, carbon nanotube (CNT) membrane has become an attractive alternate for most of water treatment methods owing supreme features such as easy operationality, low energy and expense requirement, high water permeability, permselectivity, and stability. CNTs can be grown in vertically aligned CNTs (VACNTs), transverse or horizontally aligned CNTs (HACNTs), and mixed matrix membranes (MMMs) shapes. CNT membranes are mostly synthesized by chemical vapor deposition (CVD), laser ablation (LA), and arc discharge (AD) methods. Researchers have investigated the effect of various factors on salt rejection and water flux such as dispersion quality, oscillating pressure, number of deposition cycle (while synthesizing the membrane), type of filler, fabrication method, temperature, and contact time. In this chapter, initially a general description of membrane filtration, their features in desalination, and synthesization methods are presented. Afterward, various intrinsic, thermal, and

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mechanical characteristics of CNTs (which make them pioneer in desalination goals) are mentioned. In following, the results of several studies and their key findings are noted. Finally, the challenges, perspectives, and future directions of this technology to enter desalination markets are explained.

#### Keywords

Remediation • Carbon nanotube • Membrane • Desalination • Water treatment • Wastewater

#### 1 Introduction

Limited freshwater resources, rapid population growth, industrialization, and consequently water pollution have generated significant challenges for providing clean water for human life and ecosystem (Ma et al., 2017; Taghipour & Ayati, 2017; Taghipour et al., 2017, 2019). On the other hand, global warming which is one of the fundamental agents of climate change leads to ices and glaciers melting, increasing in water level, land submergence, and finally amplification of salinity content of both freshwater and salty water (Anis et al., 2019). World Health Organization (WHO) has warned all communities about imminent peril of approximately two-thirds of world's population all over the globe to live in water-stressed condition by the year 2025 (World Health Organization (WHO), 2020). According to the reports, Middle East followed by North America and South America has the highest desalination capacity. The worldwide production capacity of desalinated water is totally 95 million  $m^3/day$  (Jones et al., 2019).

Different processes for water treatment can be divided to primary, secondary, and tertiary treatment. Primary treatment refers to preliminary treatment processes including physically separating of large particles from water (e.g., screening, filtration, centrifugation, separation, sedimentation, coagulation,

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and flocculation) (Das et al., 2014; Gerba & Pepper, 2019), secondary treatment refers to biological treatment of water including aerobic and anaerobic treatment methods (Gupta et al., 2012), and tertiary treatment benefits from advanced treatment options for further reduction in residual turbidity, metals, and pathogens (Gerba & Pepper, 2019) (such as distillation, precipitation, crystallization, membrane technologies, ion exchange, solvent extraction, and so many other advanced oxidations) (Das et al., 2014).

Each technology has its own advantages and disadvantages, but when it comes to choose a technology for water treatment in large scale, economic aspects besides efficiency play crucial roles. With respect to many inherent features such as no need to additives, low thermal energy consumption, and spent media regeneration, high separation efficiency, easy and continuous operation, membrane technology has attracted direct attention of scientists and researchers (Baskar et al., 2019; Mathew et al., 2014).

#### 2 Membrane Technology

Application of membrane technology in various industries was started in 1960s, but the first study of membrane phenomena is reported within the mid-eighteenth century (Fane et al., 2011). A membrane process (Fig. 1) is kind of particular separation in which specific compounds are permitted to pass the membrane at ambient temperature without using further chemical additives (Bazargan et al., 2015; Sadeghi, 2016; Strathmann, 2000). In this process, the filtrate or permeate is the liquid that will be allowed to cross the membrane, and the residual fluid will be called concentrate or retentate. Moreover, the generated dry sludge during filtration is named the coating (Kumar et al., 2013). The first step at successful mass transport through a membrane is the selective absorption of the feed

Fig. 1 Typical membrane process

components into the membrane, and second step is selective diffusion and eventually desorption into the filtrate under low pressure or vacuum condition (Scott, 1995).

Permselectivity is one of the main characteristics of membranes that can be specified by discrepancy between the transport rates of the existence compounds in the solution (Nath, 2017). The membrane's structure, type of the driving force, and the size, chemical nature, and the electrical charge of the particles are the leading factors that can ascertain the transport rate of species across the membrane (Kolmetz et al., 2014). The flux across the membrane is calculated by Eq. 1 (Nath, 2017):

$$Flux = \frac{Membrane permeability}{Membrane thickness} (Driving force)$$
(1)

Permeability is a criterion that represents how tight the membrane is. In porous membranes, water permeability (*A*) is calculated by the Hagen–Poiseuille equation (Eq. 2) (Werber et al., 2016):

$$A = \frac{\varepsilon r_{\rm p}^2}{8\mu\delta_{\rm m}} \tag{2}$$

where  $\varepsilon$  is the surface porosity,  $r_p$  is pore radius,  $\mu$  is viscosity of the aqueous solution, and  $\delta_m$  is the thickness of the active layer of the membrane.

In dense and non-porous membranes, water permeability can be evaluated by solution–diffusion model as is presented in Eq. 3 (Werber et al., 2016):

$$A = \frac{P_{\rm w} V_{\rm w}}{\delta_{\rm m} R_{\rm g} T} \tag{3}$$

where  $P_w$  stands for diffusive water permeability,  $V_w$  is molar volume of water,  $R_g$  stands for the gas constant, and *T* is the absolute temperature.



Based on the type of the applied driving force, membranes can be divided to electrical-potential-driven processes, concentration-gradient-driven processes, temperature-driven processes, and pressure-driven processes (Tofighy & Mohammadi, 2020).

Electrical-potential-driven processes such as electrodialysis, electrophoresis, and membrane electrolysis are used for the removal of charged compounds from a solution or suspension (Kathiresan & Doss, 2020; Mandal & Kulkarni, 2011). The electrical voltage difference drives the ions flux through the electrical field which is generated via cathode and anode electrodes. The uncharged molecules will not be affected by the electrical field, but instead cations will be attracted to the cathode and the anions will be attracted to the anode (Beier, 2014).

In concentration-gradient-driven processes such as dialysis, the fluid fluxes across a membrane due to the existing concentration difference (Youravong & Marthosa, 2017).

In temperature-driven processes such as membrane distillation (MD), the fluid is driven across the permeate and feed side of the membrane due to partial pressure difference which is in turn generated due to temperature gradient (Camacho et al., 2013).

In pressure-driven membranes, filtration occurs through small pores for capturing the pollutants due to the pressure gradient. Depending on the membrane's pore size, the target species, and their polarity, these kinds of membranes can be categorized in four groups including: microfiltration, ultra-filtration, nano-filtration, and reverse osmosis as shown in Fig. 2 (Graff, 2012; Tan & Rodrigue, 2019).

Characteristics of different types of membranes are presented in Table 1 (Belleville & Vaillant, 2016; Nath, 2017).

Beside all the mentioned advantages of the membranes, they still suffer from drawbacks in water treatment applications. In real scales, both high water permeability and selectivity are required for an ideal membrane while the combination of these two features in one membrane is hard to obtain (Park et al., 2017). It is noteworthy that based on the target compounds in some cases (such as desalination), selectivity is more important than permeability (Werber et al., 2016). When using membranes in large scales, membrane fouling is another important problem. For instance, natural organic matters (NOMs) are one of the main agents causing serious fouling and performance aggravation in membranes (Cui & Choo, 2014).

For these reasons, more effective materials are required to be used in the membranes. Previously, polyamide (PA) and cellulose acetate (CA) membranes have been developed for desalination. Results revealed that this system design generates further amount of salts besides process chemicals. In addition, constant contact of the membrane's materials with chlorine causes irreparable damages in membrane over time (Yang et al., 2019). Polymeric membranes exhibit low chemical stability and fouling tolerance (Pendergast & Hoek,



Fig. 2 Efficiency of different types of pressure-driven membranes for materials with wide size range. Reproduced from Graff (2012) with permission from Woodhead Publishing, Elsevier

Driving force	Membrane process and pore radius	Mass transfer mechanism	Permeate	Retentate	Applications
Electrical Potential gradient	Electrodialysis	Ion exchange (Donnan exclusion)	Ionized solutes, water	Non-ionic solutes, water	Separation of ions from water and non-ionic solutes
Concentration gradient	Dialysis membrane extraction	-	Small molecules, water gases, solutes, vapors soluble in the extractant	Large molecules, water components of feed insoluble in extractant	-
Temperature gradient	Membrane distillation	Evaporation/diffusion/ condensation	-	Molecules, <1 nm	Desalination, concentration
Pressure gradient	Microfiltration 0.1–1.0 µ (10– 30 psi)	Convection	Dissolved solutes, water gases ( $\sim \le 1$ nm), and polar vapors	Suspended particles, water gases	Clarification and cold sterilization
	Ultrafiltration microns) 30– 100 psi	Convection	Small molecules, water	Polymers, proteins, micelles, colloids, particulates	Concentration, fractionation of macromolecular solutions
	Nanofiltration 50– 150 psi	Diffusion/convection	Monovalent ions, water	Small molecules, divalent salts	Concentration, purification of small organic compounds, separation of selected salts
	Reverse osmosis (5–10°A) 200– 1000 psi	Solubilization/diffusion	Water, small polar solvents, salts	All solutes, water	Concentration/desalination

Table 1 Different characteristics of membranes with different driving forces (Belleville & Vaillant, 2016; Nath, 2017)

2011). Considering high operational costs, ceramic membranes are suggested for small-scale experiments (Nandi et al., 2008). Moreover, numerous hybrid membranes have been developed and utilized which in turn can cause for high operational costs (Hosseini et al., 2020; Sadeghi et al., 2016).

Producing sufficient clean water that meets the drinking water or irrigation standards is one of the challenging issues worldwide, and thus extensive researches to address this issue lead to benefit from remarkable potentials of nanomaterials (NMs) in membrane's material. The term "nanomaterials" refers to materials with size lower than 100 nm ( $\leq 100$  nm) in at least one dimension (Salisbury et al., 2018).

Hereunto, numerous NMs have been successfully utilized for water remediation goals (Beheshti et al., 2019; Ghenaatgar et al., 2019; Khadir et al., 2020a, 2020b; Khorsandi et al., 2019; Mirjavadi et al., 2019; Mohammadi et al., 2019; Mollahosseini et al., 2019; Piri et al., 2019, 2020). Among countless number of NMs (e.g., zeolites, metal/metal oxide nanoparticles, oxyacids, carbonaceous nanostructures, etc.), carbon nanotubes (CNTs) have exhibited outstanding features which have made them to be of leading materials in desalination and water purification (Ihsanullah, 2019).

#### 3 CNT Materials

CNTs are quasi-one-dimensional allotropes of carbon in a rolled-up graphene sheets with hexagonal structure (Tiwari et al., 2016). These materials contain strong sp<sup>2</sup> hybrid carbon–carbon bonds which is the strongest chemical bond (Paula et al., 2016). CNTs are classified to multi-walled carbon nanotubes (MWCNs) and single-walled carbon nanotubes (SWCNs) (Sielicki et al., 2020). MWCN consisted of multilayers of graphene sheets and was first discovered in 1991 by Prof. Iijima during arcing of graphite. Later he characterized SWCNs in the arc process in 1993 (Hassan et al., 2020). SWCNs consist of a flattish array of benzene molecules with honeycomb-shaped rings of carbon–carbon bonds (Nayfeh, 2018). A schematic presentation of SWCN and MWCN can be seen in Fig. 3.

Different comparable properties of SWCNs and MWCNs such as purity, density, and thermal and electrical-related coefficients are presented in Table 2 (Awad, 2016; Chaudhary et al., 2013; Sattar et al., 2015).

CNT by itself is a hydrophobic material which turned it to an attractive media for water treatment. Hydrophobicity besides capillarity nature causes efficient adsorption and also



Fig. 3 Perspective views of carbon nanotubes: a single-walled carbon nanotubes (SWCNs) and b multi-walled carbon nanotubes (MWCNs)

navigation of sorbates in microporous carbons (Narang & Pundir, 2018).

Owing outstanding features such as large surface area and consequently high adsorption capacity, admissible electro-conductivity, convenient functionalization, feasibility of flowing through the nanotubes without friction, and high chemical, optical, and mechanical resistance, CNTs have become a favorable nomination for the production of innovative membranes in the order of water desalination (without needing to pretreatment or post-treatment) (Suzuki, 2013).

### 3.1 Fabrication of CNT Membranes

Fabrication method has deep effects on performance of CNT membranes. Various fabrication techniques may lead to the production of CNT membranes with different morphologies and separation performance such as pore size and interconnections, porosity, surface charge and rugosity, roughness, and hydrophobicity (Awad, 2016). The concept of using CNT membranes was first raised by Sun and Crooks (2000). Based on the type of morphology, CNTs can be directly grown in three shapes including (Gao & Kono, 2019):

- (I) vertically aligned CNTs (VACNTs)
- (II) transverse or horizontally aligned CNTs (HACNTs)
- (III) mixed matrix membranes (MMMs).

The most applied synthesization methods of CNT are chemical vapor deposition (CVD), laser ablation (LA), arc discharge (AD), and other methods include electrolysis, hydrothermal/sonochemical, and template/bottom-up (Du et al., 2017).

In CVD approach (Fig. 4), cheap catalysts and carbon sources are utilized for fabrication of high yield CNT. At the applied temperature and in the presence of catalyst, the carbon sources decompose to carbon atoms on the surface of a transition metal (He et al., 2015). As a result of the generated interactions, CNTs will grow in the solution. This method in turn consisted of plasma enhanced, injection assisted, thermal CVD, radio frequency, microwave, laser assisted, hot filament, aerogel assisted, water assisted, alcoholic CVD, oxygen assisted, liquid pyrolysis, and solid pyrolysis (Das & Tuhi, 2018).

In LA approach (Fig. 5), there are several determinative parameters including target component, catalyst, light source, pressure, and temperature. In this method in the presence of inert gas and at high temperature, the laser is utilized to vaporize the graphite target and form the CNTs. Besides the main carbon resource (i.e., graphite, fullerene), the suspended carbon atoms in the atmosphere can also play role in the CNT's growth (Das & Tuhi, 2018; Du et al., 2017). Here, the catalyst acts as a cage structure opener of

Property	SWCNs	MWCNs
Graphene layer	Single	Multiple
Bulk synthesis	Multiple	Easy
Purity	Poor	High
Specific gravity (g/cm <sup>3</sup> )	0.8	1.8
Electrical conductivity (S/cm)	$10^2 - 10^6$	10 <sup>3</sup> -10 <sup>5</sup>
Thermal conductivity (W/(m K))	6000	2000
Thermal stability in air (°C)	>600	>600
Temperature coefficient of resistance $(\times 10^{-3}/K)$	<1.1	-1.37
Electron mobility (cm <sup>2</sup> /(V s))	$\sim 10^5$	$10^4 - 10^5$
Maximum current density (A/(cm <sup>2</sup> ))	109	10 <sup>9</sup>

**Table 2** Comparison betweendifferent parameters of SWCNsand MWCNs (Chaudhary et al.,2013; Sattar et al., 2015)



the carbon resource followed by cluster formation. Afterward, the clusters will be used as a stand for CNT's growth. The reaction will continue unless the carbon layer could not be able to accept further evaporated carbon for nanotube growth (Srivastava, 2006).

In AD approach, high DC voltage is exerted to catalyst-containing graphite electrodes. During the test, atmosphere includes helium gas. A micrometer can be utilized for movement of the anode electrode which is connected to the positive pole (Du et al., 2017). A schematic presentation of the AD approach for synthesization of CNTs can be seen in Fig. 6.

Compared with other methods, CVD has several superiorities. The AD and LA methods require high-energy input. Moreover, in CVD approach the size and dimension of the synthesized CNT for fabrication of a high-performance membrane are controllable. So that, dispersion and interface of the nanocarbons in the medium can be enhanced by adjustable factors such as contact time, the partial pressures, temperature, the type of the carbon feedstock and catalyst, the amount of pressure, the type of the atmosphere gas, and injection rate (He et al., 2015). In addition, carbon deposition occurs in high rate and in large synthesis area, and the produced CNTs in this method have high purity, crystallinity, and aspect ratio (Yousefi et al., 2016).

#### 3.1.1 Vertically Aligned CNTs (VACNTs) Membranes

In this method, CNTs are placed perpendicular to the support layer and are reinforced by cumulative van der Waals forces to generate stronger bonds (Li et al., 2017). Application of VACNTs was first reported by Kalra et al. (2003) by using molecular dynamics. The osmotic force-driven flow passes through the membrane due to the salt concentration gradient (Suzuki, 2013). Hinds et al. were the first researchers that reported about mass transport of CNTs in 2004. Their results



Fig. 6 Simple illustration of AD approach for fabrication of CNTs

proved rapid transportation of  $N_2$  gas and ionic ruthenium (III) hexamine (Ru(NH<sub>3</sub>)<sub>6</sub><sup>3+</sup>) solution in VACNTs with inner diameter of 6 nm (Ang et al., 2019).

For proper membrane structure and considering very versatile and tunable options, it is suggested to synthesize the VACNTs via CVD method (Fig. 7). Dense materials such as polystyrene (Lee, 2019), parylene, silicon nitride (Liu et al., 2019), and epoxy (Lee et al., 2015) are the foremost materials in using between CNTs to fill the slots and abate the water leakage. Since CVD is conducted in high temperatures, membrane substrates should be selected among high-temperature-resistant materials (Monea et al., 2019).

Steps for fabrication of VACNT membranes are as follows (Das, 2017): (1) deposition of non-conducting substrates (i.e., SiO<sub>2</sub>/Si) on the substrate, (2) deposition of catalyst on the support by electron beam evaporation, (3) oxidization of the catalytic surface in high temperature, (4) CNT's growth from catalyst in CVD system including Ar, H<sub>2</sub>, or other inert gases, (5) operation of catalyst as a nucleation site for nanotube growth, (6) filling the gaps of CNT by impermeable materials, (7) using reactive ion etching (RIE) at a suitable power for opening the backside substrate, (8) uncovering the nanotube film, (9) reusing RIE or wet chemical etching, etc., to remove excess impermeable material and opening the tips (Golshadi, 2016; Krishnakumar et al., 2012).

Owing high thermal, chemical, and mechanical durability, porous ceramic membranes have been extensively used as support for CNTs (Kayvani Fard et al., 2018).



Fig. 7 Schematic mechanism of water transport and desalination by vertically aligned CNT membranes (VACNTs)

Studies about VACNT membranes have mostly focused on desalination. Trivedi and Alameh developed VACNTs with outer diameter of 8 nm and 300  $\mu$ m length, by CVD method for desalination. A cross section of the synthesized VACNTs obtained by focused ion beam scanning electron microscope (FIB-SEM) is presented in Fig. 8 (Trivedi & Alameh, 2016).

In this study, silicon wafer was stuck on a glass surface and used as mechanical supporting material for VACNTs without any chemical reaction during whole experiments. Then by locating the wafer in a spin coater and adding 50% (w/w) poly(dimethylsiloxane) (PDMS) and xylene, VACNT–PDMS membrane was obtained. To remove volatile part of the PDMS, the membrane is placed in a 100 °C vacuum oven for 6 h. Finally, the silicon support was separated by mechanical peeling (Trivedi & Alameh, 2016).

#### 3.1.2 Transverse or Horizontally Aligned CNTs

Transverse or horizontally aligned CNTs (HACNTs) are synthesized parallel to their flat substrate and have approximately long intertube distances (Bai et al., 2018). The results of primary studies about HACNTs were published in 1998 when researchers succeeded to synthesize individual parallel SWCNTs on a Si substrate (Kong et al., 1998).

The length, aspect ratio, and diameter of HACNTs can be up to centimeters,  $10^{6}$ – $10^{8}$ , and 1–5 nm, respectively (Zhang

Fig. 8 Cross section of VACNT developed by CVD method. The image is captured by using focused ion beam scanning electron microscope (FIB-SEM) device (Trivedi & Alameh, 2016). Adopted from Graff (2012). Copyright © 2016, Springer Nature



et al., 2017). The number of shells (*N*) in a CNT can be evaluated by Eq. 4 (Todri-Sanial, 2016):

$$N = \frac{d_{\rm out} - d_{\rm in}}{S_{\rm a}} \tag{4}$$

where  $d_{out}$  stands for diameter of the outer shell,  $d_{in}$  is the diameter of the inner shell, and  $S_a$  refers to the space between shells. The typical number of shell in a HACNT is 1–5.

Owing longish lengths, infirm intertube interactions and consequently low defect density, HACNTs have exhibited excellent mechanical, electrical, and thermal properties (Li et al., 2016). After successful fabrication of HACNTs, studies focused on their growth mechanism. Numerous processes such as ultralow feeding gas flow guiding (Li & Zhang, 2019), external electrical fields (Morais et al., 2019), and fast-heating CVD (Huang et al., 2004) were developed for fabrication of guiding longish HACNTs. A well-defined HACNT with proper structure can lead to fabrication of multifunctional devices for different applications on a large scales (Neupane & Li, 2011).

Zheng et al., successfully synthesized 4-cm-long horizontally aligned SWCNs (growth rate = 11  $\mu$ m/s) on a Si substrate by catalytic CVD method. They concluded that parameters such as flow rate, type of the atmosphere gas, temperature, the initial concentration of the catalyst solution, and catalyst's size play crucial role when optimizing the experimental conditions (Zheng, 2004).

An et al., reported fabrication of 1-cm-long HACNTs on Si/SiO<sub>2</sub> substrate with 1.7 nm in diameter by CVD growth methodology. They reported notable decrease in length and density of the CNT by decrease in the growth temperature. To achieve a controllable synthesis of CNTs, researchers optimized the growth condition. They studied different catalyst concentration (0.005–0.1 M), pretreatment time (10, 20, and 30 min), and growth temperature (925, 925, 975 °

C). Results proved that increase in solution concentration has dramatically decreased the density and the length of the CNT arrays (Fig. 9). The results also illustrated that higher pretreatment time and higher growth temperature (at lower catalyst concentration) leading to the production of denser and longer CNTs (An et al., 2014).

Besides several advantages, HACNTs suffer from drawbacks such as poor ability to control chirality, bounded lengths, small areal density, and low structural ability to retain homogeneous, which limited their extensive applications in real fields (Shi & Plata, 2018). Thus, the main challenges in this field are to synthesize HACNTs in microscale size with excellent properties and appropriate structures.

Over the past few years, CVD method has been considered as one of the main approaches in HACNT's growth (Fig. 10).

To study growth steps of HACNTs, three classes have been developed including nucleation (on the atomic scale), growth modes (on the molecular scale), and CNT's distribution mechanism for CNT arrays (Page et al., 2010; Sengupta, 2018; Zhang et al., 2013). A brief description of these classes are presented in the following:

- Nucleation: When metallic nanoparticles are used as catalyst for CNT's growth, nucleation will be the prime step. This step in turn can be cleaved into the following steps: dissolution of carbon atoms, supersaturation of the carbon atom following by their precipitation, and finally CNT formation. Unceasing addition of carbon in this stage plays a vital role in CNT's growth (Gomez-Ballesteros et al., 2015).
- Tip-growth mode: Longish CNTs begin from the catalyst region, and during CVD method, catalysts are still on the growing tips of the CNTs. In this stage, getting raised from the substrate surface is the cardinal issue which can

**Fig. 9** Change in the number of distributed CNTs toward their axial orientation versus the length of the CNTs by change in the concentrations of the catalyst. Inside the diagram, the related SEM images for the fabricated CNTs are placed. The length scale of the images is  $100 \mu m$ . Adapted from An et al. (2014) with permission from Elsevier

**Fig. 10** Illustration of water injection and rejection mechanism in horizontally aligned CNT membranes (HACNTs)



Clean water

occur due to the temperature gradient (thermal buoyancy perpendicular to the substrate) among gas flow and the substrate (Fig. 11) (Zhang et al., 2014). Poor interaction of the nanocatalyst and substrate surface can be another reason for this phenomenon (Gohier et al., 2008). In the following, the CNTs will be buoyed in the gas flow, and nanocatalysts will stay at the tip ends of these CNTs to catalyze the CNT's growth uninterruptedly (Zhang et al., 2014).

3. **Base-growth mode**: Unlike the tip-growth mode, at the end of CNT's growth, the nanocatalyst will stay at the bottom of the CNT (Fig. 12). The probable reason for occurrence of this mode is due to the powerful interaction of the particles and substrate's surface. Thus, carbon precipitates from the top surface of the catalyst, and the nanotubes keep growing from the catalyst region (Sengupta, 2018).

4. Schulz–Flory distribution mechanism for CNT arrays: Schulz–Flory is a probability distribution theory which can be defined as the linear polymer's ratio with different molecular weight and length in a polymerization process (Helfferich, 2004). Preconditions of this theory are as follows: equivalent reactivity of the carbon resource molecules, controllability of the process by kinetic, and existence of a steady growth media (Zhang et al., 2013). Forasmuch as the screw-like dislocation theory, HACNTs can be considered as linear carbon polymer. Based on this theory, shorter polymers have



higher proportions than the lengthy ones (Zhang et al., 2011). Observations of the constant growth rate of horizontally aligned CNTs proved that the growth progress of these materials is being controlled by kinetic (Zhu et al., 2019).

#### 3.1.3 Mixed Matrix Membranes

Mixed matrix membranes (MMMs) are mix of solid and/or liquid NMs that the top layer is blended with polymers, porous materials (activated carbon, CNTs, etc.), or nonporous material (SiO<sub>2</sub>, TiO<sub>2</sub>, and fullerene) (Roy & Singha, 2017). MMMs benefit outstanding features of both ceramic and polymeric materials (including physicochemical stability and easiness in fabrication, respectively) to enhance efficiency of water pollution abatement by improving selectivity, penetrance, tortuosity, fouling diminution, and sufficient removal of the target compounds (Qadir et al., 2017). In addition, this technology provides higher hydrophilicity, permselectivity, and resistance against pH and temperature changes (Maghami & Abdelrasoul, 2018). Application of NMs in this technology introduces antibacterial and photocatalytic characteristics to the membrane as well. A schematic illustration of this technology can be seen in Fig. 13.

Besides all the advantages, it is noteworthy that utilizing nanoparticles are limited due to the agglomeration feature of these materials, weak adhesion to the polymeric matrix support, and consequently weak mechanical stability (Fu et al., 2019). Therefore, several fabrication methods have been developed for efficient application of nanoparticles on the surface of the membranes such as self-assembly deposition (Abdelrasoul et al., 2017), layer-by-layer deposition (LbL) (Chimisso et al., 2020), chemical grafting (Ursino et al., 2018), and physical and chemical deposition of NPs (e.g., dip coating, spin coating, hot pressing, etc.) (Xiao et al., 2016).

There is no specific phenomenon for categorization of MMMs. In some of the recent studies, structure and the position of NMs (filler) were of the notable factors in classification of MMMs (Fig. 14) as follows: (1) conventional nanocomposites, (2) thin-film nanocomposites (TFN), (3) thin-film composite (TFC) along with nanocomposite substrate, and (4) nanocomposites on the surface of the membranes (Yin & Deng, 2015).

The main steps for fabrication of MMMs are providing a homogenous solution of NMs and polymer. This goal can be achieved by three different approaches as follows (Esfahani et al., 2019):



Clean water

Fig. 13 Schematic presentation of water injection and desalination mechanism by mixed matrix CNT membrane (MMMs)



Fig. 14 Categorization of different types of mixed matrix membranes

- 1. Dissolution of polymers in the solvent and stirring for a given time to form a homogeneous polymeric solution and then adding predestined amount of NMs to the solvent and stirring for a given time.
- 2. Adding predestined amount of NMs to solvent and stirring for a given duration to form a homogeneous suspension and finally adding polymer.
- 3. Adding predestined amount of NMs in the solvent and stirring for a certain duration, then adding polymer into

the solvent in a separate container, and finally mixing NMs solution and polymer solution.

To enhance the dispersion of nanofillers (especially for hydrophobic NMs such as CNTs) and application of these materials in MMMs membranes, two cardinal factors should be considered. The first one is the ability to scatter the CNTs homogeneously in the entire matrix, and the second one is the adaptability of CNTs and the matrix (Sianipar et al., 2017).

#### 4 Application of CNT Membranes in Desalination

Fabrication of CNT nanocomposite membranes for desalination of sodium hydroxide was first reported by Shawky et al. (2011). Nowadays, CNT membranes are widely utilized as vital materials for water treatment, especially in desalination because of their rapid transportation ability (Ma et al., 2017). The water permeability of a CNT membrane can reach up to 30 thousands liters per square meter per hour (LMH/bar) (Lee et al., 2015). CNTs could decrease the size and construction expenses of membrane desalination plants up to 50% (Humplik & Lee, 2011) and lead to production of desalinated water with equal expenses to conventional water treatment methods (World Bank, 2019).

The International Desalination Association (IDA) is the precedent society in the world which has concentrated particularly on the development of desalination industries and water recycle technologies. In the annual reports of IDA in 2019, it is listed that desalination is being practiced in 174 countries all over the globe and over 17,000 plants have been contracted. Over 300 million people are dependent upon desalinated water for whole or part of their daily consumption. The cumulative installed desalination capacity and reuse capacity was 107 and 146 million m<sup>3</sup>/day, respectively (The International Desalination Association (IDA), 2020). Desalination capacity refers to the volume of high-quality produced water for utilization of human.

Although CNT membranes are not yet commercially available for desalination, it is estimated that they will be available for large-scale utilization within in the near future (likely next decade) (World Bank, 2019). Scientists have monitored and forecasted annual desalination capacity (including brackish water and seawater) versus municipal wastewater reuse capacity between 1990 and 2030 (Fig. 15) (Sanz, 2019). The results revealed that the annual desalination capacity is growing and evolving at the same rate as wastewater reuse capacity.

Concentration of total dissolved solids (TDS) can be used as an agenda for classification of feed water (Table 3) (Hanasaki et al., 2016). The worldwide installed desalination capacity for different feed water types and their contribution percentage compared with each other is presented in Fig. 16. The pure water is mainly utilized by industries which require high-quality water (i.e., food and drug industries) (Khan & Ali, 2018).

Both SWCNT and MWCNT have been used extensively in membranes for different feed water's desalination.

Kim et al. fabricated polyamide-RO membrane with 0.2 g CNTs (PA-CNT) by varying values of required acid solution (AS) (20-100 mL), temperature (25-105 °C), and contact time (3–5 h) and compared their efficiency with non-CNT-containing membrane (PA). Six different membranes fabricated in different conditions include PA-CNT1 (a bought pristine membrane), PA-CNT2 (T = 25 °C, t = 3 h, AS = 20 mL), PA-CNT3 (T = 45 °C, t = 3.5 h, AS = 40 mL), PA-CNT4 (T = 65 °C, t = 4 h, AS = 60 mL), PA-CNT5 (T = 85 °C, t = 4.5 h, AS = 80 mL), and PA-CNT6 (T = 105 °C, t = 5 h, AS = 100 mL). Applied pressure is kept at 15.5 bar, the feed solution contained 2000 mg/L NaCl, and the cross-flow velocity was 700 mL/min. The water flux and salt rejection efficiency of the fabricated membranes are presented in Fig. 17. As can be seen, all the PA-CNT membranes exhibited higher water flux than PA membrane (36.4 LMH). Furthermore, PA-CNT4 was the only membrane that showed higher salt rejection and similar water flux compared to PA membrane. The reason can be attributed to the well dispersion of CNTs in the solution (Kim et al., 2014).

Scrutinies of transmission electron microscopy (TEM) images (Fig. 18) proved that PA-CNT1, PA-CNT2, and PA-CNT3 have had a non-dispersed or entangled morphology. PA-CNT4 and PA-CNT5 had a well-dispersed CNT structures with smaller tubes. PA-CNT6 has had a non-tube structure with small spots consisting of debris of carbon materials. Existence of aggregated morphology could

Fig. 15 Annual report about the capacity of desalinated water versus municipal wastewater reuse during 1990–2030. Reproduced from Sanz (2019) (accessed date: 13 May 2020)



**Table 3**Classification ofdifferent feed water types(Hanasaki et al., 2016)

Feed water type	Total dissolved solids (ppm)
Pure water	<500
River water	500-3000
Brackish water	3000-20,000
Seawater	20,000-50,000
Brine	>50,000
Wastewater	Unknown





**Fig. 17** Water flux and salt rejection of fabricated membranes under various conditions  $(C_{\text{NaCl}} = 2000 \text{ mg/L}, P = 15.5 \text{ bar})$ . Adapted from Kim et al. (2014). Copyright © 2014, American Chemical Soci-ety (ACS)




Fig. 18 TEM images of fabricated CNTs (scale bar: 200 nm): a, b PA-CNT1, c PA-CNT2, d PA-CNT3, e PA-CNT4, f PA-CNT5, and h, g PA-CNT6. Adapted from Kim et al. (2014). Copyright © 2014, American Chemical Society (ACS)

lead to the reduction in salt rejection values (Kim et al., 2014).

Ragunath et al. (2018) fabricated CNT-immobilized membranes (CNIM) via phase inversion technology in the presence of different amount of polyvinylidene fluoride (PVDF) to utilize for desalination. In this study, concentration of CNT was 0.01 wt%, the filtrate temperature was kept in 15-20 °C range, and feed and permeate flow rates were equal to 150 mL/min. Results indicated that water flux in CNIM (which contained 0.01 wt% PVDF) enhanced from  $31.4 \text{ L/m}^2\text{h}$  to  $51.4 \text{ L/m}^2\text{h}$  when temperature is raised from 60 to 80 °C, respectively. The reason can be ascribed to adsorption enhancement and quick desorption arising from outstanding properties of CNTs. At the same condition by increasing PVDF content to 0.03 wt%, the permeate flux reduced to 27.9 and 45.01 L/m<sup>2</sup>h. This can be attributed to change in membrane's morphology and decrement of active sites for permeate flux. Moreover, water vapor flux was extremely affected by feed concentration in such a way that when feed concentration increased from 0 to 35,000 ppm, the flux decreased from 33.8 to 30.8 L/m<sup>2</sup>h, respectively (Ragunath et al., 2018).

Sun et al. (2019) constructed a CNT forest (CNTF) on a porous electrospun silica fiber layer with high durability against wetting for desalination of 1 M NaCl. An electrospun polyvinyl alcohol (PVA)–silica fiber mat (SFM) was utilized for the production of a porous fiber substrate. In this study, CVD method was carried out for the production of membranes with different orientations (vertical/horizontal) and adjustable lengths. By increasing catalyst dosage (FeCl<sub>3</sub>) from 0.1 to 1.0 mol/L, the height of vertically aligned CNTF was increased up to hundreds of nanometers, which can be ascribed to the formation of a squatty catalyst layer. Water flux of the ultrahigh VCNTF@SFM membrane was approximately  $17.31 \pm 2.3$  L/m<sup>2</sup>h at a temperature of 40 °C. By difference adding 1000 mg/L polyvinylpyrrolidone (PVP) to the stock solution, horizontally aligned CNTF was generated which could be due to a powerful fiber-surface adherence force. In various temperature differences (10, 20, 30, and 40 °C), horizontally aligned CNTF represented recognizable water fluxes equal to  $1.52 \pm 0.5, 2.73 \pm 1.0, 4.08 \pm 0.9$ , and  $8.42 \pm 1.5$  L/m<sup>2</sup>h, respectively. In all membranes, approximately complete (>99.9%) salt rejection was achieved at various temperature differences (Sun et al., 2019).

Anga et al. (2018) investigated the effect of different CNT sizes on the desalination of 1.2 M salt through slit confinements formed by HACNTs. Figure 19a depicts that increasing in membrane thickness can increase water flow. This deduction is seemingly a controversy as it is claimed in several studies (Li et al., 2017; Suk & Aluru, 2010) that the shape of nanochannels (instead of membrane thickness) plays a crucial role in distinguishing water flow. Salt rejection efficiency (Fig. 19b) has always remained 90%. This parameter measured when 80% of feed water was discharged. In additional, by considering membrane area, tubes with small diameter exhibited higher permeability than greater ones. The result can be attributed to efficient transition effect of CNT's curvatures (Ang et al., 2018).

In another study conducted by Li et al. (2019), outer wall VACNTs were utilized as membranes in RO technology for desalination of 2000 mg/L NaCl. Water flux and salt rejection of the membrane at 15.5 bar were equal to 128.6 LMH and 98.3%, respectively. Ultrahigh porosity of VACNT and also establishment of a thin PA layer on the support layer caused outstanding functionality of VACNT-RO membrane. Researchers also investigated the effect of repetitious deposition of PA layers on the functionality of PA/outer wall VACNTs membrane and concluded that increasing in deposition cycle (from 1 to 17) could decrease water flux (from 875.8  $\pm$  150.3 to 58.9  $\pm$  6.2 LMH) and increase salt rejection (from 94.9 to 98.8%) (Fig. 20) (Li et al., 2019).

Dong et al. (2018) represented fabrication of a novel superporous, superhydrophobic, and thermally resistant ceramic CNT by MD process. This membrane exhibited significant desalination potential (99.9% of Na<sup>+</sup>), water flux (37.1 L/m<sup>2</sup>h), and distillate conductivity (0.09  $\mu$ S/cm) which could be due to high surface porosity (79.1–81.1%) and superhydrophobicity by providing excellent liquid–gas interface. The wall thickness and diameter of the MWCNTs were approximately 10 and 40–50 nm, respectively (Dong et al., 2018).

Ang et al. (2019) investigated desalination efficiency of 1 M NaCl rejection by modeling transverse flow CNT membrane (TFCNT) under oscillating pressure condition. For this purpose, molecular dynamics and large-scale atomic/molecular massively parallel simulator (LAMMPS) were used. In this study, mean applied pressure ( $\Delta P_0$ ), amplitude (*A*), and period (*T*) were kept at about 136 MPa, 0.025–1.25 kcal/mol-Å, and 0.02–0.08 ns, respectively. Moreover, three different slit sizes (4.28, 5.28, and 6.28 Å) were considered. Results of water

**Fig. 19** Efficiency of CNT membranes with different sizes in desalination: **a** water flow and **b** salt rejection. Adapted from Ang et al. (2018) with permission from The Royal Society of Chemistry



**Fig. 20** Functionality of outer wall VACNT-RO membrane in terms of water flux and salt rejection based on repetitious deposition of PA (input NaCl = 2000 mg/L, operating pressure = 15.5 bar). Adapted from Li et al. (2019) with permission from Elsevier

**Fig. 21** Efficiency of TFCNT membrane in terms of water permeability and desalination  $(\Delta P_0 = 136 \text{ MPa})$ . Adapted from Ang et al. (2019b), with permission from Elsevier



permeability and desalination efficiency are illustrated in Fig. 21. As it is obvious, 3.28 Å was the critical slit size (largest size) that led to complete desalination efficiency (Ang et al., 2019).

Results demonstrated that in the period of 60,000 fs and 0.075 kcal/mol-Å amplitude, sinusoidal pressure can enhance water permeability of the membrane (CNT diameter = 6.8 Å) by 16% and bring complete salt rejection within 0.02 to 0.1 ns. This can be ascribed to the fact that at higher terms of fluctuation, resistance against reverse flow could improve. Furthermore, additional mixing of the salt caused

ions to return into the feed water and consequently decreasing the concentration polarization effects at the feed side of the membrane wall (Ang et al., 2019).

Countless studies have been reported about the application of CNT membranes for rejection of common types of salt ions. A brief summary of some of these studies is given in Table 4.

All these studies proved reliability and excellent performance of CNT nanotubes in desalination. Different properties of CNT-based and conventional membranes are compared in Table 5 (Ali et al., 2019).

Rejected ion type	Pore size (nm)	Efficiency (%)	References
Na <sup>+</sup>	0.32	100	Corry (2008)
	0.4	100	Song and Corry (2009)
	0.8	100	Jia et al. (2010)
	1.5	98	Chan et al. (2013)
Cl	0.49	100	Corry (2008)
	0.69	84	Goldsmith and Martens (2010)
	0.8	90	Ratto et al. (2011)
	1.1	86	Corry (2011)
K <sup>+</sup>	0.5	99	Song and Corry (2009)
	0.4	100	Song and Corry (2009)
	1.6	40–60	Fornasiero et al. (2008)

 Table 4
 Operation of CNT membranes in separation of various salt ions

Table 5 Characterization of different membranes and superiority of CNT membranes (Ali et al., 2019)

Items	Membrane							
	CNT-based	Reverse osmosis	Nanofiltration	Ultrafiltration	Ultrafiltration			
Application in water treatment	Water and wastewater treatment, desalination, contaminant separation	Water purification and reuse, desalination, production of deionized water	Softening, removal of hydrated ions, and NOMs	Virus rejection and colloidal particles separation	Protozoa and bacteria rejection of, suspended solids separation			
Required materials	CNTs, polymers, ceramics	Organic polymers (e.g., polyamide, polysulfone, and polyether sulfone)	Organic polymers (e.g., polyamide, polyester, and other porous polymers)	Polysulfone, acrylic, cellulose, and etc.	Polypropylene, polysulfone, polyurethane, etc.			
Pore sizes	Ca. 0.8–100 nm	Non-porous	<2 nm	2–50 nm	50–500 nm			
Water permeability	$\sim$ 7 × 10 <sup>-7</sup> L/MPa s	$\sim$ 3 × 10 <sup>-12</sup> L/MPa s	$\sim$ 4 × 10 <sup>-11</sup> L/MPa s	$\sim$ 5 × 10 <sup>-11</sup> L/MPa s	-			
Applied pressure	Varied with type of application	30–60 bar	20–40 bar	1–10 bar	<1.0 bar			
Features	<ul> <li>Less energy consumption</li> <li>Good performance</li> <li>Tolerant against harsh environmental condition</li> <li>High durability, fouling resistance, and cost-effective</li> </ul>	<ul> <li>High-energy consumption</li> <li>Good performance</li> <li>Less tolerance in harsh environmental condition</li> <li>High durability, fouling resistance, and cost-effective</li> </ul>	<ul> <li>High-energy consumption</li> <li>Good performance</li> <li>Less tolerance in harsh environmental condition</li> <li>Less durability, prone to fouling, and less cost-effective</li> </ul>	<ul> <li>Moderate energy consumption</li> <li>Moderate performance</li> <li>Less tolerance in harsh environmental condition</li> <li>Less durability, prone to fouling, and less cost-effective</li> </ul>	<ul> <li>Moderate energy consumption</li> <li>Moderate performance</li> <li>Less tolerance in harsh environmental condition</li> <li>Less durability, prone to fouling resistance, and less cost-effective</li> </ul>			

#### 5 Challenges, Perspective, and Future Direction

Numerous studies have emphasized that nanotechnology specifically CNTs have presented privileged potential in filtration applications (e.g., desalination) (Tlili & Alkanhal, 2019). The NMs propound principal profits by providing adjustable pore size, high permeability and thermal durability, targeted functionalization, and being core of subsidiary physicochemical interactions, e.g., improving hydrophobicity and consequently providing boosted selectivity besides water flux (Bhadra & Mitra, 2014). Nevertheless, commercialization of these compounds has limited by factors such as cost and toxicity. In order to transform this technology to a cost-effective and safe method, amending retention and recoverability of the NMs should be focused (Roy et al., 2020). Moreover, several other challenges should be considered in balancing parameters including NMs functionalization and fabrication, mechanical resistance, generation of homogenous dispersion of inorganic NMs in the polymeric matrix, and preventing agglomeration. Above all, health issues (such as exposure of workers while producing NMs and membranes) and finally leaching of NMs out of membranes (due to the lack of filler's stability inside the membrane) should be perceived before scaling up the membranes (Bhadra & Mitra, 2014; Esfahani et al., 2019). One of the most important issues in opting for fillers is that one's ability should not be immolated for boosting another sufficiency.

NM-based membranes especially CNT membranes offer up to 20% more productivity, or the same productivity but with 15% lesser energy consumption compared with conventional membranes. If carbon nanotubes with much higher productivity can be developed, then this could slash desalination costs to the level of conventional water treatment technologies within a decade (World Bank, 2019). According to the above-mentioned items, much efforts and optimizations are still needed to be conducted to improve antifouling and/or biofouling properties of membranes and move this technology forward to transpire into the desalination markets. To overcome this problem, several unsolved questions should be answered. For instance, how can NMs play role in fouling attenuation? What are the main interfacial interactions in adhesion of foulants on the both inside and outside surface of CNT membranes?

#### 6 Conclusion

1. Increasing need of humans to freshwater has turned application of nanotechnology to a promising approach in water treatment. Among numerous NMs, CNT membranes as a new emerging field in desalination have gained attention of researchers due to their outstanding features.

- 2. An in-depth review of the development procedure of CNT membranes proved that physicochemical modifications of CNTs and application of various fillers, ceramics, polymers, and nanoparticles via several techniques have successfully enhanced the maximum desalination efficiency of the CNT membranes.
- 3. Despite extensive studies, there are still problems with CNT membranes which demand prompt solution before entering desalination markets. Challenges with cases such as functionalization and fabrication of these membranes, homogenous dispersion of NMs inside the matrix, type of filler, and health issues when producing membranes should be addressed when investigating application of these materials in large scales.
- 4. High water permeability, hydrophilicity, permselectivity, and durability of CNT membrane against harsh environmental conditions could reduce the size and construction expenses of membrane desalination plants.
- 5. Studies have anticipated that the annual desalination capacity will evolve at the same rate as wastewater reuse capacity in the very near future.

**Conflicts of Interest** The authors declare that they have no conflict of interest.

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# Integrated Desalination Systems Coupled with Nuclear Reactors

K. V. Narayana Saibaba

#### Abstract

Water is the primary requirement for the survival of life on earth and is a critical parameter in sustainable development. Continuous supply of water and energy to the domestic and industrial needs determines the nations' economic development; however, freshwater supply is minimal. Freshwater resources have been exhausting rapidly due to the increase of population, together with urbanisation and unplanned industrial growth. The exhaust of freshwater resources led to the water crisis in many regions of the world, which may be extended to other regions in the coming future. This crisis necessitates the exploration of unconventional water production methods. Desalination is a promising method that can be used for sustainable water production; however, desalination is an energy-intensive process. The combustion of fossil fuels for energy generation releases large quantities of greenhouse gases into the environment and disturbs the natural ecosystem. Nuclear reactors can be used for clean and eco-friendly energy production. For sustainable water and energy supply, nuclear power plants can be coupled with both reverse osmosis desalination plants and thermal desalination plants. Hybrid nuclear desalination systems provide the reduction of water cost and produce high-quality water and offer a safe, economic and sustainable supply of energy and water and meet global water demand.

#### Keywords

Desalination • Nuclear energy • Desalination coupled with nuclear reactors • Multistage desalination • Multi-effect desalination • Reverse osmosis

#### 1 Introduction

Water is essential for the existence of human beings and other habitats on earth. Rapid industrialisation and urbanisation and the population driving the fast depletion of clean water resources, thereby potable water demand is growing rapidly. Many countries throughout the globe are facing water scarcity, and it is believed that countries will fight for water resources in the future. It is estimated that safe drinking water is accessible to only 20% of the world's population. The freshwater shortfall in the world is 230 billion m<sup>3</sup> per annum in 2002 and is estimated to go beyond 2000 billion m<sup>3</sup> per year by the end of 2025 (WWAP (United Nations World Water Assessment Programme), 2015). To minimise stress being put on natural freshwater resources, freshwater must be produced by alternative methods. Due to the fast depletion of water resources, it is required to find alternative renewable resources for water production. Most parts of the earth are surrounded by seawater, and researchers are working towards developing novel technologies for converting seawater into potable water. Desalination is the best option for converting seawater into usable water. Desalination technology is being used in many countries for more than 5 decades. However, its applicability on a large scale is much lower than the anticipated values due to its high energy requirements and high production cost.

#### 1.1 Desalination

Seawater desalination by distillation has been in practice since the early 1800s; however, its use has been increased from 1960 onwards with the application of the multistage flash distillation process. The total desalination capacity touched almost 99.8  $\times$  10<sup>6</sup> metre cube per day (m<sup>3</sup>/d) in 2017. The total number of desalination plants worldwide has reached 19.372 in 2017 (TOPSFIELD, 2017). The major

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problems associated with desalination plants are the disposal of highly concentrated brine left as waste into the ocean and high energy requirements for electricity and heating purposes. Fossil fuels are generally used for this purpose and impacts the environment very badly by releasing hydrocarbons. For this reason, the use of nuclear energy in desalination plants is gaining importance. Nuclear reactors, coupled with desalination plants, offer significant improvements in reducing pollution to the environment and saving non-renewable energy for future generations. The technological advancements in this sector resulting in less environmental pollution (such as noise, air pollution, etc.) and safe potable water with safe plant operation. Research in the development and use of novel desalination methods has been accelerated the growth in constructing desalination plants throughout the world for the past 30 years (Esmaeilion, 2020; Sanza et al., 2007; Sauvet-Goichon, 2007). Important developments and research works carried out in the nuclear desalination processes are represented in Table 1.

#### 1.2 Nuclear Energy

Nuclear reactors have been in use for the production of nuclear energy generation for the past five decades. Nuclear energy production was directed to meet industrial heating processes' energy requirements, domestic electrical supply and heating purposes from the early 1960s. Nuclear energy production is developing very rapidly throughout the globe due to the fast exhaust of natural resources. Since it is possible to produce energy from nuclear reactors in large quantities, International Atomic Energy Agency (IAEA) initiated a program in 1989 and directed all the member countries to evaluate the applicability of energy generated from nuclear plants for the desalination of seawater. This program's results revealed that the use of nuclear energy for the desalination process is technically viable and economically competitive. Based on these studies, IAEA framed guidelines, technical documents and safety manuals and provided the required support to member countries for the desalination process for producing water using nuclear energy.

Nuclear energy can be used for the supply of electricity as well as water. The assessments performed by the IAEA along with different nations on the applicability of electric energy produced from the nuclear plants for desalination of seawater proved to be technically feasible and economically competitive with non-renewable energy resources. Both nuclear energy and desalination technologies are being used over the past five decades; these technologies are proven, and hence, combing these two can be useful to society in different ways. Nuclear reactors and desalination plants are combined to meet the high energy requirements in desalination processes. Integrated desalination units, coupled with nuclear reactors, are gaining importance. It is expected that the growth of hybrid technology is very high in the near future. Numbers of suppliers are coming up with novel hybrid technologies (Megahed, 2003).

#### 1.3 Common Types of Reactors Used for Electricity and Desalination

Nuclear energy is an extremely reliable and stable form of energy. Nuclear power has been used for electrical power generation for the past seven decades. Russia is the first country to design a nuclear reactor for electricity generation. It constructed a 5 MW(e) power reactor in the year 1954 at Obninsk town. However, the United States of America was the first to design a commercial nuclear power plant, which was operational since 1957. This nuclear reactor was a pressurised water reactor (PWR) type, Westinghouse Nuclear Power Division designs capacity of 90 MW(e). United States of America was also the first to construct the boing water reactor (BWR) with 200 MW(e) capacity. In the year 1957, United Kingdom constructed the gas-cooled reactor (GCR) for the first time in the history with 50 MW(e) capacity. Pressurised heavy water reactor (PHWR) of 22 MW(e) was constructed in Canada in 1962. The largest nuclear power plant is situated in France; it produces as much as 70% of total electricity generation. France-based company Areva, Russia-based company Rosatom and General Electric-Hitachi alliance are in the top three list of nuclear energy makers. There are 440 operable power reactors of various types (mostly pressurised reactors) in the world till April 2020. These reactors are producing 390,000 MW(e) power. In addition to this, nearly 55 reactors with 63,000 MW capacity are in the erection stage, and some are in the planning stage with a combined electrical capacity of 118,000 MW. It is reported that 17% of the total world's electric energy is produced from nuclear technology (Al-Mutaz, 2003).

All types of nuclear reactors currently in use produce heat energy. This can be used directly for heating purposes specific to that plant or converted first into heat, then into electricity through a steam turbine generator. Some of these nuclear reactors were designed to produce both heat and electricity to meet in-house and nearby domestic needs. Different types of reactors are in use; they are water reactors, liquid metal cooled reactors and gas-cooled reactors. However, the most widely used reactor for the desalination process is the water reactor. In a water reactor, ordinary water can be used, whereas heavy water is used in pressurised water reactors. These reactors may be of heat-only reactors, specifically designed for producing heat for thermal desalination processes, or these may be of co-generation reactors

Table 1	Important
developm	ents and research works
in nuclea	r desalination

S. no.	Nature of work	References
1	Energy cost for physical desalination processes	Rognoni et al. (2011)
2	Environmental effects of concentrated brine discharge to the sea from coastal desalination plants	Clark et al. (2018)
3	Environmental impact assessment	Liu et al. (2018)
4	New technologies for nuclear desalination process	IAEA Tecdoc 1753 (IAEA (International Atomic Energy Agency), 2015)
5	Economic analysis on nuclear desalination	Tian et al. (2003)
6	Safety assessment in nuclear desalination plant	Masriera and Doval (2006)
7	Green energy from seawater desalination	Misra and Khamis (2009)
8	Technical and economic characteristics of shore-based nuclear desalination plant	Dragunov et al. (2005)
8	Environmental issues related to nuclear desalination	Anastasov and Khamis (2010)
9	Water supply from nuclear desalination	Aleksandrov et al. (2018)
10	Comparison of MED and RO nuclear desalination plants	Golkar et al. (2017)
11	Current status and future developments in nuclear power industry	Pioro et al. (2019)
12	Current status and future perspectives of nuclear desalination in Iraq	Ghazaie et al. (2019)
13	Study of various nuclear power-desalination coupling options	Dewita et al. (2019)
14	Water and power costs of various nuclear desalination coupling units	Elaskary (2013a)
15	Economic and safety aspects in nuclear desalination plants	Bouaichaoui et al. (2012)
16	Assessment of impact of nuclear desalination water shortage problems in china	Avrin et al. (2015)
17	Economic feasibility of Integration of high temperature reactor with osmosis process	Park and Kim (2013)
18	Studies on various options of coupling of reactors with desalination plants	Jung et al. (2014)
19	Economic aspects of different nuclear desalination units	Nisan and Dardour (2007)
20	Economic assessment of the possible desalination processes for the first unit of Bushehr nuclear power plant	Sadeghi et al. (2020)
21	Comparative economic assessment was performed on different nuclear desalination processes	Gadallah and El-Fawal (2009)

designed to produce both heat and electricity. Liquid metal cooled and gas-cooled reactors are used for applications where high temperatures are required.

#### 2 Nuclear Reactors Coupling with Desalination Plants

Fossil fuels were used for the generation of heat energy in the desalination process. Since these fuels are limited and must be used for other essential uses, using fossil fuels for desalination is not recommended and is not compatible with sustainable development over the long term. Moreover, the burning of fossil fuels generates large quantities of greenhouse gases and other toxic carbon emissions and dictates the use of alternative methods for desalination. In the Mediterranean region itself, it is required to produce 10 million  $m^3/d$  of additional water by the end of 2020 to overcome the freshwater shortfall. If fossil fuels were used for this purpose, then additionally 20 million tons of carbon dioxide, 200 kilotons of sulphur dioxide, 60 kilotons of nitrogen oxides and 16 kilotons of other hydrocarbons

would be entered into the environment per year (Nisan & Dardour, 2007). Considering these issues, the International Atomic Energy Association is promoting the use of nuclear energy as an alternative to fossil fuels for the desalination process. It is also providing technical assistance to associated countries in the form of publishing updated guidelines, design books and manuals on construction, operation and maintenance (IAEA, 1996, 1997a, 1997b, 1997c, 2005, 2000a, 2000b, 2000c, 2002a, 2002b). Using guidelines from the IAEA, countries like Russia, the United States, India and Japan are producing electrical energy and drinking water through these coupling technologies.

A sustainable supply of energy and water to humankind has become a big challenge due to the rapid population growth and a drastic decrease in fossil fuels throughout the globe. It was realised globally that sustainable electricity supply and water supply could be achieved with the help of nuclear power generation. For the last two decades, many countries are showing interest in using nuclear power as an alternative to fossil fuels in desalination plants. It is reported that the cost of water production has been reduced to a range of 0.4–1.8 USD/m<sup>3</sup> for various types of reactor–desalination combinations. It will further reduce due to the advancement of coupling technologies (Al-Othman et al., 2019). Implementing nuclear energy for the desalination process depends on two important factors, such as reactor and fuel used. Many countries have started R&D studies and started pilot plants for desalination using nuclear energy. Many technical, economic and safety considerations studies were conducted on the viability of using these coupling technologies (Elaskary, 2013b; IAEA, 1992, 2000a, 2001, 2008; Khalid et al., 2016; Sadeghi et al., 2020; World Nuclear Association, 2017).

#### 2.1 Nuclear Reactors—Desalination Plants Coupling Technologies

Coupling of a nuclear reactor with desalination plant technologies mainly classified into two types. They are the integration of a nuclear reactor with thermal desalination processes and integration with mechanical desalination processes (Elaskary, 2013a). Multistage flash distillation (MSF) or multi-effect distillation (MSD) comes under thermal desalination processes, whereas reverse osmosis (RO) and vapour compression (VC) processes come under mechanical desalination processes. Multistage flash distillation method is the most widely used for freshwater production through the thermal desalination process. In multistage flash distillation methods, feedwater passes through a series of stages where temperature and pressures are maintained at lower values than the previous stage. A Typical multistage flash distillation plant contains 15–28 stages (Voutchkov, 2012; Esmaeilion, 2020). The thermal energy generated from a nuclear reactor coupled with thermal desalination (MSF or MSD) processes is mainly used for the desalination process's heating purposes. However, very little electrical energy is required for operating pumps and other process equipment in the plant. The generated thermal energy is primarily used for steam production and other heat transfer processes. However, energy generated from nuclear reactors coupled with mechanical desalination (RO or VC) processes is mainly used for the desalination process's electrical heating purposes.

Nuclear Reactor-RO Desalination Plant: Desalination using reverse osmosis is a very important method for the production of water. The growth in usage of the RO method has been increasing due to the developments in membrane technology (Widiasa et al., 2009). The reverse osmosis process works exactly opposite to the osmosis, process where water diffuses from a dilute solution to the concentrated solution under a natural driving force. In the reverse osmosis method, water is diffused through the membrane from a high concentration region to a low concentration region under the influence of high pressure. RO process requires large amounts of electrical energy for maintaining a high-pressure gradient with the high capacity pressure pumps. The electric power generated from the nuclear reactor can be used to supply energy to operate high-pressure pumps in RO desalination plants. Integration of these two plants, i.e. nuclear energy with reverse osmosis, is not complex and, moreover, easy to operate. It can be used for small as well as middle range plants. Nuclear energy produced in power plants is utilised to raise the operating temperatures of the RO desalination plant, which in turn facilitates the mass transfer flux and increases the desalting rate. Due to this, the overall efficiency of the plant improves. The integration of nuclear energy with a reverse osmosis plant requires no special arrangements except the connection to the electric grid, and hence, it involves no additional costs. Therefore, these plants can be located in different places.

**Nuclear Reactor—Thermal Desalination Plant**: Thermal desalination plants require temperatures up to 130 °C or lower. These temperatures can be easily generated through nuclear energy, and hence, this type of integration of nuclear energy plant and thermal desalination plant offers the optimum results. Coupling of nuclear heat reactors with thermal desalination process requires that both plants be located in the same place to reduce cost and heat losses during transportation to long distances.

**Table 2**Comparison betweenMSF, MED and RO desalinationplants (Kutbi et al., 1986)

Name of the item	MSF	MED	RO
Pretreatment	Treated water is required for better efficiency	Treated water is required for better efficiency	Pretreatment of feedwater is compulsory. The efficiency of the plant depends on the pretreatment
Type of water handling capacity	Dirty or unclean water can be handled efficiently	Dirty or unclean water can be handled efficiently	Less efficient with dirty or unclear water
Chemical requirement	Low	Low	High
Production of sludge	No sludge produced	No sludge produced	High salinity brine is produced
Operation	Easy to operate	Easy to operate	Complex to operate
Capital investment	Very high amount of capital investment is required	High amount of capital investment is required	Lower capital investment is required compared to MSD or MSF methods
Type of energy required	Both steam and electricity can be used	Both steam and electricity can be used	Electricity is required to operate the RO plant. A little amount of thermal energy is required to operate other equipment in the plant
Energy consumption	High energy is required	Medium energy is required	High energy is required
Safety consideration	Possibility of radiation contamination as both the plants are present in the same site	Possibility of radiation contamination as both the plants are present in the same site	RO plant can be installed remotely so that nuclear contamination can be limited nuclear plant only
Pollution	Thermal pollution is high	Thermal pollution is high	Thermal pollution is medium
Location	Both nuclear and MSF plants must be located at the same site	Both nuclear and MED plants must be located at the same site	Nuclear and desalination plants can be located at different sites
Product transportation cost	Water transportation cost is required	Water transportation cost is required	Water transportation cost is not required
Total dissolved solids (TDS)	Low TDS values are obtained in the product water	Low TDS values are obtained in the product water	High TDS values are obtained in the product water
Purity of water	Good purity of water is obtained	Good purity of water is obtained	Low purity of water is obtained
Size of plant	Large size units can be used	Small size units are required	Very large size units can be used

The integration of nuclear energy plants with thermal desalination units facilitates the energy requirements for heating and electricity in the desalination plants. Besides, these units supply surplus power to the local electricity grids in a reliable and economical way. Coupling of either thermal or mechanical desalination units (MSF or RO) with nuclear reactors produces good quality potable water and minimises production costs (Al-Mutaz, 1997a, 1997b). Table 2 illustrates the advantages and disadvantages of the nuclear plant's coupling with the RO plant and nuclear plant with the MSF plant (Kutbi et al., 1986).

#### 2.1.1 Coupling of Dual-Purpose Reactor with Thermal Distillation Units (MED or MSF)

A dual-purpose nuclear desalination coupling plant produces both heat and electrical power; heat energy produced is used to increase temperatures required for the thermal desalination unit, and electricity generated is connected to the electric grid to cater to the needs of the surrounding people. The coupling mechanism of a dual-purpose nuclear reactor integrated with a thermal desalination plant by MED and MSF is schematically illustrated in Figs. 1 and 2, respectively. The basic principle involved in the two methods is the same except for the cooling circuit. In dual-purpose plants, thermal energy released is utilised for stream production in a boiler. This steam is utilised for rotating a turbine for the production of electricity. The heat energy present in the turbine's outlet water is transferred to the desalination units with the help of the cooling condenser/heat exchanger. In this configuration, both the plants are sharing common facilities and are hence prone to radioactive contamination. Coolant loop pressures are maintained in such a way that contamination will not transfer to the product. The heat removed from the condenser is transferred to the flash tank, which provides the feed temperature suitable for thermal desalination processes. Scaling inhibitors are added in MSF and MED plants to avoid scaling in the pretreatment of seawater. More stages are beneficial in the MSF system as the number of stages improves the MSF system's efficiency.

## 2.1.2 Coupling of Heat-Only Reactor with Thermal Distillation Units

The coupling mechanism of the heat-only reactor coupled with the thermal desalination plant by either MED or MSF is schematically illustrated in Fig. 3. Heat-only reactors are designed solely to produce thermal energy. The heat produced in the nuclear reactor is transfer to the boiler, where steam is produced. The steam generated from the boiler is pumped to a MED or MSF desalination system for supplying the required temperature for the desalination process. The heat produced in the nuclear plant may be directly transferred to the MED or MSF system based on the temperature and pressure used in the desalination process.

#### 2.1.3 Dual Purpose Nuclear Plant Coupling to RO Preheat System

In a dual-purpose nuclear plant, thermal energy produced is used for electricity production as well as the heat supply for various processes used in the plant. It is estimated that only one-third of this is utilised for electricity generation, and the remaining is used for thermal processes. The working principle involved in the coupling of a nuclear plant with the RO unit is schematically illustrated in Fig. 4. The heat released at the integrated plant is transported to the steam generation unit/boiler to produce high-pressure steam. This steam is passed through the turbine and generates electrical energy. Used water available from the turbine is sent to a cooling system where feed water is preheated to achieve the suitable temperature used in the RO plant. RO system takes energy primarily from electrical energy, and a small amount of thermal energy is needed for preheating purposes.

#### 2.1.4 Nuclear Power Plant Coupling with a Hybrid Thermal–Mechanical Desalination Plant

Nuclear power plants designed to generate both heat and electric power can be coupled simultaneously with both thermal and mechanical desalination plants. This coupling mechanism provides a reduction of water cost and produces

**Fig. 1** Multi-effect distillation (MED) to dual-purpose nuclear reactor coupling (IAEA, 2001)





high-quality water (Al Bazedi et al., 2019; Shaorong & Zheng, 2002). Generally, this hybrid coupling system consists of the multistage flash (MSF)/MED distillation plant, followed by the RO desalination plant. This arrangement facilitates the use of outlet water from the MSF unit as feed

water to the RO desalination plant; thereby a significant reduction in the cost of production is achieved. This hybrid coupling system offers economic and technical advantages.

This hybrid desalination coupling method is also used for achieving the benefits of each individual method. However,





this coupling mechanism is very complex and is schematically illustrated in Fig. 5. In this coupling system, the nuclear power plant is connected to the MED/MSF distillation plant, followed by a RO plant. In hybrid plants, thermal energy released in the nuclear reactor plant is utilised to stream production in a boiler. This steam is utilised for rotating a turbine for the production of electricity. The heat energy present in the turbine's outlet water is transferred to the desalination units with the help of a cooling condenser/heat exchanger. The heat removed from the condenser is transferred to the flash tank, which provides the feed temperature suitable for thermal desalination units. The used outlet water from the MDF or MED unit is supplied as feed water to the reverse osmosis unit, as shown in Fig. 5.

In India, the RO-MSF nuclear desalination plant is constructed in Kalpakkam, Tamil Nadu. The desalination plants are coupled with a  $2 \times 170$  MW(e) capacity nuclear power plant. This plant has a production capacity of 1800 m<sup>3</sup>/d of water through the MSF plant and 4500 m<sup>3</sup>/d of water through the RO plant. In 2003, 284 k m<sup>3</sup>/d of MSF and 170 k m<sup>3</sup>/d of RO desalination plants were coupled in the Fujairah seawater desalination project in the United Arab Emirates. This coupling technology allowed the use the water, power and heat in the most economical way and produced good quality water at a lower cost.

It was found that the nuclear reactor (CANDU) developed by the atomic energy of Canada is very effective for electricity generation and water production in the Middle East Countries (Al-Mutaz, 1997a, 1997b, 2001, 2002; Al-Suliman et al., 1995). Pressurised heavy water is used as a coolant in this reactor. Coupling of this CANDU PHWR with MSF and RO plant showed better results in electricity and water generation. This hybrid system provided improved water quality at a low running cost.

#### 3 Safety and Licensing Issues

Coupling of nuclear reactors with desalination plants shares many common facilities and systems, whatever the manner they are coupled. The common use of facilities may create many safety considerations, such as the possibility of radioactive diffusion in distillation plants through heat transfer circuits, the possibility of radioactive threat due to the sharing of preheated water in the RO plant. Special provisions must be made to produce water continuously during unforeseen reactor outage situations. National authorities must regulate and see that radioactive components concentration must be within the international prescribed levels.

International Atomic Energy Agency looks after the development and publication of standards related to plant design, installation and nuclear desalination plants' maintenance. IAEA publishes safety guidelines in a series of published books such as Nuclear Safety Standards (NUSS). Due to the changes and advancements in technology, IAEA publishes revised new guidelines separately as new safety standards series. These publications include information to relate to objectives, technology, concepts and methods that will promote nuclear energy use. These standards should be



Fig. 5 Hybrid MED-RO desalination system to nuclear reactor coupling (IAEA, 2001)

followed by all regulatory bodies or member countries installing and producing nuclear energy or willing to start new projects in the near future.

IAEA safety guidelines are mainly organised into three categories: safety fundamentals, safety requirements and safety guides. The primary category is the safe fundamentals that publish the basic objectives, concepts and principles required to ensure safety during the development and application of nuclear energy. This category mainly focuses on the general safety of individuals, society and the environment from nuclear exposure and other hazards. It broadcasts guidelines for the protection against radiation caused by the radioactive materials used in nuclear reactors. It publishes guidelines regarding prescribed exposure limits, precautionary measures to prevent accidents during installation and running, guidelines on minimising the hazards during accidents, future health hazards due to radioactive exposure, etc.

Safety considerations and best practices that must be followed during the installation and operation of nuclear desalination plants are documented and published in the safety requirements category. This category deals with problems associated with siting, design, operation, governmental organisation and quality assurance. This category mainly focuses on equipment and other components used for design, operation and other plant activities. Separate guidelines are published regularly for already existing hybrid reactors and research reactors. The safety guides category publishes guides based on member countries' recommendations through their experience of working with nuclear reactors. Through the above three categories, IAEA continuously monitors the plant's operations, guides the member countries to minimise the hazards during installation and operation, and directs the member countries to promote sustainable renewable resources for desalination methods.

Even though the latest hybrid technologies used to couple nuclear reactors with desalination plants offer many benefits still some of the issues such as safety, social and economic issues need to be addressed. The major safety problems associated with a nuclear reactor-desalination coupled plants could be due to nuclear plants themseves. These may include potential health hazards due to the diffusion of radioactive materials into potable water, radioactive exposure to persons working in the plant or in surrounding areas of the plant. Other environmental issues associated with effluents' disposal into the nearby water resources and environment must be considered. All these safety issues must be addressed for starting these plants in populated areas.

The impact of waste brine from the nuclear desalination plant on marine animals and the application of suitable alleviation measures need to be evaluated. In spite of the large advantages associated with these hybrid technologies, the growth of the installation of such plants is very slow due to public perception of the negative impact on human health. The governments still need to create awareness among the people residing in surrounding areas for getting approval to install such plants. Since technology and problems associated with them are dynamic, all the above-mentioned safety standards must be reviewed continuously and revised to ensure the safety of the working personnel and people residing around the plant.

#### 4 Economic Assessment

Nuclear desalination technology is proved to be a competitive solution for providing sustainable water supply in an environmentally friendly way. This technology can be used to meet the water requirements in domestic, industrial and agricultural needs. Recent developments in nuclear desalination technology have shown great improvements in water production. The water production costs have been reduced a lot during the last decade. IAEA has recommended the member countries to use the Desalination Economic Evaluation Program Software (DEEP-3) for economic feasibility surveys (IAEA, 2006; 2000b). Using this software, IAEA member countries designed various models to assess the economic feasibility and performance of different types of nuclear reactor-desalination coupling technologies. Studies such as preliminary economic evaluation and energy sources like fossil fuels and nuclear energy were also done. The results were very promising and signified the importance of nuclear desalination coupling technologies in the sustainable water supply. The nuclear desalination process is mainly classified into two types, thermal and membrane types. Studies show that around 65% of the total desalination processes used in the world are membrane-type; this percentage may further grow up in the future (Sadeghi et al., 2020). However, the selection of the desalination process depends upon factors such as plant capacity, design of the plant, feed water quality, technical expertise available in the country, energy consumption, etc. Energy consumption and economics of the plant depend on local geographical factors also hence all these factors have to be considered for the selection of the process.

#### 5 Summary

Water and energy are two important critical parameters for sustained development. Rapid growth in population, along with unplanned urbanisation and industrialisation, resulted in water scarcity throughout the globe. The release of toxic pollutants into the environment through the burning of fossil fuels for energy generation caused global warming, and it further deteriorated the availability of renewable energy resources. Desalination, coupled with nuclear reactors, offers sustainable water and energy resources to the current and future generations. Despite the fact that nuclear plant coupled with desalination plants offers many economic, energy-related benefits, it is very dangerous to work with nuclear plants because of possibilities of nuclear contamination of water. Even though stringent safety and precautionary measures are in practice in these plants, there is a possibility of unforeseen accidents. Hence, its use must be regulated by governmental and international agencies.

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### **Carbon-Based Materials for Desalination**

Marzie Fatehin and Ali Mohebbin

#### Abstract

Nowadays, the scarcity of sanitary water has become the biggest concern in the world. There are many proposed solutions to solve this problem. Water reuse and increasing water supply by desalination of salty water are promising choices. Generally, polymeric membranes are used to remove salts, minerals and contaminants from polluted and brackish water during desalination processes. The membrane-based methods are popular due to lower cost and higher energy efficiency than other methods. Recently, carbon-based nanomaterials (CBNMs) have attracted great attention with respect to the conventional membrane in the desalination process. The porous structure, chemical/ physical stability, excellent surface area, environmentfriendly, low density and antibacterial feature are exceptional properties of CBNMs, which make them supreme candidates. We reviewed the application and performance of CBNMs in the desalination processes. The survey of the current researches and available literature indicated that CBNMs can improve the water permeation flux, ion rejection, hydrophilicity, durability, desalination efficiency, specific surface area, antifouling and selectivity of the pristine membrane. Carbon nanotube, graphene and its derivatives, activated carbon, carbon nonfilter, MXene and fullerene are familiar CBNMs, which are discussed in this chapter.

#### Keywords

Desalination • Membrane • Carbon-based nanomaterialsWater permeation flux • Ion rejection

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#### 1 Introduction

The growing population and developing industrial activity increase the demand for clean, fresh and sanitary water. The scarcity of fresh water is a serious global challenge and becomes a national security issue in the coming years (Shannon et al., 2008). Water is one of the most abundant liquids on earth, and it is estimated that approximately 3% of water on the earth is fresh water. A large amount of fresh water is inaccessible in polar ice form. Natural conditions and human activity are the reason for freshwater shortage. It is obvious that the quality of water potently influences the farming production, environment and outputs of industries. These global concerns make us purify water using low-cost and high-efficient methods. Water desalination is one of the promising strategies to response for growing freshwater needs (Shenvi et al., 2015). Desalination is a natural or human-driven process for the removal of salts, minerals and contaminants from polluted and brackish water. Membraneand thermal-based techniques are two classes of water desalination by humans. The membrane-based methods are more environment-friendly, energy efficient and low cost than thermal-based methods. Due to these advantages, pure water production is increased extremely over the years by membrane-based methods (Fig. 1).

A membrane is a narrow permeable barrier, which water molecules or ions pass through its pores by higher pressure than osmotic pressure or electrical potential, respectively. According to the membrane performance, they are divided into reverse osmosis, forward osmosis, nanofiltration, microfiltration, electrodialysis, pervapouration, capacitive deionisation, ultrafiltration and membrane distillation. The porosity and pore size of membranes have an essential effect on the rejection of salt, mineral or contaminants. On the other hand, the thickness and surface of membranes change the hydraulic permeation flux. Driving force, structure and type of membrane materials are other important parameters (Chandrashekara & Yaday, 2017; Giwa et al., 2016; Qadir

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**Fig. 1** Pure water production versus years with water desalination by membrane-based technology according to the estimation of Desaldata.com. Modified after Hebbar et al. (2017)



et al., 2017; Sharon & Reddy, 2015). Therefore, much attention has been paid to membrane modification. Conventional polymeric membranes have some disadvantages like fouling, chemical and physical changes. Nanocomposites are the emerging materials to improve membranes and are divided into inorganic and organic that organic nanomaterials are the most capable to improve membrane process.

Nowadays, the carbon-based nanomaterials (CBNMs) have attracted the consideration of researchers due to their high surface area, excellent mechanical properties, large pore volumes, antibacterial feature, low density and good chemical/physical stability. They have high adsorption ability because of their hollow and porous structures and also decrease energy consumption during the desalination process. According to the position of CBNMs and structure of membrane, advanced membranes are divided into three types: freestanding CBNM membranes, membranes with modified surface and CBNMs embedded membranes.

Carbon nanotube, graphene and its derivatives, carbon nonfilter, MXene, fullerene and activated carbon are the most popular CBNMs, which reviewed in this chapter. The structure and characteristics of CBNMs are discussed in detail. The objective of this chapter is to reveal the CBNMs potential for improvement of pristine membranes, desalination performance and also for increasing desalination efficiency of other CBNMs.

#### 2 Carbon Nanotube

Carbon nanotubes (CNTs) are the most famous member of carbon nanomaterials, which introduced in 1991 by Iijima et al. At high temperatures, hydrocarbons disintegrate to fabricate carbon filaments. Then, fibres of carbon are rolled to produce single and multiwalled CNTs (Fig. 2) (Goh et al., 2013; Hebbar et al., 2017).

The cavity structure of CNTs enables them to pass quickly water molecules through their channels in the desalination process, as shown in Fig. 3.

Capillary phenomena, internal surface of CNTs, radius of pores and pore distribution have an essential role in water permeation flux and ion rejection (Goh et al., 2013). It would be interesting to know that the viscosity of liquid affects the water permeation flux and the rejection of ions so that at high salt concentration, ion rejection and water permeation rate decrease (Zaib & Fath, 2013). On the other hand, some theoretical studies show that the number of walls can influence water permeation flux and ion selection of CNTs membranes (Rizzuto et al., 2018).

According to the presented experimental and simulation results, water molecules can pass through narrow vacant space of CNTs (Talati et al., 2019). The exceptional characteristics of CNTs have made them an appropriate choice to improve the performance of conventional membranes



**Fig. 2** Multiwalled (a) and single-walled (b) carbon nanotubes. Reprinted with permission of Hebbar et al. (2017)

**Fig. 3** Performance of carbon nanotubes in water desalination process. Reprinted with permission of Hebbar et al. (2017)



(Hebbar et al., 2017). Interaction between internal surface of CNTs and salt water causes to pass water molecules and reject ions. Das et al. (2014) investigated the molecular performance of carbon nanotube (CNT) membrane in salty water desalination. They emphasised that the fabrication method of CNTs can influence its performance. Vertically aligned and mixed matrix (Fig. 4) are two kinds of CNT membranes.

Vertical type by complex fabrication process possesses compact structure with high water permeation flux, while mixed matrix CNT membranes by simpler fabrication process have loosely structure and lower water flux. The chemical vapour deposition is introduced as the best method for vertically aligned CNTs fabrication as shown in Fig. 5.

Majumder et al. (2005) indicated experimentally that liquid flow rate through CNT membranes is faster than that of hydrodynamics prediction (four to five orders of magnitude). Frictionless wall of CNTs is the reason for this high flow rate of water. There are various experimental studies, which verify the increase of water permeation flux by use of CNTs with respect to the conventional membrane. Conventional membranes have some disadvantages like the need for pressure force for the mandatory passage of water molecules and the accumulation of suspended particles. Table 1 provides the differences between CNT membrane and conventional one. Moreover, Table 2 lists obtained results of some current studies on the performance of various CNTs in water desalination process. According to the presented results, carbon nanotubes are promising choices, which can improve and modify the performance of pristine membranes.

#### 3 Graphene

Graphene is a two-dimensional structure of graphite with impressive specific surface area, low weight, electron mobility and thermal conductivity. Impermeability of graphene for small molecules has made it appropriate for fabrication super narrow membrane to sieve molecules (Zaib & Fath, 2013). Therefore, graphene as a building block is used to produce graphene oxide, CNTs and fullerene.

An et al. (2017) reviewed the ability of this green material for water desalination and in purification technologies. They investigated wetting characteristics and porous structure of graphene to contribute the explicit insight of water behaviour on the surface of this carbon-based material. Water contact angle (WCA) was used for classifying graphene as hydrophobic, hydrophilic, superhydrophobic or superhydrophilic material. In total, the WCAs of hydrophobic, hydrophilic, superhydrophobic and superhydrophilic surfaces are between 90 and 180°, between 0 and 90°, above



**Fig. 4** Vertically aligned (**a**) and mixed matrix (**b**) types of carbon nanotubes (CNTs) membrane. Modified after Das et al. (2014)

 $150^{\circ}$  and  $\sim 0^{\circ}$ , respectively. Graphene is ordinarily considered as hydrophobic material, while its WCA is  $45^{\circ}$ . The WCA of graphene surface is related to the presence of hydrocarbons on its surface, which increases with exposure to air. The graphene surface adsorbs airborne particles after exposure, and consequently, the surface energy is decreased (Fig. 6). Therefore, the most graphene surfaces, which are used in industries, show hydrophobic properties due to hydrocarbon contamination.

Recently, superhydrophobic surfaces have attracted more consideration because of their valuable properties like self-cleaning (lotus effect). There are many studies to introduce novel superhydrophobic materials. Among the recommended materials, carbon-based materials are interesting for researchers. Therefore, they have been encouraged to improve the surface of carbon-based materials to achieve superhydrophobicity.

There are many methods to produce superhydrophobic graphene, those change the wettability of graphene and modify its surface by structural and chemical techniques. One of the most common structural modification methods is to combine graphene with other materials to form nanoscale pores on its structure. On the other hand, chemical methods enhance graphene hydrophobicity. Table 3 presents the WCAs of modified graphene by chemical and structural methods.

Graphene with nanoporous structure is a good option for water desalination due to its excellent properties like high mechanical strength (especially in high pressure), high ion selectivity and low pollutant fouling. Figure 7 depicts the performance of graphene-based membranes in water desalination process.

Some simulation researches indicated that pore size distribution of graphene membranes influences the ion rejection, selectivity, water permeation flow and consequently water desalination efficiency. Thus, production of nanosized pores on the graphene surface modifies the performance of nanoporous graphene membranes compared to conventional one.

In an experimental study, nanoporous graphene monolayer was examined as a membrane for water desalination (Surwade et al., 2015). Nanopores were generated with plasma etching to study ion transport and water permeation. They introduced oxygen plasma as a suitable method to generate an appropriate pore size distribution on the graphene surface. The results exhibit high salt removal approximately 100% and fast water transfer. Three water molecules passed through the porous single layer graphene during one picosecond (ps), which is extremely a high flow rate compared to conventional surface (200 water molecules per microsecond ( $\mu$ s)).

Electron and ion beam irradiation are successful approaches to fabricate nanoporous graphene. These methods change atomic structure by atomic motion and reassemble the atoms (Dyck et al., 2020). Russo and Golovchenko (2012) presented an appropriate method for generating very small pores (3 Å) in graphene structure with beam irradiation. Furthermore, there are modelling and simulation researches on beam irradiation phenomena to figure out the formation of nanopores. Bai et al. (2016) simulated the fabrication of nanopores by oblique beam irradiation of ions and investigated the effect of the incident angle and energy of ions on the efficiency and quality of generated nanopores. In a low incident angle, pores were elliptic in shape. On the other hand, the size of nanopores increased by reducing ion energy due to enhancement in the amount of ions adsorbed. In another study, the generation of nanopores on the



Fig. 5 Chemical vapour deposition (CVD) method for production of vertically aligned carbon nanotubes (CNTs) membrane. Modified after Das et al. (2014)

graphene surface was investigated through electron and ion beam irradiation method by molecular dynamics simulation (Wu et al., 2015). They focused on the effects of beam parameters on the quality of pores and mechanical properties of graphene. The obtained results showed that generation of nanopores can be happened in supported and suspended graphene.

In addition to beam irradiation, chemical oxidation and block copolymer template are used to produce porous graphene. Kim et al. (2016) believed that these methods cannot be able to produce low cost membrane with high efficiency. Therefore, a new method was presented to fabricate high-dense graphene with uniform pore structure. After peroxidation of graphite, they produced nanopores (3 nm) by potassium hydroxide (KOH) activation. Finally, nanoporous graphene was produced by dispersing of graphite. Water permeation flux of this modified graphene was six times that of conventional graphene membrane.

The performance of CNTs and graphene membranes was compared in water desalination by Suk and Aluru (2010). They investigated the effect of pore radius on water flux and used CNTs with 2–10 nm length and 0.375–1.375 nm radius and graphene with the same radius of pores. The simulation results showed that CNT membranes have higher permeation flux than graphene membranes for smaller radius of pores, while for larger radius of pores, graphene membranes have better performance (approximately two times).

In another simulation study, in order to survey water and ions molecule behaviour in the graphene structure, Konatham et al. (2013) performed molecular dynamics

	CNT	RO	NF	UF	MF
Definition	Porous structure with vertically aligned and mixed matrix forms	Purification process to remove ions from salty water by applying pressure to overcome osmotic pressure	Removal process for molecules larger than 2 nm	Removal process for solute particles (>1–100 nm in diameter) using concentration or pressure gradients	Rejection process for macromolecules larger than 100 nm
Particular application	Desalination and selective pollutant removal pollutant	Desalination, reuse and ultrapure production of water	Removal of water hardness, dissolved organic and heavy metals	Removal of virus and colloid	Removal of bacteria suspended solids and protozoa
Materials	CNTs and polymers	Organic polymers	Organic polymers	Polysulphone, acrylic and cellulose	Polypropylene, polysulphone and polyurethane
Size of pores (nm)	0.1–2	0.3–0.6	<2	2–50	>50-500
Thickness (µm)	26	~0.1-0.2	~ 0.05	150–300	50-100
Permeation flux (mPa <sup>-1</sup> s <sup>-1</sup> )	$\sim 7 \times 10^{-7}$	$\sim 3 \times 10^{-12}$	$\sim 40 \times 10^{-12}$	$\sim 0.5 \times 10^{-10}$	-
Capability of ion rejection	Good	Good	Good	Moderate	Poor
Self-cleaning ability	Capable with or without functionalisation	Only with functionalisation	Only with functionalisation	Only with functionalisation	Only with functionalisation
Tunable selectivity	Mixed matrix only	Mixed matrix only	Mixed matrix only	Mixed matrix with ceramic reactivity	Mixed matrix with ceramic reactivity
Fouling of membrane	No	Yes	Yes	Yes	Yes
Required pressure (bar)	Negligible	30-60	20–40	1–10	<1.0

Table 1 Detailed comparison between carbon nanotubes (CNTs)-based membrane and regular membranes

MF microfiltration, NF nanofiltration, RO reverse osmosis and UF ultrafiltration

Modified after Das et al. (2014)

simulations for a graphene sheet. They evaluated the potential of mean force to monitor transposition of water and ions molecules through pores with various radii (from 3.75 to 7.25 Å), which are generated by using of some functional groups like carboxyl groups. The results indicated that ions cannot pass through pristine pores with radius < 3.75 Å, while they pass through larger pores ( $\sim 5.25$  and 7.25 Å). On the other hand, carboxyl groups can improve ion rejection for all pore radii, but by increasing pore radius and ion concentrations, the effect of carboxyl functional groups on ion rejection decreased. Nevertheless, ion rejection of this type of graphene was higher than that of CNT with similar pore distribution. Therefore, they concluded that the pores functionalised with hydroxyl groups improve the graphene desalination performance. These researches indicated that the desalination performance of graphene-based membrane

is more remarkable than polymeric membrane in water purification.

#### 4 Graphene Oxide

In total, graphene oxide (GO) is fabricated through graphene oxidation process and adding some functional groups like hydroxyl to the graphene structure (Manawi et al., 2016). Because of its hollow structure, the chemical and physical stability and high sorption capacity, GO has tremendous contribution in solar desalination and sometimes it is considered as filtration film in other desalination processes (Li et al., 2019). The mechanism of water transport through GO layers was investigated by Chen et al. (2017) using molecular dynamics simulation. The results showed that  
 Table 2
 Summaries of some
 recent studies about the application and performance of carbon nanotubes (CNTs) in water desalination processes

Type of study	Main results	References
Theoretical	Hydration of CNTs pores was affected by attraction between wall of tubes and water molecules. On the other hand, the change of channel polarity and solvent conditions mutated occupancy of CNTs channel	Hummer et al. (2001)
Experimental	Modified multiwalled carbon nanotubes by Fe <sub>2</sub> O <sub>3</sub> particles were fabricated during chemical process and investigated adsorption of salt using them. High efficiency for desalination of seawater and excellent repeatability was observed for these advanced membranes	Mishra and Ramaprabhu (2010)
Experimental	Interactions of solute and membrane are altered by immobilisation, and consequently membrane permeation and selection. Salt rejection and permeation flux enhanced to 15 and 1.85 times, respectively, after incorporation of CNTs	Gethard et al. (2011)
Experimental	The performance of thin-film composite membrane modified by single-walled CNTs, and that of conventional membrane was compared in reverse and forward osmosis processes. High ion selectivity and water flux were observed for modified membrane compared to conventional membrane during reverse and forward osmosis, respectively	Tang et al. (2020)
Theoretical	Using the density functional theory, natural bond orbital and quantum theory, the interaction between ions and simple CNTs and doped one was investigated. Ions like $Fe^{2+}$ , $Na^+$ , $Cl^-$ had covalent interaction with two types of CNTs	Tavakol et al. (2020)
Theoretical	Water permeation and salt rejection of charged and uncharged multiwalled CNTs were assessed. The performance of these two kinds of CNTs was better than that of graphene or boron nitride in water desalination process. Poor infarction between water molecules and CNTs surface molecules increased water pass through CNTs channel. In addition, charged multiwalled CNTs enhanced anchoring of water and ions molecules and consequently increased water pass flux in central region of channels compared to the near of walls	Ghoufi and Szymczyk (2020)



Air exposure duration

Fig. 6 Water contact angle changes during air exposure. CA: contact angle, Cu: copper, G: graphene and Ni: nickel. Modified after An et al. (2017)

significant water permeation flux for GO-based membrane is related to hydrogen interactions between water molecules and functional groups of GO. The GO concentration influences these interactions and consequently slip velocity of water. They also concluded that water permeation flux of GO improves after reduction.

Despite the inherent advantages of this carbon-based material, some researches have been done to improve its specifications. Parsamehr et al. (2019) introduced a new GO-based membrane, which is fabricated during carboxyl groups activation of GO and then cross-linking to polyethylenimine. Stability and salt rejection of this improved membrane increased in comparison with those of simple polyethylenimine membranes. In addition, the modified membrane showed excellent ability in MgCl<sub>2</sub> and Na<sub>2</sub>SO<sub>4</sub> rejection.

In another study, Romaniak et al. (2020) used hydrazine-treated GO to improve the efficiency of graphene-based membranes in water treatment. Actually, hydrazine affects functional groups of GO and changes filtering properties. Water permeation flux and the ion rejection rate of modified membrane were enhanced compared to graphene-based membrane. These results confirmed that CBNMs can improve desalination the efficiency of advanced membrane by combining with other CBNMs. Table 4

**Table 3** Water contact angles(WCA) of modified graphene bychemical and structural methods

	Туре	WCA (°)
Structural modification	Polyvinylidene fluoride and graphene	~153
	Three-dimensional graphene foam coated by Teflon	143–163
	Two-dimensional grating like structure	~155
Chemical modification	Octadecylamine-functionalised graphene oxide film	~163
	Perfluorodecyl-trichlorosilane-treated graphene aerogel	~160

Modified after An et al. (2017)

**Fig. 7** Performance of nanoporous graphene (a) and stacked graphene oxide-based (b) membranes in water desalination process. Reprinted with permission of An et al. (2017)



presents the summaries of recent studies on the performance of GO and modified GO in water desalination process. Based on the results, GO and reduced GO are promising choices to improve the ability of thin-film composite and thin-film nanocomposite types of membranes in forward osmosis and nanofiltration processes.

#### 5 Activated Carbon

The activated carbons are fabricated from natural sources like wood, coconut shell and charcoal. Extreme surface area, hollow form and acceptable surface reactivity have made them an appropriate choice to improve capacitive deionisation method for water desalination. Ahirrao et al. (2019) synthesised activated carbon with significant porous structure and surface area from sweet lime peels through facile method at low temperature. They prepared the supercapacitor and porous electrodes to characterise the applicability of this inexpensive activated carbon during electrode capacitive deionisation. The obtained results showed remarkable stability, excellent storage capacity of electrochemical charge, high salt rejection and appropriate energy density for this inexpensive activated carbon electrode. Electrosorption efficiency of electrodes can be enhanced by surface modification of the activated carbon using chemicals and metal oxide nanoparticles. Zou et al. (2008) investigated the potential of activated carbon and surface-modified activated carbon for desalination of brackish water. They modified the surface of activated carbon by alkaline treatment and loading TiO<sub>2</sub> nanoparticles. The treated activated carbon had higher electrosorption capacity than untreated sample (about 5%). Actually, alkaline treatment increases hydrophilic compounds, which this change increases salt ions adsorption. After loading TiO<sub>2</sub> nanoparticles on the activated carbon surface, electrosorption capacity was enhanced by about 10% due to reduction of physical adsorption. These results demonstrated that surface treatment improved the

Fabrication procedure	Main result	References
Graphite flakes were used to produce graphene oxide using Hummer's method. To improve polyamide thin-film nanocomposite membranes	Water permeation flux increased with increasing graphene oxide concentration in improved polyamide membranes. It is due to enhancing of membrane hydrophilicity and attraction changing of water molecules with membrane wall. On the other hand, with increasing concentration of graphene oxide, salt rejection enhanced especially for MgSO <sub>4</sub> solution	Bano et al., (2015)
Graphene oxide was produced using graphite by Hummer's method and then incorporated into the polyacrylonitrile thin-film composite membranes	Incorporation of graphene enhanced smoothness of thin-film composite membranes and its hydrophilicity. They reported high permeation flux (23.6 $L/m^2$ h) and low salt flux (4.4 g/m <sup>2</sup> h) compared with control samples	Shen et al., (2016)
During phase inversion process, graphene oxide–cellulose acetate was produced by dispersion of graphene oxide in formamide and mixing with acetone including cellulose acetate. Finally, graphene oxide–cellulose acetate nanocomposite membranes were prepared with different graphene oxide contents	The skin layer thickness and prepared membrane contact angle decreased by increasing graphene oxide content from 0 to 0.01 wt%. Permeation flux of membrane containing 0.01 wt% graphene oxide was higher than that of 0 wt.% (2.3 times), in addition salt retention decreased (15%)	Shi et al., (2017)
Graphene oxide was produced by natural graphite powder and reduced during a novel hydrothermal process and then attached on a polyethylene film	In this study, the key roles of functional group of reduced graphene oxide on the membrane performance were studied. The results exhibited the significant effects of hydroxyl and carboxyl groups of reduced graphene oxide on the uniform structure of membrane. This type of advanced membrane had high water permeation and salt rejection	Huang et al., (2019)
Sulphonated graphene oxide was prepared with graphene oxide nanosheets and sulphonic acid to improve polyamide thin-film nanocomposite membrane	Modified nanofiltration membrane had higher antifouling and water permeation flux (87.3%) than control sample, but salt rejection of thin-film nanocomposite membranes and that of modified sulphonated graphene oxide membrane was the same	Kang et al., (2019)
Graphene oxide nanosheets were synthesised by graphite flakes and used to modification of polyamide thin-film composite membrane by layer-by-layer deposition procedure	The water flux of polyethylenimine with positive charge and graphene oxide membrane with negative charge was two times that of compared sample membrane. NaCl, Na <sub>2</sub> SO <sub>4</sub> , MgSO <sub>4</sub> and MgCl <sub>2</sub> solutions were used to investigate the modified membrane's ability in salt rejection. At different temperatures and salt concentrations, 99.9% salt rejection with 8 kg/m <sup>2</sup> h water permeation flux was observed	Halakoo and Feng (2020)
Uniform graphene oxide (GO-0) with different flake sizes was prepared by sonicating dispersion and then embedded in the polyamide thin-film composite membrane	This study investigated the effects of flake size of graphene oxide on the structure and modified membrane performance in forward osmosis process. For graphene oxide flake size of $0.01 \ \mu\text{m}^2$ , water permeation flux and salt removal increased 1.5 and 1.6 times, respectively, compared to the size of $1.06 \ \mu\text{m}^2$	Akther et al., (2020)

desalination efficiency of activated carbon, especially loading of metal dioxide nanoparticles.

However, hydrophobicity of activated carbon is an essential problem to use it in desalination process. In capacitive deionisation method, ions are accumulated onto electrodes by activated carbon by applying a voltage and then pure water is removed (Fig. 8).

During capacitive deionisation process, the electrodes wettability must be enhanced (Luciano et al., 2020). Therefore, hydrophobicity of electrodes is reduced using various methods like adding the ion-exchange resin (Lee et al., 2009). Unlike activated carbon problems, CNTs and carbon aerogels are better alternatives for this purpose (Fellman et al., 2010; Zaib & Fath, 2013). Actually, capacitive deionisation method is used for low salinity water and has not economic efficiency for high concentration of ions.

#### 6 Carbon Nanofibres

Carbon nanofibres (CNFs) are the porous structure, which synthesised by stacking one or more graphite layer (Fig. 9) (Palmeri et al., 2011).

Electrospinning is a conventional method for fabrication of nanofibres, and this method is widely used due to the tunable precursor solution. Carbon nanofibres are considered as energy storage and an effective choice for capacitive deionisation process for water desalination. In many cases, **Fig. 8** Capacitive deionisation method by carbon aerogel. Modified after Zaib and Fath (2013)





**Fig. 9** Transmission electron microscopy image of stacked graphite layers to produce carbon nanofibre. Reprinted with permission of Palmeri et al. (2011)

polyacrylonitrile is precursor solution in the production of electrospun fibres due to its high carbon percentage. Polyacrylonitrile-based CNFs are generally modified by some activation additives like polymethyl methacrylate, polystyrene, polyacrylic acid and poly (L-lactide) to improve their specific surface area and pore size. Liu et al. (2016) prepared two types of CNFs during electrospinning method and modified with polymethyl methacrylate and polystyrene as additives. These prepared CNFs had different pore sizes and structural characteristics. They also studied the pore size distribution effect on the performance of modified CNFs in capacitive deionisation desalination. Two modified CNFs had significantly higher capacitance than the control sample, and also their specific surface area was higher. Therefore, they had excellent performance and high stability in capacitive deionisation desalination.

In another study, Zhang et al. (2018) developed an effective method to produce hierarchical porous CNFs during electrospinning process. Polyacrylonitrile nanofibres were used to fabricate CNFs. The modified CNFs had high electrical conductivity, high specific surface area (223.9 m<sup>2</sup>/g) and excellent salt capacity (36 mg/g), which their sorption capacity is retained after more cycles. They also reported that the efficient deionisation time of prepared CNFs was more than that of conventional CNFs.

In another work, Jashni et al. (2019) embedded CNFs into the polyvinyl chloride membrane matrix and fabricated an exchange membrane based on a novel electrodialysis mixed matrix cation method. The application of CNFs enhanced obviously the hydrophilicity of membrane surface, membrane selectivity, water permeation flux, salt rejection and conductivity of ions compared to the conventional polyvinyl chloride membrane. The surface of pristine membrane was rougher than that of CNF-based membrane. In addition, the CNFs' concentration effect was investigated on the prepared membranes performance. The results indicated that increasing CNF content influenced the ability of modified membrane by a concentration dependent manner.

Therefore, carbon nanofibres could enhance the efficiency of electrodes in capacitive deionisation desalination and improved the desalination ability of ordinary polymeric membranes.

#### 7 MXene

MXenes are two-dimensional metal carbides with formula of  $M_{n+1}X_nT_X$ , where T represents a surface functional group and *n* is 1–3 (Fig. 10). MXenes are produced by extraction of "A" layers from layered carbonitrides or carbides using HF treatment, which distinguished as MAX. M, A and X refer to early transition metals like Ti, an A-group element like Al, and carbon or nitrogen, respectively (Tian et al., 2019; Wang et al., 2015; Xie et al., 2016).

Because of exceptional features of MXenes like high surface area, environment-friendly, antibacterial nature and hydrophilicity, MXenes have attracted the researcher's attention to use them in water desalination. Ihsanullah (2020) summarised new production methods for MXenes and MXene-based materials, their application in desalination and their mechanisms for ion rejection in a comprehensive review article. Among 30 reported MXenes compositions, titanium (Ti)-based MXenes are familiar for applications in desalination. Titanium-based MXenes exhibit low contact angle ( $<21.5^{\circ}$ ) and high stability during desalination processes. They can also improve conventional membranes to enhance ion rejection. According to the reported results,  $Ti_3C_2T_r$ -based membranes exhibited high water permeation flux and salt ion removal (various ion sizes with different charges). Vacuum-assisted filtration method is appropriate to fabricate  $Ti_3C_2T_x$ -based membranes, while the thickness of the membrane is an essential parameter in water permeation. Furthermore, the distance between  $Ti_3C_2T_r$ -based membrane's layers influences the water flux rate. Sometimes, modification of Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub>-based membranes enhances the water permeation flux, so that this flux for Ag@Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub>based membranes is higher than that of pristine membrane (about 3.5 times). In addition, ion size and charge have

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different effects on the ion rejection, and this rejection can be improved by modifying the charges of membrane surface (Ihsanullah, 2020). On the other hand, MXene materials can be used to improve the desalination efficiency of pristine and modified membranes. A GO-MXene-TiO<sub>2</sub> membrane has higher water permeation flux than pristine GO membrane and GO-TiO<sub>2</sub> membrane, about 7.5 and 2.5 times, respectively (Al-Hamadani et al., 2020).

Table 5 summarises the performance of various MXenes for ion rejection and water permeation in some MXenes-based membranes. Therefore, MXene-based membranes can remarkably develop various water desalination processes based on their promising features.

#### 8 Fullerene

Structural, chemical and physical properties of fullerene and its derivatives have attracted the researcher's attention since 1990. Fullerene is third allotrope of carbon and possesses various types, which  $C_{60}$  is the most valuable of them. This distinctive fullerene has spherical shape and hollow form, which includes 12 pentagons, 20 hexagons and 30 C=C bonds (Pan et al., 2020; Sherigara et al., 2003). Figure 11 illustrates the chemical structure and transmission electron microscopy (TEM) image of fullerene.

High selectivity, ion rejection and water permeation flux are fetching properties of fullerene to use them for modifying conventional membranes (Ma et al., 2016). The rejection and permeation percentages or flux of some fullerene-based membranes are provided in Table 6.

In new theoretical research, Ji and Zhang (2019) explored the application and desalination mechanisms of fullerenebased membrane by molecular dynamics simulation.

**Fig. 10** Scanning electron microscope images of the  $Nb_2CT_x$ , top view (a) and cross-sectional view (b). Reprinted with permission of Byeon et al. (2016)



**Table 5**Summary of ionrejection and water permeationflux for some MXene-basedmembrane

М.	Fatehi	and	Α.	Mohebbi

Type of membrane	Ion rejection percentage	Water permeation (L/m <sup>2</sup> h bar)
$Ti_3C_2T_x$ -based membranes supported on $\alpha$ -Al <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> SO <sub>4</sub> : 75.9 MgSO <sub>4</sub> : 67.3 NaCl: 55.3 MgCl <sub>2</sub> : 46.1	22.4 L
$Ti_3C_2T_x$ -based membrane	-	38
Ag@ $Ti_3C_2T_x$ -based membrane	Methyl green: 92.32 Rhodamine B: 79.93 Bovine serum albumin: 100 NaCl: 25.8 MgCl <sub>2</sub> : 41.3 AlCl <sub>3</sub> : 49.5	420
$Ti_3C_2T_x$ -based membrane supported on anodic aluminium oxide	Bovine serum albumin: 100 Rhodamine B: 85 Evans Blue: 90 Cytochrome: 97	1084
Self-cross-linked Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> -based membranes	NaCl: 98.6	0.0515
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	Methylene blue: $98.0 \pm 0.9$	10.6
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /poly(vinyl alcohol)	Methylene blue: 97.2 $\pm$ 0.8	7.4
MXene slit membranes (molecular dynamics simulation)	Monovalent salt ions: 100	400
Modified after Ihsanullah (2020)	· · · · · · · · · · · · · · · · · · ·	

**Fig. 11** Chemical structure (a) and transmission electron microscopy image (b) of fullerene. Reprinted with permission of Manawi et al. (2016)



Transportation of water molecules was investigated through nanopores of modified membrane.  $C_{80}$  fullerene was used to fabricate polymerised fullerite membrane, also NaCl considered as salt with concentration of 37.125 g/L. The simulation results revealed a superior water permeation flux and ion removal for the modified membrane, and these values were higher than those for previously reported membranes. The temperature of desalination influenced the dipole orientation and collective water motion and consequently water permeation from membrane. In another study, Foroutan et al. (2019) encapsulated fullerene by CNTs and studied its performance for water permeation and ion rejection by molecular dynamics simulation. They emphasised that ion rejection was approximately 100% in this modified nanochannel. In addition, the obtained water permeation flux ( $16.22 \pm 0.85 \times 10^{-19} \text{ m}^3$ /s) was higher than

 Table 6
 Rejection and

 permeation percentages or flux of
 some fullerene-based membranes

Type of membrane	Rejection percentage	Permeation percentage or flux	References
Fullerene-based sulphonated polyvinyl alcohol nanocomposite membrane	Cu <sup>2+</sup> : 73.2%	-	Keçili et al. (2020)
Fullerene (10 wt%)-based poly (2,6-dimethyl-1,4-phenylene oxide) membrane	Estrone: 95%	-	Keçili et al. (2020)
Fullerene-based sulphonated polystyrene	_	Methanol: 50% lower than polystyrene membrane and 30% lower than Nafion@117 membrane	Keçili et al. (2020)
Fullerene-based polyphenylene isophthalamide membranes	-	Water: 99.9%	Pulyalina et al. (2020)
Fullerene (5 wt%)-based polyphenylene isophthalamide	-	Methanol: 84–241 g/m <sup>2</sup> h	Dmitrenko et al. (2019)
Polyhydroxylated fullerene-based thin-film nanocomposite membrane	Na <sub>2</sub> SO <sub>4</sub> : 95.6% MgSO <sub>4</sub> : 93.6% MgCl <sub>2</sub> : 89.9% NaCl: 22.6% LiCl: 16.3%	Water: 6.7 L/m <sup>2</sup> h bar	Shen et al. (2019)

that of natural channels. Different hydrostatic pressures were applied, and the optimal results are obtained under 5 MPa. The applied forces on the fullerene by water molecules and interaction between fullerene and the wall of CNTs influenced the fullerene motion through CNTs channels. Liu et al. (2018) used C60 molecules to improve the desalination performance of GO membrane. They grafted fullerene on the GO surface by lithiation reaction, and consequently, the water permeation flux of prepared membrane increased compared to GO membrane. NaCl rejection rate is also reported by about 89.66% for this modified membrane.

According to the aforementioned results, it can be concluded that fullerene is an excellent candidate to improve the performance of conventional membranes and other carbon-based nanomaterials in water desalination processes.

In conclusion, carbon-based nanomaterials enhance water permeation flux and ion rejection of pristine membranes, which are the main goals of water purification and desalination processes due to their porous structure, high surface area, low surface energy, excellent chemical/physical stability and low cost.

#### 9 Conclusions

The lack of fresh water has become a major problem in the present and future, while water covers about 71% of earth's surface. Therefore, the use of seawater or polluted water and determining useful materials for water purification are

urgent. Desalination of saline water is an attractive solution for this problem, especially by membranes. Membranebased desalination methods are environment-friendly with low cost and high efficiency. In total, these porous materials pass water and sieve ions. The pore size and thickness of membranes can influence their performance in water permeation or ion rejection; moreover, the membranes have some disadvantages like fouling. Therefore, the membranes were developed by researchers. In this regard, nanomaterials especially carbon-based nanomaterials have attracted a lot of consideration. The excellent chemical/physical stability, porous structure, low density, antifouling feature, high surface area, antibacterial property and low surface energy are the distinguished properties of carbon-based nanomaterials (CBNMs). The valuable studies have investigated the performance and application of CBNMs to improve the performance of conventional polymeric membrane.

This chapter reviewed several current published studies to reveal the important roles of CBNMs on the desalination performance of membrane and other CBNMs. Graphene and its derivatives, carbon nanotube, carbon nonfilter, fullerene, MXene and activated carbon are familiar CBNMs, which can enhance water permeation and ion rejection of membranes. Nanoporous structure of graphene and its derivatives has made them one of the best choices to modify pristine membrane and improve desalination performance of other CBNMs like carbon nanotubes. Surface modification of graphene, reduction of graphene oxide or adding some

functional groups to it can increase desalination efficiency. However, some researchers have believed that water permeation flux of carbon nanotubes (CNTs)-based membranes is higher than that of graphene-based one, which can be obviated by optimising the pore radius of graphene membrane. CNTs are ever appropriate option to modify pristine membranes, but moving water molecules through their pores depends on the interaction between water molecules and internal surface of CNTs and also CNT fabrication method. Vertical CNTs have a better desalination performance than mixed matrix types. Fouling of CNT-based membrane does not occur, unlike regular membrane processes. On the other hand, the required pressure for this type of modified membrane is negligible compared to the pristine one. Sometimes, CNTs are modified by some chemical methods, which enhance their desalination efficiency. Carbon nanofibres (CNFs) and activated carbons are mostly used in capacitive deionisation desalination process because of their capacitance. CNFs can ameliorate the specific surface area, capacitance, stability, salt capacity, deionisation time, surface hydrophilicity and electrode efficiency compared to conventional polymeric membranes in capacitive deionisation methods. However, surface hydrophobicity of activated carbons is the main problem in capacitive deionisation desalination. Therefore, wettability of activated carbon-based electrodes is increased by reduction of electrode hydrophobicity. Fullerene and its derivatives are another fetching candidates to improve the performance of membrane processes, which cause an increase in water permeation flux and salt rejection. In addition, they can increase the ion rejection percentage by CNTs to approximately 100%. Finally, MXene, emerging 2D metal carbides with exceptional properties, was reviewed as advanced membranes. Titanium-based MXenes exhibit high water permeation rate and ion rejection for different sizes and charges of ions.

This review discussed that using CBNMs can improve water permeation flux, ion rejection, salt selectivity, durability, and decrease in fouling of membranes in water purification and desalination processes. This information is potentially essential for future studies and researchers to improve membrane processes and to solve the freshwater scarcity problem.

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# **Microbial Desalination**

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# Abstract

Several investigations have established that due to population expansion and land use, fresh water scarcity has increased tremendously placing immense burden on desalination process as the major alternative remedy to freshwater supply. The increase in problems related to energy across the globe due to exhaustibility of fossil fuels. Hence, there is a need to search for other sources of energy to meet up with the growing demand. Globally, the existing methods of desalination are not cost effective; hence, scientists are searching for alternative ways to reduce the financial burden in the setup of water desalination techniques. One of the suggested areas is in the utilization of microorganisms. One of the suggested methods is in the area of energy conservation, when using water to produce energy via utilization or engaging the use microbial desalination. Therefore, this chapter intends

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to provide comprehensive information on the application of microorganisms for the water desalination. Several types of microorganisms that could be applied for water desalination were also highlighted. The modes of action through which they exhibited their action were also discussed. The principles involved in the process of desalination were also elucidated.

#### Keywords

Microbial desalination • Modes of action • Techniques • Water scarcity • Desalination techniques

# 1 Introduction

Several people have witnessed acute shortage of portable water in many parts of the world mostly due to salt contents and contamination of oceans, lakes, and rivers (Shatat & Riffat, 2014). Thus to tackle this situation, water desalination techniques are deployed to satisfy human and animal water requirements. Studies have revealed that desalination is commonly utilized as an alternative means of providing drinking or portable water in areas that are experiencing scare supply of clean water. Water scarcity is estimated to affect one-fifth of the world's population, and this condition is predicted to be worsening due to many factors such as population explosion, climate change, industrial and household usage, and urbanization. The method of desalination can be deployed for wider range of primary source of water like seawater, groundwater and estuarine water containing inorganic ions, borate, bromide, sodium, potassium, and iodide of potential concern to humans (Cao et al., 2009).

Over the years, different technological advancements have been witnessed in the methods of water desalination such as the conventional methods like thermal, reverse osmosis, electrodialysis reversal systems, to the advanced method such as microbial-based desalination bioelectrochemical systems

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(BES) that uses bacteria to generate renewable energy as electricity, methane, and hydrogen. The conventional methods are more energy-dependent and expensive compared to the advanced method. Microbes in the anaerobic salty soil wetlands transform exudates in the roots of plants or dead plant material into  $CO_2$ , electrons plus protons. These products can be harvested via cathode and anode connections and converted to energy source. The energy can be harnessed as a potential desalination technique plus power generation utilizing bacteria and biodegradable organic matter. The microbial fuel cell is known to produce current by degrading microorganisms forming organic compounds and equally utilized in the treatment of waste waters (Xincao et al., 2009).

Studies have shown that due to population expansion and land use, fresh water scarcity has increased tremendously placing immense burden on desalination process as the major alternative remedy to freshwater supply. It has been shown that 18,000 desalination plants are been used globally producing average rate of 86.8 million m<sup>3</sup>/day. The large development seen in desalination technology is mainly due to the rapid expansion and improvement in desalination technology with energy requirement and lower cost of production (Gede Wentena, 2016). Surva et al. (2015) have revealed that there is increasing rate of energy problem across the globe due to exhaustibility of fossil fuels; hence, researchers are beginning to harness other sources of energy to meet up with the growing demand. One of the suggested methods is in the area of energy conservation, when using water to produce energy via utilization or engaging the use microbial desalination. The authors have revealed that wastewater-based three-chambered microbial desalination cell could solve both energy and water crisis offering greater efficiency than the other such as thermal distillation, reverse osmosis, freezing, and electrodialysis.

Globally, the existing method of desalination is not cost effective; hence, scientists are searching for alternative ways to reduce the financial burden in the setup of water desalination techniques. One of the suggested areas is in the utilization of microorganisms such as *Saccharomyces cerevisiae* yeast to provide a sustainable energy supply, reduced toxic generation, and water desalination process with reduced cost. Though commercialization of this technology has suffered tremendous setbacks due to several factors like pH instability, low current output, and membrane biofouling plus polarization, scientists still believe that if all these challenges are resolved, microbial desalination holds a promising potential in the near future (Yahiaoui et al., 2020).

Therefore, this chapter intends to provide a broad fact on the use of microorganisms for the water desalination. Several types of microorganisms that could be applied for water desalination were also highlighted. The modes of action through which they exhibited their action were also discussed.

# 2 Principles Involved in the Process of Desalination

The principles of desalination centres on the doctrines establishing a technology for large-scale desalting of water. Some of the already established procedures include reverse osmosis, vapour reheat distillation plus hyperfiltration extremely important for water treatment during water shortage and contamination. Rozendal et al. (2008) revealed a new approach in water desalination using microbial-based techniques and also generated electrical current from the organic matter plus bacteria in another solution representing (anode). The authors showed that reduction in the salinity may require greater amount of anode solution; hence, the use of membranes will further increase ion exchange capacities. Thus, it was concluded that desalination of water is conceivable using equivalent amount of anode solution plus salt water.

The basic principle of desalination involves the removal saline water source plus devotes energy to two distinct streams, one with salt-free fresh water and salty brine. A reliable and proper desalination technology utilizes lower quantity of energy to generate a huge volume of fresh water plus small amount extremely concentrated brine. In a study, the performance of single- and double-membrane microbial desalination cell was used to investigate the operation mode flow in water desalination. The authors revealed that there was increased generation of energy after the cycle; hence, the technology is eco-friendly, safe, and cost effective for desalination (Cheng et al., 2006). Another study by Zhang et al. (2010) revealed that analyte solution in the microbial desalination cell acts as buffer, catalyst, and substrate for the microbes. Studies have revealed that two kinds of microbes are utilized in this process, those using synthetic facilitated electron transfer and those that do not (exoelectrogens).

APHA (1998) have revealed that traditional wastewater purification methods are energy consuming and lot of resources wasted in the cleaning process. Scientists hence have developed microbial nutrient recovery cell technology as a desalination technique and also regenerate lost nutrients in waste water management. This technique has proven to be efficient, self-stimulated approach and sustainable wastewater purification strategy using bioelectrochemical system. Zhang and He (2012) described the microbial fuel cell-based technologies as emerging innovations for the applications in bioremediation, seawater desalination, biosensors plus hydrogen production. **Microbial Desalination** 

# 2.1 Air Cathode Microbial Desalination Cell

In air cathode microbial desalination cell, the most usually used oxidizing agent in cathode chambers is oxygen provided by bubbling air via water in the cathode chamber. The process involved in the different microbial techniques for desalination and principles involved were elaborated in Table 1

## 2.2 Biocathode Microbial Desalination Cell

Cheng et al. (2006) demonstrated the utilization of biocathode in cathode chamber, by engaging microbes as oxidizing agents.

#### 2.3 Stack-Structure Microbial Desalination Cell

In this arrangement similar to stacked electrodialysis cells, series connections are made in the cathode and anode sections. In the operation, ions move out from desalination chamber to freshwater chambers, forming one dilution section plus two concentrated sections (Chen et al., 2011).

# 2.4 Microbial Electrodialysis Cell

In this classification, microbial desalination cells are reorganized to generate  $H^+$  from the cathode compartment while desalination process is going on (Mehanna et al., 2010). 2.5 Microbial Electrolysis Desalination and Chemical-Production Cell

In this setup, acids and alkaline solution are generated during desalination. Application of large quantity of current will induce a potential difference and create water splitting into hydrogen and hydroxyl ions.

#### 2.6 Microbial Capacitive Desalination Cell

In microbial capacitive desalination cell, stimulated carbon membrane associations are used to absorb deionized ions and stored in electrical double layer capacitors. Electrical neutrality is the hallmark of this setup via the free movement of cations during desalination.

# 2.7 Upflow Microbial Desalination Cell

This arrangement is made up of fitted cylindrical sectors containing analyte solution and anode electrode. Oxygen is been utilized as electron acceptor due to exposure to air (Zhang et al., 2010).

# 2.8 Osmotic Microbial Desalination Cell

In osmotic microbial desalination cells, desalination plus dilution concept is into practice utilizing osmosis combined with electric potential gradients which assist in lowering salinity in the desalination chamber.

**Table 1** Different microbialtechniques for desalination andprinciples involved

Microbial technique	Principle	Authors
Bioelectrochemical system		APHA (1998)
Microbial fuel cell	Activity of microbe	Zhang and He (2012)
Biocathode microbial cell	Engaging microbes as oxidizing agents	Cheng et al. (2006), Croese et al. (2011), Al-Mamun et al. (2018), Wen et al. (2012)
Stack-structure microbial cell	Electrodialysis	Chen et al. (2011)
Microbial electrodialysis cell	Electrodialysis	Mehanna et al. (2010)
Upflow desalination cell	Utilization of oxygen as electron acceptor	Zhang et al. (2010)
Microalgal biocathode	Utilization of high carbon by Chlorella vulgaris	Zamanpour et al. (2017), Powell et al. (2009), Wang et al. (2020)
Bacterial biocathode	Transformation of CO <sub>2</sub> into carbon derivatives using acetogenic bacteria	Saeed et al. (2015), Zaybak et al. (2013)

# 2.9 Submerged Microbial Desalination-Denitrification Cell

In this arrangement, combined direct nitrate elimination and desalination exist in groundwater desalination using two chambers of cathode and anode. Nitrate ions move as a result of electrical potential generation into the anode chamber and later reduced by denitrifying bacteria.

# 2.10 Principles Entailed in the Process of Desalination

The principle behind the procedure involved in desalination entails the organic migration of waste water to the anode compartment where there is increased microbial population and creation of biofilm, and in the course of the degradation of biological matters, there is production of electricity (Elimelech & Phillip, 2011; Lovley, 2012; Yuan et al., 2017). Electrons that migrated to the cathode produced electrostatic energy when present in water that is salty propel the anions near the positively charged and negatively charged electrode (Kim & Logan, 2013). This process provides double advantage of treatment of waste and production of bioelectricity (Fritzmann et al., 2007; Sophia et al., 2016).

There are two primary processes that are mostly used in desalination, that are;

- i Thermal desalination process
- ii Membrane desalination process.

**Thermal desalination process**: This desalination process is also known as distillation. It is a primitive way of converting sea water into drinkable water. This method is widely used because it is usually cost effective. This application mode is centred on the boiling of salty water and allowing it to evaporate then the condensed water would be collected to obtain pure and drinkable water (World Health Organization, 1993). These entail the following:

**Multi-stage Flash Distillation**: This entail boiling of water speedily when force of vapour exists quickly lowers beyond the vapour pressure of the fluid at a specific temperature. Subjecting the water to heat remains foremost to it being introduced into the flash compartment, the steam produced is compressed to the surfaces that are in interaction with the fresh water. Multi-stage flash is usually simple to build and easy in functioning; the plant is also referred to as the energy intensive process, owing to the fact that it entails both heat and mechanical energy.

**Multi-effect distillation**: This process occurs in succession of vessels and utilizers. It operates on the principles of evaporating and condensation at lowered standard pressure. This process operates at reduced temperature of 70 °C,

leading to reduced corrosion of cylinder and the establishment of scale on the cylinder surface.

**Vapour compression evaporation**: This process does not use steam heat. The heat used in evaporating the feedwater is gotten from the density of the vapour and not through uninterrupted heat exchange from stream released during a boiling process.

**Cogeneration system**: This is a process in which energy source can function differently doing various things, for example, production of electrical power and removal of salt from water sample. This process entails the production of high-pressure steam to function in the turbines and the steam is formed when boiled at 5408 °C. There is reduction in the temperature and energy level when the vapour expands the turbines.

**Renewable energy distillation/solar water desalination**: This process is used for small-scale setups. Energy gotten from the sun is utilized in evaporating renewed water from salty water. Steam gotten during this process condenses on a transparent glass or covering and is latter obtained as fresh water from the condensate. This process is usually adopted in parched areas where there is lack of fresh drinkable water (Bodzek, 2000; Worldwide Desalination Research and Technology Survey, 1994).

**Membrane process**: Membrane process utilizes penetrable film to mobilize either water or salt to form two zones of varying concentration to form fresh water. This is usually employed in community waste water treatment. This could be categorized into the following.

**Reversed osmosis:** This method utilizes pressure as propelling force that pushes the water via a temporarily penetrable sheath into a concentrated saltwater. Practically, the feedwater is being pumped into a confined vessel, in contradiction to the membrane, to pressure it (Logan et al., 2006). As the water product passes via this membrane, the feedwater and brine solution become extra and extra concentrated. A reverse osmosis consists of four major process which are;

- (i) Pretreatment system.
- (ii) Pressurization.
- (iii) Membrane separation.
- (iv) Post-treatment stabilization.

**Pretreatment**: Feedwater entering was being pretreated to be well matched by the members by eliminating suspended solids, regulating pH, and addition of a threshold eradicators to regulate scaling initiated by some elements like calcium sulphate.

*Pressurization*: The pump elevates the force of the pretreated feedwater to a functional pressure that is suitable for the film and the saline nature of the feedwater.

*Membrane* separation: The penetrable membrane hinders the path of liquefied salts while allowing the removal of salt from produced water to go through. **Post-treatment stabilization**: The product water gotten from the film assembly normally needs pH adjustment and the removal of gas previously and then transferred to the circulation system as drinking water. The product goes through a ventilation column where the pH is raised from a value of about 5 to a value near 7, which make the water drinkable and useable (Logan et al., 2006; Kutty, 1995).

#### Electrodialysis

This electrodialysis process contains film with positively charged ions and anions exchange groups. In the pressure of straight current electric field, positively charged ions and anions move to their corresponding electrodes so that there will be formation of ion-rich and ion-exhausted streams in other places within the membranes (Logan et al., 2006; Furukawa, 1999).

#### **Membrane Distillation**

The principle here is the use of thermal and membrane technologies. Here, the variation in the temperature is formed amongst the supplied solution that is to contact the surface on the side of the micropermeable sheath and there remains left over space on the other side. Variation in the temperature leads to the variation on the vapour pressure and causing migration of vapour formed via the membrane on the concentration surface (Alabdula'aly & Khan, 1997).

#### Secondary/Alternative process

This method can be tackled in two different ways which are freezing and ion exchange. In the course of freezing, liquefied inherent salt in the feedwater are divided in the period of production into crystals below measured conditions. Before the entire water is freezed, the mixture is washed and rinsed, eliminated the salt in the remaining water while, for ion exchange, organic and inorganic solids are found to interchange one-type cation or anion restrained on the surface of another kind of cation found in the solution (Logan et al., 2006).

#### (A) Types of microbial desalination

**Osmosis MDC**: Osmosis microbial desalination cell was developed during research by replacing anode exchange membrane with osmotic membrane. In this system, water is pressured out of salty water due to the increased osmotic pressure of the fundamental compartment and ions are circulated round the membrane. In respective of the diluted salt water in the middle compartment, lack of membrane selective paths brought about the ability of unwanted ions to deter the effectiveness of the microbial desalination cells (Zamanpour et al., 2017; Zhang et al., 2012).

Zhang and He (2015) stated that this system is a distinct type of membrane because it exploits the osmotic liquid movement from the positively charged electrode to the salty water, thereby allowing dilution process to take place and desalination of the water sample (Zhang & He, 2015; Zhang et al., 2011; Zhao et al., 2006). This kind of microbial desalination, where there is separation of the positively charged electrode compartment and middle compartment by AEM through a forward osmosis membrane (FOM). The forward membrane permits the passage of liquid and decreases transmission of ions from the central compartment of anode then the cathode compartment (Kim & Logan, 2013; Zhao et al., 2006). They found out that the salt was really not eliminated but became concentrated. They also discovered from their study that high-energy method of osmotic MDC had an outcome of greater water desalination effective in the generation of power via salty water and reduced conductivity (Saeed et al., 2015).

Osmotic MDC is widely known in performing these key objectives, which are; anode exchange membrane is replaced with forward osmotic membrane thereby improving the dilution of salty water (desalination), dilute salt water by increasing water flux thereby enhancing the remover of biological matter from waste water, potassium ferricyanide is utilized to speed up reaction allowing this forward osmotic membrane to be cost effective compared to the anode electrode membrane (Zhang et al., 2012). Although forward osmotic membrane was found to be more susceptible to fouling that ion exchange membrane, with a consequence of escalation in the internal osmotic resistance thereby reducing water flux (Kim & Logan, 2013).

#### Biocathode MDC

Biocthode MDC is an advanced system of electrodes which encourages the reduction in electrochemical reactions and prime catalysis is done by microorganisms (bacteria and algae). Microorganisms perform reduction presses in the cathode or present on electrode (Croese et al., 2011). This type of MDC does not need high catalyst of its lower maintenance, self-regenerating properties and production cost (Al-Mamun et al., 2018; Zhang et al., 2012). The bacteria performed as catalyst in the compartment because they were active to electrode and causing oxidation reduction reaction resulting in the increase in the removal of salt from water (Wen et al., 2012).

There was advancement in biocathode microbial desalination cell which comprises of the use of cathode, middle, and anode compartment. This reaction was related to microbial fuel cell only that middle compartment remains filed with salty sea water which initiates the desalination reaction by potential gradient amid the anode and cathode, and then fresh water is produced from the sea water owing to migration of ions to the negatively charged and positively charged electrode (Brastad & He, 2013). The biocathode is subjected to optimal conditions; start-up time of the microbial desalination cell could be remarkably reduced, consequently increasing the general performance of the cell (Luo et al., 2017; Tchobanoglous et al., 2003).

Nitrifying and denitrifying bacteria and algae could be used as biocatalyst in biocathodes because they can produce electron acceptors at negatively charged electrode which is needed in reducing reaction (Clauwaert et al., 2007). Some microalgae biocathodes could remain used in appropriating nutrients that were missing and melted biological substance in order to produce bioenergy by the microalgae biomass (Arana & Gude, 2018; Gude, 2016). Literature has shown that specialized microorganisms which are known as electrotrophs possess the capacity in gaining electrons directly of via the negatively charged electrode, thereby exploiting various routes for electron acceptors for example iron, carbon dioxide,  $O_2$ , or nitrate (Saeed et al., 2015; Zaybak et al., 2013).

**Microalgal biocathode MDC**: *Chlorella vulgaris* is high and accessible (microalgae) was found to utilize high carbon content such as bicarbonate and carbon dioxide to produce oxygen under favourable condition leading to the production of solar power (Powell et al., 2009; Zamanpour et al., 2017).

**Bacterial biocathode MDC**: The use of bacterial biocathode microbial desalination as electron in biocathode is a complicated and strenuous method. However, anaerobic facultative autotrophic biocathodes have proven to be more effective and virtually achievable method established via pre-improvement of heterotrophs (Zaybak et al., 2013). This deviceallows  $CO_2$  to be transformed into carbon-based derivatives via replacing hydrogen by negatively charged electrode that serves as source of energy and source of electron source with the aid of acetogenic bacteria (Saeed et al., 2015).

#### Staked MDC

Staked microbial desalination is exceedingly convenient owing to their level of productivity in recovering more energy when equated to other microbial desalination system and is inexpensive. In these setup, bacteria oxidizes the biological matter in the anode compartment thus leading to extraordinary retrieval of energy (Shehab et al., 2013).

Researchers sated that this system could be upgraded by multiple sets of ion exchange membrane inserted amongst the cathode and anode compartment, so as to improve the effectiveness of charge transmission and then permit the highly saline water to run across sequence of microbial desalination cells that supports the removal of more salt (Al-Mamun et al., 2018; Gude et al., 2013).

Subsequently, stacked arrangement operates based on bioelectrochemical response, whereby fluctuating of systems plus functioning strictures like assembly of electrodes, whichever sequence or parallel and hydraulic movement methodologies may disturb the procedure of salt removal (desalination). Disruption of pH amongst the cathode and anode compartments could affect the desalination process when more than one compartment is used. When there is great decrease in the pH level at the anode level, this could lead to reduced performance of the microbial population in the compartment of the positively charged electrode, but an increase pH level in the compartment of the negatively charged electrode compartment may result in great loss of potency and reducing the general course productivity (Cheng et al., 2010).

#### **Upflow MDC**

Upflow cell is exceptional kinds of cell where mixtures of solution inside the compartment can be accomplished deprived of shaking and recover 100% water (He, 2011; Qu et al., 2012). This technique enables microbes present in the positively charged compartment to stay in the supernatant method and effectively perform extreme oxidation of biological material (He et al., 2006; Saeed et al., 2015). Promoter (catalyst) utilized in the upflow microbial desalination cell is combination of carbon and platinum; this mixture deposits on the exterior layer (Jacobson et al., 2011). The upflow system is made up of cylindrical unit which is comprised of twofold sections where internal section full of pellets from graphite then delivered an advanced superficial part for oxidation reaction to be done. Two bars of graphite were absorbed as current collectors in the graphite granules which are known to tolerate conduction of electrons. The external unit which contains salty water symbolizes desalination compartment which was extra secured by cation exchange sheath cylinder, thereby allowing superficial part of the amplified to take part in the desalination process from stage to stage (Jacobson et al., 2011). The benefits of upflow are that there is usually effective fluid mixing within the compartment, very easy to scale up, and this system improves desalination.

#### Photo-MDC

This system improves energy yield of microbial desalination cell by employing sunlight potential as an initiator without increasing the cost of functioning. Heamatite nanowire photo-anodes are some of the examples of this type of microbial desalination cell and are known to be useful in proficient setup (Qian et al., 2014) nanowire-bacteria hybrids for unassisted fixing of solar energy CO<sub>2</sub> (Liu et al., 2015; Zang et al., 2014). Researchers have presented high-performance photo-microbial desalination cell where the anode is improved with nanostructure. Then, they perceived that there was maximum density of the current of the photo-MDC in the course of this method remained at 8.8  $Am^2$  double the unmodified microbial desalination cell by means of 20 g/L original concentration of briny. Despite the concentration of salt discovered in the central compartment remained beneath 1.4 mg/L of waste, remover of salt was 96%. There could be greater attraction in the combination of MDC with biophoto-electrochemical cell.

#### **Capacitive MDC**

This type of microbial desalination cell is engrossed on the usage of carbon materials with greater exterior area at two electrodes. There is usually likelihood of variation between the permeable anode and cathode. Capacitive deionization as technologically advanced into two subsequent techniques which are desorption and adsorption. Here, the ions were primarily separated from saline and desalination of water concurrently takes place. Moreover, the adsorption techniques show two electrical layers made together on charged cathode and anode owing to migration of ions separated from water sample. Then, there is substitution between the electrodes present in the solution, and there is total discharge of zero voltage by the electrode, there is supply of energy, and ions were released into solution producing unwanted water steam (Wen et al., 2012).

Researchers stated that there was formation of doubled layer capacity on the high surface area of electrodes, and there was adsorption of ions when salt solution was placed amongst the anode and cathode. The ions found in the saline were adsorbed on exterior areas of the electrodes by dualcoating capacitors, as soon as the potential gradient was disconnected, ions were allowed to return into the solution. There was immediate deionization of the salty water irrespective of the pollution of the cathode and anode compactly through the salt (Saeed et al., 2015; Santoro et al., 2017; Zhang & He, 2012); this type of membrane was developed by the introduction of membrane capacitive deionization which is cationic selective membrane incorporated on the negatively charged electrodes and anions exchange membrane was incorporated on the positively charged electrons (Zaybak et al., 2013).

One main stated flaw in the performance of this system is increased concentration of salt anolyte and catholyte. The result in this buildup of ions alters pH of the catholyte and the anolyte resulting in reduced metabolism of the microorganisms. Therefore, there would be need for replacement of catholyte and anolyte and reapplication of water (Al-Mamun et al., 2018; Forrestal et al., 2012; Saeed et al., 2015).

#### **Bipolar membrane MDC**

In bipolar membrane system, desalinate water also generate hydrochloric acid as well as sodium hydroxide concurrently. This system consists of a cathode exchange membrane and an anode exchange membrane layered together to produce a single membrane. When subjected to appropriately high electricity potential variation, water splits to protons and hydroxyl ions by the border of the layered ion exchange membranes, thereby resulting in the discharge of hydroxl ions in the direction of the anode compartment and the protons close to the side where there is combination through chloride ions entering from the middle section to form the HCl (Forrestal et al., 2012; Buck, 2014).

This reaction is crucial in evading reduction of pH in the anode compartment, thereby undertaking a cogent role in the microbial desalination cells (Forrestal et al., 2012). The limitation posed on the MDC could be dealt with by the use of exterior voltage to manoeuvre the bipolar membrane. Studies have shown that the least voltage needed to separate

water is 0.83 V but, practically, the essential voltage surpasses 1.2 V; then, it could be concluded that the possible variation produced by MDC only is insufficient to measure up to the condition of bipolar membrane functioning and exterior voltage of approximately 1 V is required to function properly. Bipolar membrane is more prone to biological pollution due to their contact to saline in the anode compartment (Kim & Logan, 2013; Mehanna et al., 2010). Splitting of water at the bipolar membrane is intricate in removing salt from water. The degree of saltwater is also great due to the cost of preservation of anolyte pH (Chen et al., 2012). This system has high ion selectivity, long-lifespan membrane of biofilm, increased level of salt remover, and maintenance of preferred pH in the anode compartment.

#### **Recirculation MDC**

Recirculation microbial desalination cell is a system where solution of anolyte and catholyte is usually uninterruptedly recirculated via the MDC in order to counterbalance pH value (Luo et al., 2011). Numerous investigations have been performed with the objective of eliminating the inequity of pH in the microbial desalination cell compartments. These include adding of buffers and the initiation of increased volume of anolyte (Al-Mamun et al., 2018). Researchers have shown that the recirculation of catholyte and anolyte has encouraging energy production and salt water removal. Recirculation system functions having 50 mM of phosphate buffer generated 335 extra power; hence, at ambient temperature and favourable situations, cell worked with at 25 mM produced 53% additional power (Luo et al., 2011). Therefore, increased buffer concentration is not very important for the improvement of the density power, so then, an optimum concentration should be acknowledged.

One main difficulty in this system is the hindrance of the membrane in cooperation with hydroxyl and proteins manufactured during redox reaction, in cathode and anode compartments (Kim & Logan, 2013). Oxidation of the biological matter discharges protons that are not capable to migrate to the cathode compartment, while hydroxyl ions are formed by means of the reduction reaction (Luo et al., 2011; Qu et al., 2012). This may result in frequent pH imbalance within the cell, where there is increase in pH in the cathode and reduction in the anode.

#### Modes of action of microbial desalination

The mechanism of action of desalination is established on the relationship amongst living microscopic entities and electrodes via the reduction of biological matter (Logan et al., 2017; Schröder et al., 2015). Microbial desalination cell exploits the possibility variance generation from oxidation made by anode producing bacteria to initiate the movement and removal of salt ions. Overtime the bacteria utilize the biological matters in the waste water thus, making the clean. Electricity is produced and water desalination is done concurrently (Xiaoxin et al., 2009; Schröder et al., 2015; Sevda et al., 2015).

The rate at which desalinated cells work is being quantified by its efficiency in salt removing ability Jacobson et al. (2011), overall salt removing rate (Chen et al., 2012) specific desalination rate (SDR) current density (Jacobson et al., 2011; Qu et al., 2012); removal nitrogen (Zhang & He, 2015; Zuo et al., 2017), removal of biological materials (Luo et al., 2017) Faradaic potential, pH discharge and the anode in the microbial environment (Qu et al., 2012).

Forrestal et al. also explained the mode of action of microbial desalination by stating that the setup is comparable to that of a MFC which comprises of dual compartments with cathode, two electrodes, and anode. Then with the third compartment which is divided by an anion exchange membrane, cation exchange membrane, a marginal, peripheral circuit, that is accountable for the anaerobic and aerobic and procedures at both corresponding electrodes. There is increase on the carbon-based matter in the anode compartment thereby leading into the creation of biofilm that produces an electric current. The pollutants in the slush are then oxidized by the aid of the biofilm via stringent adherence to anode, releasing together the electrons and the protons from the bio-slush and the production of current of atoms by circuit conveyance that are composed of electrodes. Electrical current is then created by the probable variance produced amongst the anode and cathode owing to the presence of  $O_2$  in the cathode compartment (Forrestal et al., 2012).

Sevda et al. (2015) also explained the mode of action of microbial desalination by stating that the anode cavity and the cathode cavity are usually parted by ion exchange membrane. Desalination cavity is made when the ion exchange membrane makes a border on the salt solution. Organic materials in the waste water go into the anode part where formation of biofilm takes place owing to the increased microbial load that oxidizes organic material, causing in the fabrication of electrons at the electrode and liberating protons into the solution (Chen et al., 2011). The electrons formed migrates via an exterior resistance to the electron acceptor at the cathode, water is produced.

Microbial desalination is a unique technology that utilizes energy gotten from the metabolism of electroactive microorganisms usually bacteria and algae at the process of degraded biological material in order to provide ecological safe water (drinkable). This process allows various techniques to take place concurrently. This process includes desalination of water, waste water treatment, and also the creation of electricity. Microbial desalination Cells (MDCs) comprise of an electrochemical material that has three compartments (Cao et al., 2009).

The anode part consists of an electrode which is usually enclosed with a biofilm which oxidizes the biological materials present in the waste water, thereby transmitting electrons from the biological material (substrate) to the electrode. Thereafter, the electrons utilize an exterior circuit locate the area where the cathode resides; this is where reduction reaction is done. There is usually movement of ions imposed by electric potential. Consequently, desalination initiated when positive ions migrate via the cathode and the negatively charged ions then migrate within the anion exchange membrane from the salty area to the anodic area (Wen et al., 2012). Cao et al. in their study used 9 cm<sup>2</sup> cross section of a cell with the aid of salty volume compartment of 11 mL; there was 90% elimination of salt (Cao et al., 2009).

Several researches have shown that cubic along with tubular devices, amassed cells in the course of using batch recirculation, and biologically produced cathodes increases the release of water due to the application of exterior voltage (Chen et al., 2011; Mehanna et al., 2010a, Kim & Logan, 2013a; Jacobson et al., 2011a). This could also be achieved by forward osmosis, interchange of ion resin in the partitions, or through microfiltration procedures (Lovley, 2012; Zhang & He, 2012; Zhang et al., 2010, 2012; Zuo et al., 2017).

The reaction that happens in the cathode is one of the most crucial reaction phases in this setup. Studies have shown that applying oxygen reduction method in the cathode section has been of greater benefit by utilizing the oxygen as electron acceptor (Lu & Li, 2012). Zhao et al. (2006) in their studies stated that there are three primary things that disrupt the proper activity of cathode which are pH, concentration of the catholyte, and the lead (Pb) of the catalyst. The reduction of oxygen in diverse pH environment is usually made possible by the present Pt, gold, and silver metals which is often as promoter in electrochemical devices (Ge et al., 2015). Liu et al. (2015) revealed decreased in the operational oxygen in the system having used MnOx as a substitute catalyst instead of using the former costly metals (Lu & Li, 2012; Zhao et al., 2006). Additional transitional metals were also worked on by researchers (Wang et al., 2015; Zhang et al., 2016) amongst other metals (Vij et al., 2017), added that iron was used also as the other properties where altered (Vij et al., 2017; Yuan et al., 2015b).

Cao et al. (2009) used ferricyanide catholyte as electron acceptor in the growth of microbial desalination cells. There were about ninety-four 94% elimination of salty water and  $2 \text{ W} \cdot \text{m}^{-2}$  production of energy was attained, thereby meaningfully increasing the activity of the system when compared to when osmosis reverse is used in the cathode reaction. However, owing to how expensive the reagents are, the application of ferro-ferriccyanide catholyte would be possible in the technical approach if the redox mediator is less expensive and easy approach was implemented for the

rejuvenation of the catholyte solution as soon as it is exhausted. There could be the implementation of two different approaches to establishment of microbial desalination cell (MDC) innovation in producing drinkable and eco-friendly water using only low energy. These methods are:

**Operation of MDC by the use of reduced oxygen in cathode reaction**: Operating the microbial desalination by the use of reduced oxygen in cathode reaction is usually set up as air dispersion cathode prepared of carbon nanofibres and Fe nanoparticles as catalyst is usually appropriate and less expensive electricity for eco-friendly uses.

**Operating the MDC by the use of Ferro-ferricyanide redox pair in cathode reaction**: This method employs one ferro-ferricyanide redox catholte as a substitute to oxygen reduction. This was done in order to improve the obtainable potential in the microbial desalination cell thereby allowing enhanced functioning.

Specific examples of authors that have utilized microbial desalination for the management of water

Alhimali et al. (2019) highlighted the importance of microbial desalination cell which is a microbial electrochemical system as a proficient means of utilization in desalination with lower energy consumption. Recently, more attention has been focused on microbial desalination cell; hence, the authors investigated bioelectricity generation plus behaviour of ions transportation in microbial desalination cell. From the results obtained, they suggested that optimization of the salt transfer with ion diffusion coefficients is needed to boost microbial desalination cell performance.

Carmalin and Bhalambaal (2016) revealed that microbial desalination cells are a modified version of microbial fuel cells that utilize organic compounds from exoelectrogenic bacteria in wastewater to generate energy. The authors showed that enhancement in water desalination plus energy generation can be stimulated by activated carbon.

Chojnacka et al. (2012) suggested that algal biomass could be a very good source of renewable and valuable active substances with diverse range of industrial applications to meet up with the growing demand on energy and water. The authors revealed that the commercial and productivity value of microalgal biomass and seaweeds is huge; thus, microalgae can be harnessed foe proficient supply of energy and water desalination. It should be noted that not all techniques are capable of removing natural ionic contaminants, though most techniques remove significant amount, but some remnant of pathogenic waste may still be found or transfer.

Wang et al. (2020) showed that algae biocathodes have received tremendous attention as an alternative substitute to abiotic cathodes known to cause release of toxic chemicals and very costly to use. The authors deployed the use of *Chlorella vulgaris* microalgae to treat landfill leachate under different conditions. From the results obtained, they suggested that algae biocathodes boosted the sustainable efficiency microbial desalination cells and equally generated bioenergy. Ummy et al. (2016) revealed that yeasts such as *Saccharomyces cerevisiae* are potential biocatalysts due to their physiochemical properties as non-pathogenic organisms in microbial fuel cells utilization. The authors suggested that further development is needed to make yeast a robust material for energy production and desalination agent.

Also, modifications of microbial fuel cells (MFCs) to microbial desalination cells (MDCs), research has helped in the extensive study and design of these MDC devices leading to a considerable upgrade over the years (Kim & Logan, 2013; Sevda et al., 2015). Brastad and He (Brastad & He, 2013) investigated the efficacy of the use of MDCs in purification of water. MDCs were reported to remove a significant of arsenic, copper, mercury, and nickel, and it was concluded that MDCs can be used to reduce water hardness. Xincao et al. (2009) studied the use of a microbial fuel cell having acetate as its substrates in desalination of water having various original salt concentrations which varies from 5 to 35 g/L. The bacteria cells were recorded to have eliminated salt present in water to about 90%, by utmost production of 2 W/m<sup>2</sup> of the microbial desalination cells. During the desalination process, there was a decrease in voltage as the cycle proceeded while after the desalination, the microbial cells had an improved resistance of 970  $\Omega$ from the initial 25  $\Omega$ . A new, energy producing and cost-effective desalination procedure was recorded by the authors. The use of stacked microbial desalination cells (SMDCs) in attaining potable water by evaluating the desalination rate was also investigated by Chen et al. (2011). At to 0.0252 g/h, it was reported that the frequency of desalination was increased when a two-desalination chamber of stacked microbial desalination cell was used. The SMDCs were recorded to have a greater external resistance which was 1.4 times greater than a single desalination chamber. Jacobson et al. (2011) performed a four-month study set to investigate an uninterruptedly active upflow MDC to eliminate salt and create electricity. They observed that in four days, there was a > 99% salt elimination achieved and the production of 62 mA of energy and a total dissolved solid (TDS) removal rate of 7.50 and 5.25 g for TDS salt solution volume and wastewater volume, respectively. It was also observed that a 30.8 W/m3 power density at its highest was also produced in this process. They concluded that the UMDC technique produced a significant increase in energy and further reduced the TDS.

Over a period of eight months, Luo et al. (2012) reported the several changes entailed in the properties of membrane and the different microbes found at the anode of an MDC. They recorded a decline by 27% of the desalination proficiency, and a reduction of MDC function was also recorded after a while evident by 47% deterioration in density of the current produced. It was also observed that scaling and fouling occurred at the cathode and anode of the membrane, respectively, with the phylum Proteobacteria being dominant at the AEM which improved system resistance thereby lowering ion transfer, and thus, the translation to energy efficiency is reduced. In order to reduce the problems of the migraton of ions which is caused by the MDC systems, Forrestal et al. (2012) investigated a novel microbial desalination cells; capacitive adsorption capability (C-MDC) having the ability to transfer ions to an adsorptive activated carbon cloth which are in turn used as electrodes, and they are further used for electrochemical ion adsorption purposes. It was discovered that in a single batch desalination cycle, 69.4% salt was eliminated without addition of salt to the anodes and cathode, and the total dissolved solids (TDS) (61-82.2 mg) were reduced by the adsorption of 1 g activated carbon cloth, which served as the electrode.

Mehanna et al. (2010) demonstrated the effectiveness and salt elimination capability of MDC. It was recorded that MDC reduces the conductivity of salt water prior to reverse osmosis treatment procedure in achieving potable water. They studied a three-sized chamber air cathode MDC and discovered that with the reduction of the conductivity of the salt water, there was a production of a 480mW<sup>-2</sup>power density when 1 gL acetate solution was used.

Morel et al. (2012) compared the desalination abilities of the classic MDC (C-MDC), and the MDC compacted with ion exchange resin (R-MDC). The R-MDC showed a 1.5–8 times better desalination potential rate with a low concentration of NaCl (10–2 g/L) than the classic MDC(C-MDC). It was recorded that due to the ion exchange resin which served as a conductor, there was an upsurge in ohmic resistance of R-MDC which was 55–272% lowered than that of the C-MDC; therefore, R-MDC stopped the increase in the resistance of the solution. It was concluded that R-MDC is best appropriate for water/waste water with decreased salinity.

Stoll et al. (2015) investigated that after one hour on the carbon electrode, there was a removal of 36 mg per gram of salt removal from the water produced. In an improved development by Wen et al. (2012), aerobic bacterial cells were used as a catalyst on the cathode of MDC and were compared with an air cathode MDC to ascertain its efficiency in waste water treatment and desalination. Under the same conditions, the voltage (136 mV greater than air cathode MDC) and the columbic efficiency at 96.2  $\pm$  3.8% were reported to be significantly increased in the bacterial catalyzed cathode MDC than when compared to the air cathode MDC. With anode solution of 0.441 L, the salinity of 39 ml of salty solution was decreased by 92% in the biocathode catalyzed MCD, and this method was concluded to be more efficient method in water desalination. In a further

enhancement of MDC technology, Yuan et al. (2015b) in a synergistic approach to eliminate chemical oxygen demand (COD) for water treatment and efficiency in energy, MDCs, and forward osmosis (FO) were employed in desalination of waste water. This was carried out by a pretreatment at the MDC anode, and furthermore, the effluent was sent to the forward osmosis for conclusive retrieval. The authors discovered that in the saline water, the conductivity reduction was improved to 99.4% by the MDC-FO technology, and it also reduced COD, and the waste water volume was also reduced by 93% and 65%, respectively, when compared to a single MDC. It was also recorded that the reduction in NaCl had an effect on the reduction of the COD. This is a hopeful method used for predesalination and waste water treatment of saline water. The synergistic utilization of forward osmosis and MDC (FO-MDC) in pretreatment of wastewater was studied by the use of mathematical predictive models. This was done by incorporating FO with MDC to the model. They reported that key parameters were detected, and this was further attained by amending definite waste water of domestic origin with 500 mg glucose and a buffer of 50 mM NaHCO<sub>3</sub>. They concluded that with appropriate proficiency, desalination could be effective and achieved (Yuan et al., 2015b).

In an in situ removal of nitrate from wastewater, it established a submerged microbial desalination cell serving in the removal of nitrate, water treatment, and energy production (Zhang, 2012). A total of 90.5% nitrate was removed and a current density of 3.4 A/m<sup>2</sup>. It was reported that the external resistance was 10  $\Omega$  at a 12 h retention time. It was concluded that SMDC is promising for the elimination of nitrate in ground water.

# 3 Conclusion and Future Recommendation to Knowledge

This chapter has provide a comprehensive information on the application of microorganism for the water desalination. Several types of microorganisms that could be applied for water desalination were also highlighted. The modes of action through which they exhibited their action were also highlighted. The principles involved in the process of desalination were also elucidated. During this study, it was discovered that commercialization of this technology has suffered tremendous setbacks due to several factors like pH instability, low current output, membrane biofouling plus polarization, scientist still believe that if all these challenges are resolved, microbial desalination holds a promising potential in the near future. Also, during this study, detailed information on principles involved in the process of microbial desalination such as air cathode microbial desalination cell, biocathode microbial desalination cell, stack-structure microbial desalination

cell, microbial electrodialysis cell, microbial electrolysis desalination and chemical-production cell, microbial capacitive desalination cell, upflow microbial desalination cell, osmotic microbial desalination cell, submerged microbial desalination-denitrification cell was also highlighted. This study also established that microorganisms are sustainable tools that could be applied for maintenance of a cleaner environment (Ukhurebor, et al., 2021a, 2021b; Osemwegie, et al., 2021; Adetunji, et al., 2018a, 2018b, 2017, 2014, 2019).

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# Graphene Composite Membrane for Water Desalination

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## Abstract

Water is the basic necessity in the everyday life of human beings. There is a problem of freshwater scarcity due to pollution caused by human interruption, urbanization, population growth, change of lifestyle, changing in climate with frequently occurring natural calamities such as floods and droughts and also feckless use of water. The research community is focused on water filtration as well as desalination in order to produce huge amount of purified water within very short period of time and low cost. Currently, researchers are giving interested in graphene and its derivative-based composite membrane for water filtration as well as desalination due to some specific properties of this membrane. These composite membranes are generally prepared by incorporating some polymer matrices into the graphene derivatives. Previously, graphene has become a formal candidate for water purification and desalination process due to its hardness, high thermal activity and good conductor of electricity. Like other materials, it has also some disadvantages such as proper distribution of pores on the graphene sheets. The ability of salt rejection by graphene decreases with the increase in pore size and pressure. Recently, graphene oxide (GO)-based composite membrane has become a novel material for water purification and desalination because of its large-scale production in industry, easy fabrication method and high mechanical strength. GO is the derivative of graphene containing oxygenated functional groups such as epoxy, carbonyl, hydroxyl and carboxyl group which increases the hydrophilic properties and acts as functional reactive sides. This lesson summarizes the recent progress of graphene-based composite

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membrane, and their fabrication methods for the application of water desalination draw special attention for effective challenges and future prospectus.

## Abbreviations

aGO	Amino-graphene oxide
aPES	Amino-polyether sulfone
APTS	3-Amino-propyl triethoxysilane
BSA	Bovine serum albumin
CNT	Carbon nanotubes
CVD	Chemical vapour deposition
DMAc	Dimethylacetamide
DNA	Deoxyribonucleic acid
f-GO	Functionalized graphene oxide
GO	Graphene oxide
GOF	Graphene oxide framework
Gt	Graphite
GtO	Graphite oxide
HPEI	Hyper-branched polyethylenimine
LbL-SA	Layer-by-layer self-assembly
MB	Methylene blue
MBR	Membrane bioreactor
MDS	Molecular dynamics simulations
MF	Microfiltration
MO	Methyl orange
NF	Nanofiltration
NG	Nanographene
NMP	N-methyl pyrolidone
OMWCNT	One-dimensional oxidized carbon nanotubes
PA	Polyamide
PAN	Polyacrylonitrile
PDA	Polydopamine
PAH	Polyallyaminehydrochloride
PEC	Polyelectrolytecomplex
PECVD	Plasma-enhanced chemical vapour deposition
PEG	Polyethylene glycol
PEI	Polyethylenimine
PES	Polyethersulfone

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PSF	Polysulfone	
PVDF	Polyvinylidene fluoride	
RB	Rhodamine B	
rGO	Reduced graphene oxide	
RO	Reverse osmosis	
TFC	Thin-film composite	
UF	Ultrafiltration	
UV	Ultraviolet	

### 1 Introduction

Water scarcity is a critical environmental problem across the globe. According to a report by WHO, a billion people lack safe drinking water at home and it is severe in poorer countries.

There is scarcity of consumable water in the globe due to rapid population growth. The underground and seawater contain a large amount of contaminated particles, salts and various unwanted particles. The cost of separating these particles is rather high, and therefore, this water cannot be used as consumable water in large scale (Lapworth, 2012; Wang et al., 2012a). In order to disinfect these infected water as well as desalination of seawater, researchers are applying various techniques which will be economically favourable and environmental friendly. They developed several methods such as reverse osmosis, ion exchange and distillation which have been used for water purification (Wang et al., 2012a). These methods have certain limitations: self-pollution, financially not feasible, more power consumption and adoption of acute resources (Wang et al., 2012a). Some economically developed countries, like Saudi Arabia and Israel, have implemented plants for reverse osmosis process (Harrington, 2014). In 1990s, this process was implemented in place of distillation method due to huge power requirement in distillation process (Spiegler & El-Sayed, 2001). A polymer membrane is used in reverse osmosis process with particular structural gaps. In this process, seawater is applied forcefully from one side of the membrane which permeates about 250 million gallons of freshwater per day and impermeable to hydrated ion like sodium and chloride ion (Aghigh et al., 2015). Since this process utilizes more current, this method was not appropriate for large-scale production of pure water (Dickie, 2007). Various membrane-based desalination processes which are categorized according to their membrane pore size and permeability are shown in Fig. 1 (Schilling, 2013). In order to eliminate the problems created by various membrane filter processes, researchers have developed a new technology called nanotechnology which has created a new platform for water filtration and desalination (Xu et al., 2012a). Some of the nanomaterial such as zeolite and carbon

nanotube (CNT)-based membrane used for water purification are inefficient due to its low water permeability (Hu et al., 2011) and low salt rejection (Fornasiero et al., 2008, 2010), respectively, as compared to graphene. As a consequence because of some unique characteristics, graphene and graphene-based composite membranes have become a challenging material for water purification and desalination (Cohen-Tanugi et al., 2013). This chapter describes about the recent development of various types of graphene-based composite membrane, its fabrication process and mechanism for water desalination and pointed out their future prospectus.

# 2 Water Desalination Membranes: A Challenging Effect

The better performance of the membrane for desalination generally depends on its fouling resistance, water flux and salt rejection. The high-operating costs of reverse osmosis process are because of the low water flux and high energy consumption. Further, it was noticed that by decreasing the pressure does not affect the membrane performance. Cohen-Tanugi et al. (2014) developed an ultra-permeable membrane in order to increase the water permeability performance of RO. They observed that the water permeability increases three times more by reducing the pressure vessel through 63 and 44% in favour of seawater and brackish water RO plants severally without affecting energy consumption and permeate recovery. The consumption of energy was also reduced by 15% and 63% for seawater and brackish water, respectively, at the same permeate production per vessel. Due to the overpriced value of energy, the decrease in energy consumption plays a crucial effect for this process. The key problem generates during desalination process is membrane fouling which causes the diminution of the RO membranes, activity (Greenlee et al., 2009; Matin et al., 2011). Generally, membrane fouling occurs due to precipitation of some insoluble salt on the membrane surface, blocking of the feed steam spacer, filling of pores present on the membrane surface by the minute particles, interaction of solute with the membrane surface. (Amiri & Samiei, 2007). A biofilm is also developed by the aggregation of the biopolymers or some microorganisms such as bacteria, fungi and algae on the coating area throughout reverse osmosis process which is a most challenging effect for seawater desalination technology (Misdan et al., 2012; Yang et al., 2011). The formation of biofilm causes the lower water productivity, shortening of membrane lifetime, requirement of high pressure for water output and utilization of high energy (Matin et al., 2011). In order to increase the membrane performance, various oxidative solutions like chlorine which is used for discharging the increased coating



Fig. 1 Various types of membrane filter processes according to their pore size

biological film. When this membrane is treated with chlorine solution, it reacts over polyamide (PA) on the coating area and stops the formation of biofilm (Alayemieka & Lee, 2012; Avlonitis et al., 1992; Cadotte, 1980; Gabelich et al., 2005; Misdan et al., 2012). Due to the repetition of this process, it chemically deteriorates the polyamide layer and decreases the TFC-RO performance for salt rejection (Maruf et al., 2012), even after addition of small amount of chlorine to the supply water (Robert, 2006).

# 3 Physicochemical Synthesis of Graphene and Its Properties

In 1962, graphene was first detected in electron microscopes supported on metal surfaces. Later on the two physicists Andre Geim and Konstantin Novoselov won the Nobel Prize in 2010 for their innovative experiment with respect to the 2D material (Boehm et al., 1962; Hancock, 2010; Novoselov et al., 2004). The researchers gave significant focus on graphene for its various applications including water desalination. In 2013, Lockheed Martin patented under the trademark "perforene" for this application of graphene (Boretti et al., 2018). Graphene is a single two-dimensional sheet of graphite. In graphene, each carbon atoms are  $sp^2$  hybridized and are bonded together forms a hexagonal honeycomb-like lattice structure which gives it many amazing characteristics. The carbon-carbon bond length of graphene is 0.142 nm (Solutions, 1958). The carbon atoms of graphene are joined by each other through  $\sigma$ —bonds, and they have also  $\pi$  orbital through which the free electron can delocalize; as a result, graphite is the good conductor of electricity. The three-dimensional arrangement of graphene layer one above the other having interplanar gaps 0.335 nm is called graphite (Surwade et al., 2015). Kim et al. (Bae et al., 2010) developed roll-to-roll production of monolayer 30-inch graphene films having 97.4% optical transmittance. It is strongest, lightest, most conductive and transparent and has good mechanical stability which allows high water flux, more water transport, high salt rejection, need low pressure and broad serviceable conditions than previously possible. The fabrication of graphene is based on bottom-up and top-down methods. In bottom-up method, graphenes are prepared by the chemical reaction of small organic molecules or atoms. Silicon carbide, thermal CVD and PECVD are generally used for the epitaxial growth of graphene on various metal surfaces (Wang et al., 2013). The methods such as chemically derived graphene, chemical reduction and electrochemical synthesis are used in top-down method for better yield and good quality of graphene (To Rid Water of Salt, MIT Group Taps Thin Carbon & Computing, Office of Science, U.S., 2014). Large-scale graphenes are also prepared by the reduction of graphene oxide which is formed from the exfoliated graphite oxide (To Rid Water of Salt, MIT Group Taps Thin Carbon & Computing, Office of Science, U.S., 2014). Nanoporous graphene used for water desalination process is generally prepared by creating nanopores within the unsaturated carbon atoms passivated by the chemical functional groups. The initial methods, i.e. electron beam exposure, were mainly used for creating





nanopores. Recently, some other experimental procedures were developed by the researchers, for example diblock copolymer modelling, helium ion drilling, and chemical inscribing to carry out the distribution of pore size precisely as well as higher porosity (Bell et al., 2009; Bieri et al., 2009; Garaj et al., 2010; Kim et al., 2010). The fabricated nanoporous graphene has capability for DNA sequencing and gas separation (Blankenburg et al., 2010; Hauser & Schwerdtfeger, 2012; Postma, 2010; Schrier & McClain, 2012; Xu et al., 2012b), as well as it has a potential role for water desalination. Using classical molecular dynamics simulation Grossman and his co-workers (Bae et al., 2010) reported that the rate of rejection of salt from water by using single-layer nanoporous graphene, increases twofold to threefold higher in magnitude, i.e. range 10-100 L/cm<sup>2</sup>/ day/MPa, when it was compared with diffusive RO membranes. With all of the above-mentioned advantages of NG sheet in RO application, there are still some unknowns with all of the above-mentioned advantages of NG sheet in RO application. The results obtained for NG membrane are stimulation-based yet there has not been obtained any practical result for this novel membrane. Another problem is that the flow of water through NG membrane increases with increasing the number of pores on the graphene sheet as a result the mechanical stability of the membrane decreases. So the primary concerns for the NG sheet membrane are properly managed and distribution of pores on the graphene sheets in addition to scaling up the manufacture of the graphene sheets. In order to sort out the problems created for this method, comprehensive examination and further investigation have to be carried out. Nanocarbon-based materials, i.e. graphene and CNTs, act as antimicrobial surfaces which inhibits the bacterial growth when come in direct contact with cells (Akhavan & ElhamGhaderi, 2010; Kang et al., 2007; Liu et al., 2011; Tu et al., 2013; Vecitis et al., 2010). Liu et al. (2011) studied the antibacterial activity of four types of graphene derivatives, i.e. graphite oxide (GtO),

reduced graphene oxide (rGO), graphite (Gt) and graphene oxide (GO) against the bacterial model 'Escherichia Coli'. They observed that graphene oxide has the largest antibacterial activity succeeded by GtO, rGO and Gt consecutively under equal focusing and incubation conditions. Furthermore, graphene is an inactive matter and it melts very slightly in organic solvents (Stankovich et al., 2007). Hence, it was suggested that for practical utilization of graphene in several applications, the fresh graphene should be chemically modified (Stankovich et al., 2007; Sun et al., 2011). Graphene oxide is an oxidized form of graphene that contains oxygen-containing functional groups such as carboxyl, carbonyl, hydroxyl and epoxy groups on its edges and basal planes (Lerf et al., 1998) (Fig. 2). The presence of such types of functional groups empowers the hydrophilic properties of GO and increases its solubility in water, which also makes an expedient and profitable process for the preparation as well as molecular separation of GO membrane (Huang et al., 2013; Szabó et al., 2005). These oxygenated functional groups also take part for further surface modification of GO nanosheet for which corresponding functional GO-based composite membrane can be prepared with more suitable separation performance. Due to these advantages as well as high surface area of GO nanosheets, researchers developed different types of GO-based composite membrane and considered as a potential graphene-based material for water purification (Hu & Mi, 2013), desalination (Feng et al., 2016a), gas separation (Kim et al., 2013a; Shen et al., 2016a) and evaporation(Hung et al., 2014).

# 4 Preparation of GO

The synthesis of GO involves through two consecutive steps, i.e. oxidation of graphite followed by exfoliation of graphite oxide, as shown in Fig. 3. From the various literature review, we found that there are various methods of



Fig. 3 Schematic illustration of GO preparation processFig. 3 Schematic illustration of GO preparation process

preparation of GO have been reported. In 1859, Brodie (Brodie, 1859) first synthesized GO by using graphite as starting material which was oxidized by using fuming HNO<sub>3</sub> and potassium chlorate (KClO<sub>3</sub>) as the oxidant. Later on, this method was revised by Staudenmaier which was more efficient and suitable method for the production of GO. In this method, he added KClO<sub>3</sub> fractionally in graphite solution and then acidified this mixture with concentrated H<sub>2</sub>SO<sub>4</sub> (Staudenmaier, 1898). Both these methods produce toxic gases which were insecure and environmentally unfriendly. In 1937, Hofmann improved the production process of GO by substituting non-fuming HNO<sub>3</sub> in place of fuming HNO<sub>3</sub> during the oxidation process (Hofmann & König, 1937). After Hofmann, in 1958 Hummers and Offeman focused on further development of this procedure, who served graphite with anhydrous mixture of KMnO<sub>4</sub>, NaNO<sub>3</sub> and concentrated  $H_2SO_4$  (Hummers Jr et al., 1958). This process was safe, productive and complete within 2 h. Later on, it was modified by Kovtyukhova et al. (1999) for complete oxidation of graphite. First, they treated graphite powder into a mixture solution consisting of concentrated H<sub>2</sub>SO<sub>4</sub>, P<sub>2</sub>O<sub>5</sub> and K<sub>2</sub>S<sub>2</sub>O<sub>8</sub> at 80 °C. Then this solution was filtered, washed and dried completely. After that, this pre-oxidized graphite further oxidized by using Hummers' method which than exfoliated to produce GO flakes. Although this process produces toxic gases, still this method was more efficient and gives better yield within very short period. Recently, some other techniques have also been developed for producing GO (Luo et al., 2009; Marcano et al., 2010; Peng et al., 2015).

# 5 Preparation of GO Membrane

There are various methods of preparation of GO membranes which are based on high aspect ratio and also degree of dispersion of GO. These methods include layer-by-layer (LBL) self-assembly method, filtration-assisted method and casting/coating–assembly method. Filtration-assisted method is the simple way for the preparation of GO in which GO is homogeneously suspended in a solution, after that it is filtered by using a porous support to obtain a uniform coating. The filtration process can be vacuum- or pressure-assisted. By using this method, mainly free-standing GO membranes are prepared (Dikin et al., 2007; Hung et al., 2014; Joshi et al., 2014). This method provides a prospective way to enlarge the production of GO membrane and also controls over the microstructure as well as wideness of the membrane. Some other simple methods belonging to casting/coating--assisted method have also been developed for the production of GO membranes. These methods consist drop-casting (Shen et al., 2016b), spin-coating (Robinson et al., 2008), spaying-coating/casting (Nair, 2012) and dip-coating (Wang et al., 2016). By using spin-coating method, Kim et al. (2013a) prepared few-layered GO sheets on a polyether sulfone (PES) substrate for the purpose of gas separation. Large area and ultrathin GO sheets were also prepared by Robinson and his co-workers (Robinson et al., 2008) via modified spin-coating technique. They reported that these membranes have outstanding mechanical properties. By varying the concentration of GO in solution its membrane thickness could be controlled on nanometer scale. In this method, individual GO nanosheet within the GO membranes holds strongly by hydrogen bonding and Van der Waals force. Recently, researchers gave their interest on fabrication of GO laminate via layer-by-layer deposition of GO nanosheets. These GO nanosheets within the GO laminate are held together by either covalent bonding or electrostatic interaction (An et al., 2011; Hu & Mi, 2013). Hu et al. (Hu & Mi, 2014) prepared bilayer GO-PAH composite membrane by layer-by-layer deposition of GO and poly(allylamine hydrochloride) on the porous PAN substrate which act as a membrane supporter. Since the surface of GO nanosheet is positively charged and PAH is positively charged, so both are held together through electrostatic force of attraction. They reported that the fabricated GO membrane show high water permeability, high salt rejection ( $\sim 99\%$  for sucrose) and possessed a close-packed structure in solution having low ionic strength. Since this process is renewable, cost-effective and environmentally friendly, so it is conventionally used for the preparation of GO membranes. Apart from these methods, some additional methods have also been implemented for the preparation of GO membrane such as evaporation-assisted method (Chen et al., 2009; Guan et al., 2017), templating-assisted method (Wei et al., 2008), shear-induced alignment method (Akbari et al., 2016), Langmuir-Blodgett assembly method (Cote et al., 2009; Li et al., 2008) and hybrid method (Shen et al., 2016c). From the description of the aforementioned methods of preparation of GO-based membrane, it concluded that the shape of the GO membrane and its membrane performance towards separation generally depends on the conditions and methods used during the fabrication process. Therefore by using reformed fabrication condition and suitable fabrication technique, a preferable GO membrane can be achieved for a specific practical application.

#### 6 Characterization of GO membrane

It is very important to characterize the physical and chemical properties of graphene oxide membrane for further application. Various characterization techniques have been utilized for the structural determination of GO membrane and its performance for separation. These characterization techniques are mainly thermogravimetric analysis (TGA), powder X-ray diffraction, X-ray photoelectron spectroscopy (XPS), scanning electron microscope (SEM), transmission electron microscopy (TEM), atomic force microscopy (AFM), Raman spectroscopy, Fourier transform infrared spectroscopy (FT-IR), contact angle measurement and measurement of zeta potential. Surface morphology, especially uniformity and roughness on the surface of the membrane, is studied by TEM, SEM and AFM. Thickness and cross-sectional morphology of the membrane are also studied by applying these methods (Hu & Mi, 2013; Li et al., 2008). Studies of the contact angle and TGA are very useful to get the information regarding the hydrophobic or hydrophilic nature of the surface and thermal stability of the membrane, respectively (An et al., 2011; Feng et al., 2016a). Chemical composition and microstructure of the membrane are studied by FT-IR, Raman, XPS and XRD. These techniques are the most commonly used techniques for this purpose (Guan et al., 2017; Huang et al., 2014). The charge on the surface of the membrane is determined by using surface zeta potential measurement. Mechanical stability of graphene oxide membrane is determined by stress-strain curve. Furthermore, tensile strength and Young's modulus are also characterized by this curve (Huang et al., 2013). An experimental method for the precise determination of d-spacing of graphene-oxide membrane in the aqueous environment is reported by Mi et al (Zheng et al., 2017). Various characterization methods used to characterize GO membrane are outlined in Table 1.

# 7 Recent Development of GO-Assisted Desalination Membranes

GO membranes are classified into three categories for the purpose of water desalination such as free-standing GO membranes, supported-GO membranes and incorporated-GO membrane. If the GO membrane is directly used as a separating layer without attaching any other material, it is categorized as free-standing GO membrane. In the second category, either an inorganic or a polymeric substance assists GO membranes for operating the membrane performance. The third employs that the GO nanosheets are directly fused with polymer casting solution in the time of production. Recent development and **Table 1** Various methods used to characterizeGO membrane

Methods used for Characterization	Information obtained by characterization of GO membrane	References
SEM AFM TEM TGA CA FT-IR Raman Spectroscopy XPS XRD Surface Zeta potential	Identification of detail structure of its surface as well as its cross-sectional area Information obtained about the roughness of membrane surface and its uniformity Explained the detail structure of its surface and its cross-sectional area Thermo resistance of the membrane Hydrophobic and hydrophilic characteristics of the membrane surface Functional groups present on the	Hu and Mi (2013) Hu and Mi (2013) Cote et al. (2009) Sun et al. (2011) Feng et al. (2016a) Chen et al. (2009) Chen et al. (2009) Zheng et al. (2017) Huang et al. (2013) Huang et al. (2013)
Stress–strain curve Integrated quartz crystal microbalance with dissipation and ellipsometry	membrane surface and its chemical structure Pointed out GO's existing capacity in composite membrane Quantitative analysis about the composition of element present in the membrane Determine the structure of the crystal along with its d-spacing Recognized about the charges on the membrane surface Young's modulus, mechanical strength and hardness Precisely calculate the d-spacing of membranes in a water medium	DDikin et al. (2007)

performance of the above three categorized GO-based desalination membranes are summarized in Table 2.

# 7.1 Free-Standing GO Membrane

Free-standing GO membrane is mostly used in water desalination application because of some specificity in structure. Generally, GO membranes are formed in the form of laminate in which the GO nanosheets are packed and stacked one above the other and there forms an interlayer gap called d-spacing (Fig. 4a). GO membranes have high tensile strength and pliability due to their tiny thickness (Dikin et al., 2007; Eda & Chhowalla, 2010; Geim, 2009; Robinson et al., 2008). From the various researches, it was also concluded that due to the existence of the oxygen-containing functional groups these membranes selectively permeate water molecule without any obstacle, but do not allow liquids, vapours, gases and also helium. The formation of interlayer gaps between the GO sheets is also possible due to the presence of these functional groups in GO (Cerveny et al., 2010; Jeong et al., 2009; Lerf, 2006; McAllister et al., 2007). Xu et al. (2013) prepared graphene oxide with TiO<sub>2</sub> composite NF coat which contains pore size of 3.5 nm by using vacuum filtration process. They reported that this membrane successfully rejects ( $\sim 100\%$ ) methyl orange (MO) and Rhodamine B (RB) from water. A Cu-assisted GO NF membrane was prepared by Nair et al. (2012) via spin- or spray-coating method with their interlayer slits reduced to 1 nm. They reported that the acquired membrane penetrates water molecule approximately 10<sup>10</sup> times faster relative to helium and in penetrable to liquids, vapours and gases, along with helium. Sun et al. (2013a) prepared free-standing GO NF membrane minimized to intersect space 0.85 nm and applied for the partition of copper salt and sodium salt from organic contaminants. From classical molecular dynamics simulation (MDS), Nicolai's group (Nicolaï et al., 2014) reported that free-standing GO membrane acts as an ideal candidate for water desalination with 100% salt rejection and its permeation ability increases twice as compared to the current RO techniques.

#### 7.2 Supported-GO Membrane

Although free-standing GO membrane has performed well in water purification and desalination application, a supported-GO membrane has also a great role in membrane separation application as well as it improves the antibacterial activity (Liu et al., 2011; Perreault et al., 2014; Tu et al., 2013), antifouling properties and more reluctant to chlorine

Types of GO Membrane	Name of the GO Membrane	Membrane Classification	Fabrication Method	Membrane performance	References
Free-standing	GO/TiO <sub>2</sub> Cu-assisted GO GO membrane	NF NF NF	Vacuum filtration Spray/spin coating Drop-casting	Remove MO and RB 100%, flux: 7 L/m <sup>2</sup> .h water permeation 10 <sup>10</sup> times more selective permeation sodium salt than copper salt and organic content	McAllister et al. (2007) Xu et al. (2013) Sun et al. (2013a)
Supported	aGO/GO/aPES aGO/GO/PA GO-TiO <sub>2</sub> /PSF GO/PAN GO/PA	RO RO (TFC) Photocatalytic NF pervaporation RO (TFC)	Self-assembly LBL deposition Self-assembly LBL deposition UV Self-assembly LBL deposition Pressure-assisted self-assembly drop-casting GO covalent bonded membrane surface	NaCl rejection: 98% Water flux: 28 L/m <sup>2</sup> .h NaCl rejection: 96.4% Water flux: 14 L/m <sup>2</sup> .h Reject MB: 90% Flux: 45 L/m <sup>2</sup> .h IPA/water : 70 wt%, 99.5%, Flux: 4137 g/m <sup>2</sup> .h Reject NaCl: 97.8% Flux: 1.5 L/m <sup>2</sup> .h.bar	Kim et al. (2013b) Choi et al. (2013) Gao et al. (2014) Xu et al. (2013) Perreault et al. (2014)
GO-modified GO-modified	GO/NMP/PSF GO/DMAc/PVDF GO/NMP/PSF i-f-GO/PSE HPEI-GO/PGE GO/PECs GO/OMWCNTs/PVDF GO/DMAc/PES APTSf-GO/PVDF GO/DMAc /PVDF	UF UF NF UF NF UF NF UF MF	Phase inversion Phase inversion Phase inversion Phase inversion Dynamic self-assembly LbL blending Phase inversion Phase inversion Phase inversion Phase inversion	Rejection BSA: 99% Flux: 450 $L/m^2$ .h Rejection BSA: 79% Flux: 26.49 $L/m^2$ .h Rejection Na <sub>2</sub> SO <sub>4</sub> : 72% Flux: 50 $L/m^2$ .h Rejection BSA: 95% Flux: 135 kg/m <sup>2</sup> .h RejectPEG 20000: 85% & PVA 30000-70000: 90%, Flux: 153.5 $L/m^2$ .h Reject Mg <sup>2+</sup> 92.6% and Na <sup>+</sup> 43.2%, flux: 7.1 kg/m <sup>2</sup> .h MPa, & 8.1 kg/m <sup>2</sup> .h MPa Flux: 410 $L/m^2$ .h Antifouling BSA: 98.3% Dye rejection: 96% Flux: 53 kg /m <sup>2</sup> .h Rejection BSA 57% Flux: 401.39 $L/m^2$ .h Yeast suspension 80% Flux: 324.5 1 /m <sup>2</sup> .h	Xu et al. (2014) Zhao et al. (2014) Zinadini et al. (2014) Ganesh et al. (2013) Lee et al. (2013) Sun et al. (2013) Yu et al. (2013) Zhao et al. (2013) Zhao et al. (2013) Yeh et al. (2015)

 Table 2
 Outline regarding the current analysis of various GO-based water desalination membrane and its impact on the membrane performance

(Choi et al., 2013; Kim et al., 2013b). Since small quantity of GO nanomaterials is needed to customize the surface of GO membrane, so it is inexpensive and eco-friendly. The surface modification of GO membranes occurs by applying layer-by-layer self-deposition technique either covalently or non-covalently, as shown in Fig. 4b. For non-covalently modify the membrane, GO could be used directly or through bridging material (i.e. aGO or TiO<sub>2</sub>) depending on the net charge on the membrane surface. Elimelech et al. (Perreault et al., 2014) prepared GO/PA (TFC) membrane which has sturdy and good impact for absorbing water as well as destroying microorganisms that carry diseases. They also observed that the bacterial cell undergoes deactivation up to 65% after one hour on direct contact with the modified membrane without disturbing the flow of water as well as disallowance of salt. It was also studied that, although there is an increase in hydrophilic character of the modified membrane, there is not any improve of water flow rate. This is because the rate of flow of water is independent of the surface improvement, but depends on the solution diffusion model of PA (TFC) polymer (Elimelech & Phillip, 2011). Kim et al. (2013b) fabricated a non-covalently bonded aGO/GO/aPES RO membrane by successive deposition of negative part GO NPs layer and positive part amino-GO (aGO) on an amino-polyether sulfone (aPES) membrane surface. They reported that this fabricated membrane rejects salt 98%, and permeates water 28 L/m<sup>2</sup> h, as well as it has more resistance to chlorine relative to primary PA RO

membrane. When GO and a GO multilavers were coated on the polyamide (PA) thin-film composite (TFC) membrane surface via layer-by-layer self-assembly process, it acts as chlorine resistant as well as antifouling property (Choi et al., 2013). In addition, when this membrane was matched with unmodified membrane, it was found that the rate of flow of water was enhanced by 10% and there is a slight decrease of salt rejection by 0.7%. By applying the same technology, a photocatalytic membrane GO-TiO<sub>2</sub>/PSF was prepared by Gao et al. (Gao et al., 2014). They fabricated the membrane by depositing GO layer on the exterior part of a polysulfide membrane followed by TiO2 in the presence of UV and sunlight radiation. It was also observed that GO undergoes partial reduction after treating with ethanol/UV. They reported that the modified membrane exhibits decomposition of organic contaminates in the presence of UV and sunlight and effectively rejects 90% methylene blue (MB), along with this modified membrane increases the flow of water to 45  $L/m^2/h$  under UV, due to more membrane hydrophilicity caused by photoactivity of GO-TiO<sub>2</sub>. Preparation of GO-modified PAN composite membrane through pressure-assisted self-assembly method was guided by Hung et al. (2014) and studied their pervaporation separation for isopropyl alcohol (IPA)/H<sub>2</sub>O solution. They observed that during the pervaporation separation of 70 wt% IPA/water solution, the fabricated membrane shows an excellent work: nearly 99.5 wt% water recovery and the water permeation flux reaches 4137 g/m<sup>2</sup>/h. The high selectivity of this composite membrane towards permeation of water molecule and avoiding of IPA molecule is because of the highly ordered and packed arrangements of the GO laminates. By applying vacuum filtration method, Huang and co-workers (Feng et al., 2016b; Xu et al., 2016) developed GO/Al<sub>2</sub>O<sub>3</sub> and (GOF)/Al<sub>2</sub>O<sub>3</sub> composite membrane and covalently joined by polydopamine (PDA) and 1, 4-phenylene diisocyanate (PDI), respectively. These modified membranes are very robust, reproducible, and as well as they selectively permeate some molecules and ions. Due to the great adhesive ability of PDA and PDI, the GO nanosheets were strongly bound onto the support surface. Both the composite membranes show appreciative performance for seawater desalination with high ion rejection over 99.7% and water outflow for 3.5 wt% seawater.

## 7.3 GO-Incorporated Composite Membrane

Apart from the two aforementioned methods, researchers concentrated on altering polymeric membrane with GO nanosheets by applying several techniques in order to enrich the outlet of water through this membrane and also to increase its antibacterial activity as well as its tensile strength (Lee et al., 2013; Wang et al., 2012b; Yu et al., 2013; Zhang et al., 2013). The GOs used to incorporate into the polymer matrix are either functionalized or used directly, as shown in Fig. 4c. Several researchers used various modifiers to functionalize GO such as hyper-branched polyethyleneimine (HPEI) (Yu et al., 2013), 3-amino-propyl triethoxysilane (APTS) (Xu et al., 2014) and isocyanate (i) (Zhao et al., 2013a). Xu et al. (2014) prepared APTS f-GO/PVDF by adjoining different ratios of GO and f-GO to the hybrid polyvinylidene fluoride (PVDF) via phase inversion induced by immersion precipitation technique. When this membrane was correlated with the GO/PVDF, it was observed that the obtained membrane exhibits high water flow rate, more tensile strength and also elongation-at-break increases by 69.01 and 48.38% due to the sturdy covalent bonding interaction between the faces of functionalized GO and PVDV. They also observed that this fabricated membrane has the capability for the rejection of bovine serum albumin (BSA) about 55%. The identical phenomenon was launched by Yu et al. (2013) and revealed the fabricated membrane HPEI f-GO/PES exhibited high tensile strength and Young's modulus. The hybrid membrane also showed an effective antibacterial performance and slightly decreases pure water flux as compared with pure membranes. The potentiality of this prepared membrane to reject polyvinyl alcohol (PVA) 30,000-70,000 and PEG 20,000 by about 90% and 85%, respectively, was also enumerated by them. By applying the same methodology, Zhang et al. (2013a) prepared an i-GO/PSF ultrafiltration membrane and studied its membrane performance as well as antifouling properties. They reported that the fabricated membrane has more negative zeta potential and also increases its hydrophilic character along with smoothness of the membrane, which result for enhancing its antifouling properties. They also observed that this membrane has the ability to reject bovine serum albumin (BSA) and ovalbumin by about 95% and 6%, respectively. Some researchers also focused on to incorporate unfunctionalized GO into polymer matrix. By applying phase inversion technology, they incorporated the GO into different polymeric coatings. These are graphene oxide / N-methyl pyrolidone (NMP)/PSF (Ganesh et al., 2013), and graphene oxide /di-methyl acetamide (DMAc)/PES (Zinadini et al., 2014) NF membrane, graphene oxide/onedimensional oxidized carbon nanotubes (OMWCNTs)/ PVDF (Zhang et al., 2013), and graphene oxide/DMAc/ PVDF UF membrane (Zhao et al., 2013b), GO/DMAc/ PVDF (Zhao et al., 2014) MF membrane and GO/NMP/PSF (Lee et al., 2013) membrane bioreactors (MBRS) and studied their extraordinary impact. Ganesh et al. (2013) prepared the graphene oxide /N-methyl pyrolidone (NMP)/PSF and studied its water flux and rejection flux. They investigated that the permeability of water increases due to increase in hydrophilic character of the membrane as well as the formation of more number of microvoid on the membrane



Fig. 4 Overview of a Free-standing GO membrane, b Supported-GO membrane, c Incorporated-GO membrane

surface by doping GO during phase inversion method. They also observed that this fabricated membrane rejects NaSO4 by 72% with loading of GO about 2000 ppm, at 4 bar pressure and the trend of salt rejection increases with rise in the PH of feed solution. A novel GO/di-methylacetamide (DMAc)/PES membrane was developed by Zinadini et al. (2014), and they stated that the membrane has high dye removal capacity by 99% and significant improve of water flow rate by 65.2 kg/m<sup>2</sup> h due to large hydrophilic character of the prepared membrane. Li et al. (Zhang et al., 2013) investigated the synergistic effect between the two-dimensional GO and one-dimensional oxidized carbon nanotubes (OMWCNTs) on permeation and antifouling performance of polyvinylidene fluoride (PVDF). They studied that long and tortuous OMWCNTs bridge the adjacent GO and inhibit their aggregation which improves its antifouling performance and hydrophilicity of the fabricated membrane. The modified membrane has bigger pore density which plays a positive role in permeation flux and the pure water flux recovery achieved 98.28% for membrane with the ratio of 5:5 (GO/OMWCNTs). Zhao et al. (2013b) prepared the modified GO/DMAc/PVDF UF and reported that this fabricated membrane increases flow of water about 79%, equivalent to 26.49 L/m<sup>2</sup> h, as well as it rejects BSA about 44.3%. The better antifouling properties of this UF membrane are because of the change of surface hydrophilicity and smoother surface with a higher efficient filtration area.

# 8 Advanced Mechanical Sturdiness of GO membrane and Its Stability in Aqueous Environment

In recent times, GO membrane has drawn considerable attention due to their remarkable performance in separation. The separation ability of GO membrane is much higher than the existing traditional membranes. Despite the promising application of GO membrane for its performance in separation, still some critical challenges exist for practical applications. The most significant challenge is to determine the stability of intermediate spacings between adjoining GO nanosheets during application in aqueous medium. This problem arises because of the hydrophilic properties of GO sheets and the electrostatic repulsion of negatively charges GO sheets on hydration. By this way, the GO membrane gradually damaged after working over a particular time period. Basically, the interlayer nanochannels are deteriorated and disassembled (Sun et al., 2013b; Yeh et al., 2015). It is essential to increase the structural toughness of GO membrane by creating strong bonding between the GO nanosheets for its real-world approach for water purification. Stability of GO membrane in aqueous environment has gained considerable attention, and many productive contributions are made to solve this issue. Recently, scientific communities have developed stable GO membrane which is applicable in aqueous environment by insertion of various cross-linking agents in between the adjoining GO nanosheets. These are bonded either by covalent bonds or electrostatic force of attraction. The first chemically cross-linking agents that inserted between GO nanosheets utilizing di-valent ions and polyallylamine (PAA) were fabricated by Park et al. (Park et al., 2008, 2009) which significantly improved the mechanical stability of GO membrane as compared to the pristine GO membrane. Some other cross-linked GO membranes were developed by Mi et al. (Hu & Mi, 2013, 2014) that showed outstanding stability towards the treatment of water. This cross-linked membrane was prepared by layer-by-layer deposition of 1, 3, 5-benzene tricarbonyl trichloride (TMC) and positively charged PAH as cross-linkers. Zhao et al. (2015) prepared a gelatin (GE) cross-linked GO membrane for the separation of water/ethanol which interlink the GO nanosheets through hydrogen bonding or by electrostatic and hydrophobic interaction. In recent past, Lim et al. (2017) fabricated a stable PEI cross-linked TA-GO membrane by covalent bonding between tannic acid (TA)-functionalized GO membrane and polyethylenimine (PEI) which act as cross-linker that showed outstanding performance in an aqueous environment. By applying vacuum-assisted filtration method, Zhang et al. (2017) prepared an innovative graphene oxide framework (GOF) membrane by covalent cross-linking of GO nanosheets with chemically cross-linking agent isophorone diisocyanate (IPDI). The obtained IPDI-GOF membranes improve the membrane stability as well as it increases the permeability of water (80  $L/m^{2}/h$ ) under appreciably low pressure (1.0 bar) and readily removes organic dye molecule up to 96%. From various experimental studies, researchers also observed that the reduced GO sheets might also improve the sturdiness of GO membrane by increasing the  $\pi$ - $\pi$  interaction among the GO nanosheets. Yang et al. (2017) designed a PDA-rGO membrane by coating polydopamine (PDA) layer onto the reduced GO laminates, which not only strengthen the membrane stability but also excellently permeate the water (36.6 L/m<sup>2</sup>/h) along with maximum rejection of salt about 92% in forward osmosis (FO) desalination. The outstanding performance of water flux with high salt rejection rate is

possible due to the compressed nanochannels as well as increased hydrophilic properties on the surface of reduced GO laminates.

# 9 Conclusion

Graphene and its derivatives have higher properties and show excellent potential to be operated in membrane separation. Graphene-based composite membrane has excellent performance in water purification and desalination. Various types of graphene oxide membranes such as free-standing GO membranes, supported-GO membranes and incorporated-GO membranes have been utilized in several procedures for better membrane performance towards desalination application. After the incorporation of GO, there is an increase in several membrane properties such as tensile strength, antibacterial activity, thermal properties, selectivity, water flow rate and antifouling properties, as compared to the pristine membrane. GO-assisted composite membrane shows a positive performance for split of mono-valent and di-valent ions and dehydration of solventwater mixture and dyes discharge. It was observed that the coupling of graphene oxide and polymers is advantageous for the development of membrane properties which helps the development of effective GO-based desalination membrane through various techniques. Though graphene oxide nanosheet is an auspicious development barrier for the fabrication of graphene oxide-assisted desalination membrane, there is also have some disadvantages. These are surface defects, unideal alignment, mechanical instability and assembly. The most effective challenge that was faced by the researchers is large-scale wholesale production of an ultrathin high permeability GO membrane. More research should be carried out to further understand the structural stability, membrane interaction and fabrication process for better membrane separation performance. Although there are challenges and problems, graphene-based composite membrane provides one of the most promising tools for water purification and desalination and solves the expected global water crisis.

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# **Renewable Energy-Assisted Desalination**

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# Abstract

The present water and energy crisis facing the world at large with the ever-growing population is one that demands careful attention by the research community. The treatment of seawater and brackish water by integrating renewable energy technologies into desalination processes holds a promising future for availing freshwater in areas of water scarcity across the globe. This chapter captures the different desalination technologies (such as thermal and membrane technologies) and different renewable energy technologies (like solar, wind and geothermal energies) that can be integrated into the process of water treatment for salt removal. Utilizing renewable energy technologies in desalination systems will serve as alternative where grid electricity is not available, reduce environmental pollution and cost.

# Keywords

Desalination • Solar energy • Wind energy • Geothermal energy • Hybrid system • Future prospects

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#### 1 Introduction

Water is very important in sustenance of living creatures; nevertheless, most of the water sources around are unfit for consumption due to high salt content. Freshwater is approximately 3% of the total water on earth while the remaining percentage accounts for the saline water. Figure 1 shows the total freshwater available from total water available on earth (World Bank Group, n.d.). This share of freshwater accounts for the lake, groundwater, glacier and ice cap. Also, due to the climatic changes resulting from greenhouse gases, the freshwater source will only get bad coupled with worldwide freshwater shortage. The predictions by World Health Organization (WHO) show that by 2025 half of the world's population will be faced with limited freshwater supply because of the climatic changes. The reasons behind this water shortage are high drought, increase in population, more contamination of available clean water sources, and high penetration of saltwater into fresh groundwater sources.

Desalination is a process in which saline or brackish water is separated into two parts: freshwater and brine concentrate (Buros, 2000). According to Krishna (Krishna, 2004), it is the process of removing dissolved salt from seawater or brackish water to produce freshwater, thereby enhancing the water quality and making it fit for human use. Its major aim is to make freshwater available for domestic (such as drinking and food preparation), agricultural use (such as irrigation) and industrial uses (such as mineral refineries). Desalination proffers solution where there is limited supply of freshwater to satisfy the need for portable water. However, it is energy-intensive and costly when compared to other freshwater sources (wastewater, surface water and groundwater treatment) even though the seawater and brackish water sources are largely available and inexhaustible. Most of the desalination plants around the world are usually powered by non-renewable energy sources to purify saltwater for various applications (Emrah, 2015).

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**Fig. 1** Percentage of total freshwater available from total water available on earth



However, currently installed and operational desalination plants worldwide emit around 76 million tonnes of carbon dioxide annually, and it has been predicted to rise to around 218 million tons by 2040. This estimation shows a negative impact of fossil-powered desalination plant presently and in future terms which is not good enough and needs to be tackled. Also, increased costs, increased energy demands, environmental challenge and increased maintenance conditions have made desalination non-attractive option. Nevertheless, various countries faced with water crisis have shown that it is a good alternative of solving the problem and can be worked on to reduce or eradicate if possible its drawback. Hence, this work discusses the treatment of water using renewable resources to generate power for desalination process and this strategy will eliminate the bottleneck resulting from conventional desalination systems that are powered by fossil fuel.

# 2 Desalination Technologies

Presently, there are two main classes of desalination technologies: thermal and membrane technologies. Both classifications require energy to effectively carry out water purification which could either be conventional energy source or renewable energy supply. Each of these technologies has its subdivisions which operate based on different techniques. These main desalination techniques are shown in Table 1. There are other emerging technologies which are not popularly used such as freezing, dew vaporization and ion exchange techniques. Globally, thermal and membrane technologies have a capacity of approximately 7 billion gallons per day early two decades ago, with equal percentage in thermal and membrane technologies. This capacity has risen to approximately 12% in a year between

Table 1	Desalination	technologies	and	processes
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Thermal technology	Membrane technology
Multi-Stage Flash Distillation (MSF)	Electrodialysis (ED)
Multi-Effect Distillation (MED)	Electrodialysis Reversal (EDR)
Vapour Compression Distillation (VCD)	Reverse Osmosis (RO)

1972–1 999(U.S. Department of the Interior & Bureau of Reclamation, 2003).

Thermal technologies employ the principle of evaporation and condensation to heat up the saline water, whereby the water released in form of vapour is condensed to obtain the freshwater leaving behind the salt. It is also called distillation. This has been utilized in treating seawater but because of the expensive cost it has not been employed in treating brackish water. Several sub-categories of this technology include multi-stage flash distillation (MSF), multi-effect distillation (MED) and vapour compression distillation (VCD) (U. S. Department of the Interior & Bureau of Reclamation, 2003).

Membrane technologies employ the principle of dialysis and osmosis in separating salt from water via a membrane or filters. Membrane and filters work by selectively allowing or prohibiting the passage of certain substances. There are various forms of membrane with different particle sizes. Membranes that permit large particles to go through are employed during pre-treatment. Membrane technologies are further divided into electrodialysis/electrodialysis reversal (ED/EDR) and reverse osmosis (RO) (U.S. Department of the Interior & Bureau of Reclamation, 2003). These technologies have been used for treating brackish water, seawater is not left out as several forms of them are been employed in its treatment. Figure 2 shows the contribution of desalination technology across the whole world (Esmaeilion, 2020) where ED and RO desalination techniques account for over 50% of all desalination capacity. A good number of them now utilize filtration system before the membrane to eliminate impurities that affect lasting filtration process.

# 3 Renewable Energy-Assisted Desalination

Energy can be generated from inexhaustible resources such as wind and sun known as renewable energy sources. Renewable energy technologies and desalination plants are different systems that can be coupled together in several ways where the energy generated from the renewable energy system in form of thermal energy, electricity or shaft power is used to power the desalination plant. Figure 3a and b



Fig. 2 Contribution of desalination technology across the whole world

(Ahmadi et al., 2020) shows the combinations of renewable energy sources to desalination technologies and the percentage of their usage worldwide in 2017, respectively. Desalination process can be driven by thermal or electrical means as the primary energy type which can be from geothermal, tidal, solar energy or wind energy source where solar energy accounts for over half, wind energy accounts for three-tenth while geothermal and tidal energy accounts for the 19% remaining share tagged as others. The status of desalination system powered by renewable energy technologies is shown in Fig. 4 (Ahmadi et al., 2020).

Wind and photovoltaic (PV)-powered RO systems have been well-researched and have commercial application while geothermal-powered membrane distillation (MD), windpowered ED and ocean-based RO are still at basic research level. The major drawback of solar and wind energy sources is their unpredictability and dependency on weather and climate (Esmaeilion, 2020). Application of renewable energy as energy source for desalination process offers lots of advantages (Mohammed & Zhengming, 2014) as it will be of help to regions with limited water supply and less grid power supply. This is common in developing countries and islands. The benefits of this technology include less cost, less pollution, non-depleting source, abundant in nature (Fdasm, 2014). Also, most of these developing countries have access to some of these renewable energy resources such as high sunlight energy and wind among others.

The gradual rise in desalination capacity across the globe presently is over.

70 million  $m^3/day$ , use of renewable energy option can lead to a significant reduction in greenhouse gases. Furthermore, the inexpensive attribute of renewable energy solutions makes it a better option, particularly in rural areas with low population density and poor facilities for portable



Fig. 3 a and b: Combinations of renewable energy sources to desalination technologies and the percentage of their usage worldwide in 2017, respectively



Fig. 4 Status of desalination system powered by renewable energy technologies

water and electricity transmission and distribution. The effective combination of renewable energy technology with large-scale desalination plants at affordable cost has the potential of solving the present-day environmental problems and in near future (Jenny, 2003).

Implementation of combined effort from solar and wind energy in desalination systems has been utilized and seems to be promising more than other renewable desalination combinations (Ahmad & Rached, 2018). Also, several works have been captured in the literature in this regard (Al-Hussaini & Smith, 1995; Al-Shammiri & Safar, 1999; Baltas et al., 1996; BenJemaa et al., 1998; Caruso & Naviglio, 1999; Delyannis & Belessiotis, 1996; El-Nashar, 1992; Garcia-Rodriguez, 2002, 2003; Garcia-Rodriguez & Gomez-Camacho, 2002; Goosen et al., 2000; Hanafi, 1994; Kamal et al., 1999; Leitner, 1998; Mathioulakis et al., 2007; Mohsen & Al-Jayyousi, 1999; Rheinlander et al., 1998; Rodriguez-Girones et al., 1996; Rognoni & Trezzi, 1999; Singh et al., 1995; Tiwari et al., 1997; Voivontas et al., 2001; Zaki et al., 1993). Desalination driven by biomass, hydropower and geothermal energy is still at the developmental stage through using biomass and hydropower holds no future as this will require more freshwater than it could produce during desalination.

# 3.1 Solar-Assisted Desalination Process

This method applies solar energy in producing water with low concentration from seawater or brine. The benefit of using solar radiation over other renewable energy option is that its thermal energy can be directly used to power desalination plants without the unavoidable energy loss related to energy conversion process (Kalogirou, 1998). The solar energy desalination method can be in two ways: indirect solar (i.e. solar PV) desalination process in which electricity is generated from solar cells to drive the desalination process and direct solar desalination process which utilizes direct heat from the sun to perform desalination process.

#### a. Direct Solar Desalination Process

This process employs direct sun rays to produce portable water via the use of solar stills or solar pond. It involves the use of boiler in heating up saltwater, thereby converting the water to gaseous form, which is captured and then channelled into sets of condensing coil. This is also referred to as solar thermal distillation. The quantity of water produced is a function of the solar surface area and the angle of incidence. Thermal desalination requires lots of energy especially in locality where the salt concentration in the water is much, and these areas have ample supply of solar energy. Water shortage with high sun intensity makes solar energy the right option to solve the water shortage problem.

#### i. Solar Still

This is a very simple desalination techniques that are economical and well-preferred to conventional methods that require more energy input. It uses the principles of evaporation and condensation in purifying water. The unclean water is heated up using direct sunlight energy causing a temperature rise, thereby converting the water into steam. The steam water evaporates leaving behind impurities and in turn slowly condenses forming clean water droplets that can be utilized. They are employed in communities with limited water supply where clean water needs to be produced from dirty water. The solar still system comprises of sloping transparent glass and black trough. The salty/impure water is introduced into the trough where it is heated up using energy from the sun to evaporate water which condensates and is collected from the sloppy glass. It can be employed to produce clean drinkable water at a small-scale level (Mohammed & Zhengming, 2014).

Figure 5a and b shows a schematic diagram of a modified solar still and conventional solar still (Gupta et al., 2017). The modified solar still has incorporated in it cuprous oxide nanoparticle to increase thermal conductivity of water, white paint coating on the walls to increase solar reflectivity of solar radiation inside the basin and use of thin water film on the glass cover to reduce temperature.

Different configurations of solar still have been reported in the literature, and they include pyramid single basin, tabular, single effect, double effect, hemispherical, spherical, inverted absorber, double basin, pyramid multi-basin, triple basin, evacuated multi-stage and multi-basin solar still (Ravishankar et al., 2013). Advantages of solar still include the cost-effectiveness; the source of energy which is the sun is freely available and low maintenance. However, the disadvantages include the low production efficiency, boiling does not occur in solar still making it unable to break down toxic substances and destroy microorganisms, the wide-area sloppy glass cover may attract insects and the desalination process can only be achieved at some hours of the day resulting from unpredictable nature of solar energy.

## ii. Solar Ponds

These are water body with small depth that receives and stores solar energy (Yogi et al., 2014). It is a large-scale energy collector with integral heat reservoir for distributing heat energy. It applies the principle of heating up water by the sun rays causing it to expand and rise as it becomes



Fig. 5 a and b: Schematic diagram of a modified solar still and conventional solar still, respectively

lighter or less dense. As it rises upward to the surface it loses its heat to the air via convection. The colder water at the surface moves downwards to replace the warm water at the bottom creating a convective circulation that ensures water mixing to release heat. Hence, solar ponds are designed to reduce convective effect in other to store the solar energy received by the pond by dissolving salt in bottom zone making it too heavy to rise. Examples of solar ponds include lakes and artificial basins. They are advantageous because they can be constructed anywhere, less expensive and construction process is less complex (Saifullaha et al., 2012). They are the best heat storage option that is convenient and inexpensive and has the capacity that spans through the season. It can effectively store more heat when compared with a water body of the same size because the salinity gradient prevents convection current. Some of the drawbacks of solar ponds include the need for large expanse of land, high level of solar energy input, large supply of saltwater and constant maintenance. The three basic layers of a solar pond are outlined below:

- i Top layer;
- ii Middle layer; and
- iii Bottom layer.

These layers are structurally shown in Fig. 6 which shows the schematic view of a solar pond (Sajjadnejad et al., 2020).

#### The Top Layer

This layer is also known as the upper convective zone. This is the surface region that is at atmospheric temperature with small amount of salt. It serves as the solar receiver and is relatively shallow in depth. It requires constant flushing with freshwater to prevent the piling up of salt since the lower layer usually diffuses through the saline gradient over time (Fisher et al., n.d.).

#### The Middle Layer

This is also referred to as gradient layer which is known as a non-convective region where the salinity grows from top part to the lower part of the region. It is also called the insulation region with high salinity than that of the top layer. In this part, the saltiness increases with depth as well as the temperature. It is much thicker and occupies more than half the depth of the pond (Farshad & Mohammadnab, 2018). The water in this region cannot rise or sink thereby restricting convection current, hence acting as transparent insulator (Osamah et al., 2020; Saifullah & Arif, 2012). This transparent insulator allows sunlight to reach the bottom layer entrapping it there.

# **Fig. 6** Schematic view of a solar pond

This layer or region is hot with high amount of salt. It receives and stores energy in form of heat. The surface of this layer is dark, thereby increasing the absorption of sun energy. Therefore, it serves as heat storage zone (Saifullah & Arif, 2012). The thickness of this layer determines the amount of heat that can be stored and used later. The heat at this level is transferred to pipes circulating through the pond to extract heat energy.

#### b. Indirect Solar Desalination

This system is vital in improving the freshwater productivity. The resulting productivity from direct solar desalination system is poor to be applied in non-residential operation. Indirect solar desalination uses two separate systems;

- i a solar collection array and
- ii a separate conventional desalination plant.

#### Solar Photovoltaic Desalination System

Photovoltaic systems convert energy from the sun into direct current electricity using semiconductor materials that show photovoltaic effect (Ridha, 2015). A solar cell is the basic unit of PV that performs this operation. Solar cell relies on a quantum–mechanical process called photovoltaic effect to generate electric current. A typical solar cell comprises of a p–n junction formed in a semiconductor material. Figure 7 shows schematic diagram of PV basics (Sajjadnejad et al., 2020). In order to have a higher output, solar cells are joined







together to form a module, modules are joined together to form a panel, and panels are joined together to form an array.

Solar photovoltaic system is used in connection with established desalination techniques like RO, MSD, multiple-effect evaporation, or ED. The application of PV cells with ED is more attractive than RO for rural communities where solar energy and salty water are obtainable round the season (Nwaigwe et al., 2019). Battery energy storage units can be incorporated into the PV system to stabilize the power fed into the input desalination system. The schematic illustration of photovoltaic–reverse osmosis (PV-RO) system with battery storage is shown in Fig. 8 (NREL, 2016). The battery bank can also serve as backup to the system in the absence of solar radiation due to the unpredictable nature of the sun.

# 3.2 Wind-Powered Desalination

Wind is a moving air that results from uneven heating of the earth's surface by the sun. This gives rise to air flowing from high-pressure regions to low-pressure regions. The larger the atmospheric pressure gradients, the stronger the wind, the higher the wind speed, and thus, the greater the power that can be captured from the wind.



**Fig. 8** Schematic diagram of photovoltaic–reverse osmosis (PV-RO) system with battery storage
The wind turbine traps the wind's kinetic energy in a rotor comprising of two or more blades joined to an electrical generator via a mechanical means. The turbine, which is usually placed on top of a tower to improve the energy capture, generates electrical and mechanical energies that are utilized in driving desalination system. The amount of energy that can be generated by a turbine from the wind depends on wind speed, air density and swept area/blade radius. Dense air exerts more pressure on the rotors resulting in high power output and high wind speed translates to more power. It is important to locate wind turbines in area of with regular wind supply than areas with seasonal high speed (Donev et al., 2020). Desalination techniques suitable to be driven by wind turbines are RO, MVC and ED. Electricity generated from the wind can be employed in driving both the compressor and the heating element in the vapour compression process and to run the pump in the RO process (Ghaffour et al., 2015). Wind-powered RO process is the most developed system even though we have very few pilot plants of wind-driven MVC and ED (Mehdi & Gleb, 2009). Rent-a-port green energy designed a wind-powered desalination plant that couples wind energy to RO unit that purifies seawater for irrigation use (Rent-A-Port Green Energy, n.d.). A layout for wind-powered RO is shown in (Fig. 9, Clayton et al., 2014). Wind-powered desalination is one of the most promising application of renewable energies for desalination that offers reduction in desalination carbon and energy footprint (Water world (016), n.d.).

While the intermittency of wind as an energy resource is inherently unavoidable and stands in direct conflict with a preference for steady-state operations, the problems of intermittency in wind-powered desalination can be technically solved by incorporating battery energy storage units into the system. The battery bank can also serve as backup power supply to the whole desalination system in the period 249

of low wind energy availability mostly night-time. Regardless of the difficulties associated with intermittent power production, it seems that wind power desalination represents a promising coupling of innovative technologies (Andy et al., 2008).

#### 3.3 Geothermal-Powered Desalination Process

This is another mature technology even though not common as the solar or wind energy type that holds a promising future for desalination process. Investigation with respect to geothermal-powered desalination has been carried out theoretical using preheated water from thermal springs or hot groundwater for either indirect desalination with electric generation first or direct desalination by heating the feed water (Moshen, 2010). Geothermal heat can be employed in powering MSF, MED, MD and adsorption desalination (AD) technologies. Figure 10 is an illustration of geothermal-powered MED and AD desalination systems (Missimer et al., 2016).

Its utilization in MD and AD is still at their developmental stage (Nwaigwe et al., 2019). This system utilizes geothermal energy to power desalination process at a competitive cost. It is more advantageous in terms of thermal storage which is not needed because of its continuous and predictable nature (Barbier, 2002; Missimer et al., 2016; Report, 2001), and it also has constant stable energy (Eltawil et al., 2008). Heat storage is costly and from technical standpoint is not much in size (Al-Karaghouli & Kazmerski, 2011). A high-pressure geothermal type permits the direct utilization of shaft power on mechanically powered desalination, while high-temperature geothermal type can be utilized to produce electrical power to power RO or ED system. It has been used in combination with solar still in hybrid system (Ghaffour et al., 2011).





Fig. 10 Geothermal-powered MED and AD desalination systems

# 4 Hybrid Renewable Energy-Powered Desalination

Renewable energy sources can be combined to obtain a hybrid energy system that is capable of powering desalination process. They offer a viable and cleaner option to conventional systems. There are lots of studies on hybrid system comprising of wind turbines and solar PV. A proposed hybrid system for powering house load has been studied by Mohammed and his colleagues and this idea could also work for desalination technology. Figure 11 shows a modified proposed hybrid (wind and PV) renewable energy system which can power desalination unit (Mohammed et al., 2019). Figure 12 shows modified hybrid (wind, geothermal and PV) renewable energy system that can power desalination unit (Mohammed et al., 2019). Also, hybrid system PV/wind/diesel/battery power system for RO desalination unit has been studied by Xu and his colleagues where determination of the lowest levelized cost of energy was their major interest (Xu et al., 2019). Economic analysis of hybrid PV-wind-RO shows a meaningful reduction in the cost of water production (Khan et al., 2018). Combination of solar still with wind-water heater in a hybrid system is advantageous in terms of ensuring system operation during day and night even in cloudy days with good wind and enhanced distillation output (Moh'd et al., 2016). In the City of Kerkennah Island, freshwater was supplied from a desalination unit powered by a hybrid renewable energy system comprising of wind turbine, photovoltaic generator and fuel cell (Smaoui & Krichen, 2014). A more recent research shows that the use of hybrid renewable energy

system coupled to RO desalination unit offers great benefits in terms of cost reduction and zero pollution with high reliability for remote areas (kiehbadroudinezhad et al., 2020). In addition, hybrid solar–geothermal energy source for seawater desalination has been proposed and it offers potential benefits such as it allows geothermal heat regeneration and eliminates the need to build a thermal storage for night-time operation (Yousefi et al., 2017). Investigation by Voropoulos et al. (2004) and friends' shows that a hybrid system consisting of solar still coupled with solar collector field and hot water storage tank gave a higher freshwater output with the advantage of supplying hot water from its storage tank.

#### 5 Limitations and Future Prospects

The intermittency nature of wind and solar energy resources is a challenge to the utilization of their full potential in desalination units. This is caused by the unpredictable weather and climate changes. For instance, the amount of solar radiation that reaches any part of the earth is influenced by geographical location, time of the day, season, local landscape and local weather (US Department of Energy, 2013). Also, during summer we have abundance of sunshine and less in winter. The shorter days in the winter and fall reduce solar energy generation. Cloud, snow and foliage affect the amount of solar energy generated to power a desalination unit. High initial or upfront cost is another major challenge of solar energy, geothermal energy and wind energy utilization in water treatment even though in the long run they offer to save cost.



Fig. 11 Modified proposed hybrid (wind and PV) renewable energy system which can power desalination unit



Fig. 12 Modified hybrid (wind, geothermal and PV) renewable energy system that can power desalination unit

The future prospects should focus on the application of different combinations of hybrid renewable energy powered desalination system as it offers the benefit of better efficient system. This should be properly designed and located to tackle the drawback of individual energy system. Also, exploring some fragmentary research areas with respect to desalination technologies like forward osmosis, dew vaporization, freeze desalination can provide opportunity for enhancing efficiency of desalination process (Esmaeilion, 2020).

## 6 Conclusion

Water scarcity can be devastating as most of the domestic and industrial activities rely on it. Availing freshwater from seawater and brackish water to solve water scarcity issue is needful but proper treatment is also a necessity. Utilizing desalination technologies with integration of renewable energy technologies in proper water treatment has gained lots of research interest in recent times, and several works have been done in this regard. The implementation of research results in locality facing water challenges which require in-depth study and investigation on cost analysis and process optimization of all processes involved such as water collection, treatment, storage and distribution and energy requirement. This is very important as it will help the government and policymakers to have better knowledge of what it takes to implement such technologies to avail portable water for affected regions.

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