

Carbon Nanotubes Composite Membrane for Water Desalination

Shabnam Taghipour, Ali Khadir, and Mohammadhossein Taghipour

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

Department of Civil Engineering, Sharif University of Technology, P.O. Box 11155-9313 Tehran, Iran e-mail: sh.taghipour70@student.sharif.edu

A. Khadir (\boxtimes)

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](#page-19-0); Taghipour & Ayati, [2017](#page-20-0); Taghipour et al., [2017](#page-20-0), [2019](#page-20-0)). 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](#page-17-0)). 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](#page-21-0)). 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\)](#page-19-0).

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,

S. Taghipour (\boxtimes)

Young Researcher and Elite Club, Yadegar-e-Imam Khomeini (RAH) Shahre Rey Branch, Islamic Azad University, Tehran, Iran

M. Taghipour

Department of Materials Engineering, University of Tabriz, P.O. Box 51666-16471 Tabriz, Iran

and flocculation) (Das et al., [2014;](#page-18-0) Gerba & Pepper, [2019](#page-18-0)), secondary treatment refers to biological treatment of water including aerobic and anaerobic treatment methods (Gupta et al., [2012\)](#page-18-0), and tertiary treatment benefits from advanced treatment options for further reduction in residual turbidity, metals, and pathogens (Gerba & Pepper, [2019](#page-18-0)) (such as distillation, precipitation, crystallization, membrane technologies, ion exchange, solvent extraction, and so many other advanced oxidations) (Das et al., [2014\)](#page-18-0).

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](#page-18-0); Mathew et al., [2014\)](#page-19-0).

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](#page-18-0)). 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;](#page-18-0) Sadeghi, [2016;](#page-20-0) Strathmann, [2000\)](#page-20-0). 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\)](#page-19-0). 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](#page-20-0)).

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](#page-20-0)). 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](#page-19-0)). The flux across the membrane is calculated by Eq. 1 (Nath, [2017\)](#page-20-0):

Flux =
$$
\frac{\text{Membrane permeability}}{\text{Membrane thickness}} \text{(Diving force)} \qquad (1)
$$

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](#page-21-0)):

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

where ε is the surface porosity, r_p is pore radius, μ is viscosity of the aqueous solution, and $\delta_{\rm m}$ 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](#page-21-0)):

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

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](#page-21-0)).

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](#page-19-0); Mandal & Kulkarni, [2011\)](#page-19-0). 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](#page-18-0)).

In concentration-gradient-driven processes such as dialysis, the fluid fluxes across a membrane due to the existing concentration difference (Youravong & Marthosa, [2017\)](#page-21-0).

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\)](#page-18-0).

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](#page-18-0); Tan & Rodrigue, [2019\)](#page-20-0).

Characteristics of different types of membranes are pre-sented in Table [1](#page-3-0) (Belleville & Vaillant, [2016](#page-18-0); Nath, [2017\)](#page-20-0).

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](#page-20-0)). 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](#page-21-0)). 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](#page-18-0)).

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\)](#page-21-0). 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\)](#page-18-0) with permission from Woodhead Publishing, Elsevier

[2011\)](#page-20-0). Considering high operational costs, ceramic membranes are suggested for small-scale experiments (Nandi et al., [2008](#page-19-0)). Moreover, numerous hybrid membranes have been developed and utilized which in turn can cause for high operational costs (Hosseini et al., [2020;](#page-18-0) Sadeghi et al., [2016\)](#page-20-0).

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 (≤ 100 nm) in at least one dimension (Salisbury et al., [2018\)](#page-20-0).

Hereunto, numerous NMs have been successfully utilized for water remediation goals (Beheshti et al., [2019](#page-18-0); Ghenaatgar et al., [2019](#page-18-0); Khadir et al., [2020a](#page-19-0), [2020b](#page-19-0); Khorsandi et al., [2019](#page-19-0); Mirjavadi et al., [2019](#page-19-0); Mohammadi et al., [2019](#page-19-0); Mollahosseini et al., [2019](#page-19-0); Piri et al., [2019](#page-20-0), [2020\)](#page-20-0). 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](#page-19-0)).

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 sp² hybrid carbon–carbon bonds which is the strongest chemical bond (Paula et al., [2016\)](#page-18-0). CNTs are classified to multi-walled carbon nanotubes (MWCNs) and single-walled carbon nanotubes (SWCNs) (Sielicki et al., [2020](#page-20-0)). 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](#page-18-0)). SWCNs consist of a flattish array of benzene molecules with honeycomb-shaped rings of carbon–carbon bonds (Nayfeh, [2018](#page-20-0)). A schematic presentation of SWCN and MWCN can be seen in Fig. [3.](#page-4-0)

Different comparable properties of SWCNs and MWCNs such as purity, density, and thermal and electrical-related coefficients are presented in Table [2](#page-4-0) (Awad, [2016;](#page-18-0) Chaudhary et al., [2013;](#page-18-0) Sattar et al., [2015\)](#page-20-0).

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](#page-19-0)).

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\)](#page-20-0).

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\)](#page-18-0). The concept of using CNT membranes was first raised by Sun and Crooks ([2000\)](#page-20-0). Based on the type of morphology, CNTs can be directly grown in three shapes including (Gao & Kono, [2019\)](#page-18-0):

- (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](#page-18-0)).

In CVD approach (Fig. [4\)](#page-5-0), 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](#page-18-0)). 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](#page-18-0)).

In LA approach (Fig. [5\)](#page-5-0), 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;](#page-18-0) Du et al., [2017](#page-18-0)). Here, the catalyst acts as a cage structure opener of

Table 2 Comparison between different parameters of SWCNs and MWCNs (Chaudhary et al., [2013](#page-18-0); Sattar et al., [2015\)](#page-20-0)

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\)](#page-20-0).

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\)](#page-18-0). A schematic presentation of the AD approach for synthesization of CNTs can be seen in Fig. [6.](#page-6-0)

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\)](#page-18-0). 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\)](#page-21-0).

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\)](#page-19-0). Application of VACNTs was first reported by Kalra et al. ([2003\)](#page-19-0) by using molecular dynamics. The osmotic force-driven flow passes through the membrane due to the salt concentration gradient (Suzuki, [2013\)](#page-20-0). 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 $(Ru(NH_3)_{6}^{3+})$ solution in VACNTs with inner diameter of 6 nm (Ang et al., [2019](#page-17-0)).

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](#page-19-0)), parylene, silicon nitride (Liu et al., [2019](#page-19-0)), and epoxy (Lee et al., [2015](#page-19-0)) 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\)](#page-19-0).

Steps for fabrication of VACNT membranes are as follows (Das, [2017\)](#page-18-0): (1) deposition of non-conducting substrates (i.e., $SiO₂/Si$) 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_2 , 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;](#page-18-0) Krishnakumar et al., [2012](#page-19-0)).

Owing high thermal, chemical, and mechanical durability, porous ceramic membranes have been extensively used as support for CNTs (Kayvani Fard et al., [2018](#page-19-0)).

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 $300 \mu \text{m}$ 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](#page-7-0) (Trivedi & Alameh, [2016](#page-21-0)).

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](#page-21-0)).

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](#page-18-0)). 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](#page-19-0)).

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\)](#page-21-0). Adopted from Graff ([2012\)](#page-18-0). Copyright © 2016, Springer Nature

et al., 2017). The number of shells (N) in a CNT can be evaluated by Eq. 4 (Todri-Sanial, [2016](#page-21-0)):

$$
N = \frac{d_{\text{out}} - d_{\text{in}}}{S_{\text{a}}}
$$
 (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\)](#page-19-0). After successful fabrication of HACNTs, studies focused on their growth mechanism. Numerous processes such as ultralow feeding gas flow guiding (Li & Zhang, [2019\)](#page-19-0), external electrical fields (Morais et al., [2019](#page-19-0)), and fast-heating CVD (Huang et al., [2004](#page-19-0)) 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](#page-20-0)).

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\)](#page-21-0).

An et al., reported fabrication of 1-cm-long HACNTs on $Si/SiO₂$ 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](#page-8-0)). 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](#page-17-0)).

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\)](#page-20-0). 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\)](#page-8-0).

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;](#page-20-0) Sengupta, [2018](#page-20-0); Zhang et al., [2013\)](#page-21-0). A brief description of these classes are presented in the following:

- 1. 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\)](#page-18-0).
- 2. 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. [\(2014\)](#page-17-0) 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](#page-9-0)) (Zhang et al., [2014\)](#page-21-0). Poor interaction of the nanocatalyst and substrate surface can be another reason for this phenomenon (Gohier et al., [2008\)](#page-18-0). 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](#page-21-0)).

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\)](#page-9-0). 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\)](#page-20-0).

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](#page-18-0)). 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\)](#page-21-0). 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](#page-21-0)). 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](#page-21-0)).

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_2, TiO_2) , and fullerene) (Roy & Singha, [2017\)](#page-20-0). 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](#page-20-0)). In addition, this technology provides higher hydrophilicity, permselectivity, and resistance against pH and temperature changes (Maghami & Abdelrasoul, [2018\)](#page-19-0). 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](#page-10-0).

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\)](#page-18-0). 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\)](#page-17-0), layer-by-layer deposition (LbL) (Chimisso et al., [2020\)](#page-18-0), chemical grafting (Ursino et al., [2018\)](#page-21-0), and physical and chemical deposition of NPs (e.g., dip coating, spin coating, hot pressing, etc.) (Xiao et al., [2016](#page-21-0)).

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](#page-10-0)) 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\)](#page-21-0).

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](#page-18-0)):

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\)](#page-20-0).

4 Application of CNT Membranes in Desalination

Fabrication of CNT nanocomposite membranes for desalination of sodium hydroxide was first reported by Shawky et al. [\(2011](#page-20-0)). Nowadays, CNT membranes are widely utilized as vital materials for water treatment, especially in desalination because of their rapid transportation ability (Ma et al., [2017\)](#page-19-0). 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](#page-19-0)). 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\)](#page-21-0).

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^3 /day, respectively (The International Desalination Association (IDA), [2020\)](#page-20-0). 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](#page-21-0)). 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](#page-20-0)). 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](#page-12-0)) (Hanasaki et al., [2016\)](#page-18-0). The worldwide installed desalination capacity for different feed water types and their contribution percentage compared with each other is presented in Fig. [16.](#page-12-0) The pure water is mainly utilized by industries which require high-quality water (i.e., food and drug industries) (Khan & Ali, [2018\)](#page-19-0).

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 $^{\circ}$ 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.](#page-12-0) 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\)](#page-19-0).

Scrutinies of transmission electron microscopy (TEM) images (Fig. [18\)](#page-13-0) 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](#page-20-0)) (accessed date: 13 May 2020)

Table 3 Classification of different feed water types (Hanasaki et al., [2016](#page-18-0))

Fig. 17 Water flux and salt rejection of fabricated membranes under various conditions $(C_{\text{NaCl}} = 2000 \text{ mg/L},$ $P = 15.5$ bar). Adapted from Kim et al. ([2014](#page-19-0)). 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](#page-19-0)). Copyright © 2014, American Chemical Society (ACS)

lead to the reduction in salt rejection values (Kim et al., [2014\)](#page-19-0).

Ragunath et al. [\(2018](#page-20-0)) 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 L/m²h to 51.4 L/m²h 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^2 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/m²h, respectively (Ragunath et al., [2018](#page-20-0)).

Sun et al. ([2019\)](#page-20-0) 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 $(FeCl₃)$ 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 ± 2.3 L/m²h at a temperature difference of 40 °C. By 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 ± 0.5 , 2.73 ± 1.0 , 4.08 ± 0.9 , and 8.42 ± 1.5 L/m²h, respectively. In all membranes, approximately complete (>99.9%) salt rejection was achieved at various temperature differences (Sun et al., [2019](#page-20-0)).

Anga et al. ([2018\)](#page-17-0) investigated the effect of different CNT sizes on the desalination of 1.2 M salt through slit confinements formed by HACNTs. Figure [19a](#page-14-0) 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;](#page-19-0) Suk & Aluru, [2010](#page-20-0)) 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](#page-17-0)).

In another study conducted by Li et al. ([2019\)](#page-19-0), 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 ± 150.3 to 58.9 ± 6.2 LMH) and increase salt rejection (from 94.9 to 98.8%) (Fig. [20\)](#page-15-0) (Li et al., [2019](#page-19-0)).

Dong et al. ([2018\)](#page-18-0) represented fabrication of a novel superporous, superhydrophobic, and thermally resistant ceramic CNT by MD process. This membrane exhibited significant desalination potential $(99.9\% \text{ of Na}^+)$, water flux
 (37.1 J/m^2) and distillate conductivity (0.09 uS/cm) (37.1 L/m²h), and distillate conductivity (0.09 μ 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](#page-18-0)).

Ang et al. ([2019\)](#page-17-0) 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 (ΔP_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\)](#page-17-0) 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\)](#page-19-0) 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](#page-17-0)), 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\)](#page-17-0).

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](#page-17-0)).

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](#page-16-0).

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](#page-16-0) (Ali et al., [2019\)](#page-17-0).

Rejected ion type	Pore size (nm)	Efficiency $(\%)$	References	
$Na+$	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^+	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\)](#page-17-0)

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 \times 10 ⁻⁷ L/MPa s	\sim 3 \times 10 ⁻¹² L/MPa s	\sim 4 \times 10 ⁻¹¹ L/MPa s	\sim 5 \times 10 ⁻¹¹ L/MPa s	$\overline{}$		
Applied pressure	Varied with type of application	$30-60$ bar	$20 - 40$ bar	$1-10$ bar	< 1.0 bar		
Features	• Less energy consumption • Good performance • Tolerant against harsh environmental condition • High durability, fouling resistance, and cost-effective	• High-energy consumption • Good performance • Less tolerance in harsh environmental condition • High durability, fouling resistance, and cost-effective	• High-energy consumption • Good performance • Less tolerance in harsh environmental condition • Less durability, prone to fouling, and less cost-effective	• Moderate energy consumption • Moderate performance • Less tolerance in harsh environmental condition • Less durability, prone to fouling, and less cost-effective	• Moderate energy consumption • Moderate performance • Less tolerance in harsh environmental condition • Less durability, prone to fouling resistance, and less cost-effective		

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\)](#page-21-0). 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](#page-18-0)). 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](#page-20-0)). 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](#page-18-0); Esfahani et al., [2019\)](#page-18-0). 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\)](#page-21-0). 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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