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Application of carbon nano-materials in desalination processes

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ABSTRACT

Scarcity of freshwater resources increases the importance of seawater and brackish water desalination processes. However, still, large amount of specific energy requirement, leading to high operational costs, presents a big challenge in adopting desalination technologies. This challenge can be addressed by considering the newly emerging nanomaterials especially those made from carbon. This paper presents a comprehensive literature survey and review that brings those carbon nano-materials (CNMs) into focus which directly participate in desalination processes. The structural and functional properties of CNMs, their fabrication into membranes, their formation into bucky papers, and their composite electrodes are some of their usages in desalination processes which are exploited. The survey and analysis of the available literature shows that CNMs can enhance capacity and efficiency of next generation desalination systems, particularly reverse osmosis, membrane distillation, capacitance deionization, and forward osmosis.

Keywords: Desalination; Carbon nano-materials (CNMs); Membrane nanocomposites; Bucky papers

1. Introduction

Scarcity of fresh water is among the world's biggest problems of today. Presently, about three billion people on earth are unable to reach clean drinking water [1]. It is predicted that two third of all the countries on the globe will be suffering, to a certain extent, from water shortage by 2015 [2]. The problem of access to safe drinking water is further complicated by climate change, rapid industrialization, population growth, and contamination of existing water resources [3]. Many solutions are proposed to solve this grand issue of water scarcity, including repair of existing infrastructure, improvement of water distribution systems, conservation of existing water sources, and rainwater harvesting. Although these solutions can make the existing water resources more efficient, yet they are unable to increase them. Water supply can only be increased beyond hydrological cycle by the application of saline water desalination and water reuse [4]. Since seawater carries the ability to provide unlimited steady supply of water without interfering the naturally occurring freshwater ecosystems, therefore, there has been an increased growth in the number of seawater desalination plants in the last couple of decades [2,5,6]. By 2016, desalination is expected to provide more than 38 billion m^3 freshwater [5].

Desalination processes can be broadly classified into thermal (phase change) and membrane-based processes as shown in Fig. 1. Phase change processes

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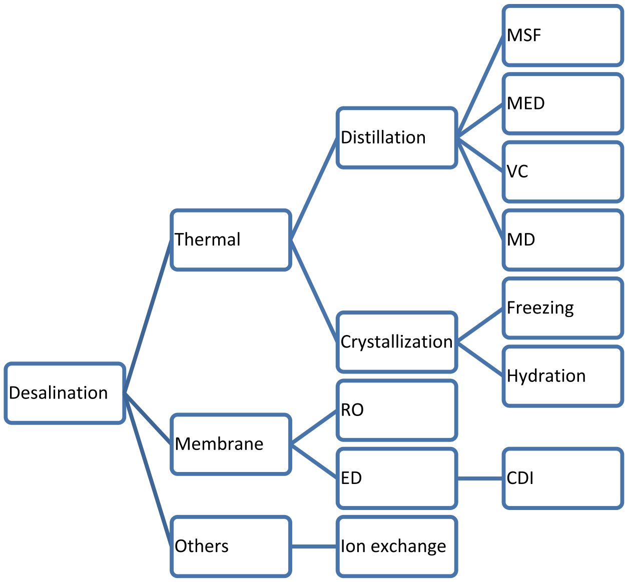

Fig. 1. Classification of desalination technologies [9].

include multiple-effect distillation (MED), multi-stage flash (MSF), freezing, vapor compression, and humidification–dehumidification (HDH), whereas the reverse osmosis (RO), electrodialysis (ED), forward osmosis (FO), capacitance deionization (CDI), and the membrane distillation (MD) are popular membrane-based processes [7]. Among these technologies, RO and MSF are of ultimate importance because they constitute 53 and 25% of total desalination capacity, respectively, Fig. 2 [6,8]. MED is expected to be the future leading thermal technology, while HDH, MD, FO, and CDI are still categorized as emerging ones.

MED, MSF, and RO consume at least 26.4, 23.9, and 3.6 kWh of energy for each cubic meter of clean water produced, respectively [11]. This high-energy consumption is the biggest barrier in expanding desalination facilities. Therefore, an increase in the energy efficiency and decrease in the costs related to both desalination and pretreatment processes are required to cope with the challenge of increased global demand of clean water. This can be achieved by optimizing the components and minimizing the thermal losses and fouling (which leads to periodic cleaning) of MED and MSF plants. Similarly, RO plants can be made economical by devising methods for maximizing the flux of water through RO membranes, minimizing fouling (bio-fouling and salt deposition), and decreasing the pressure required for the process [12].

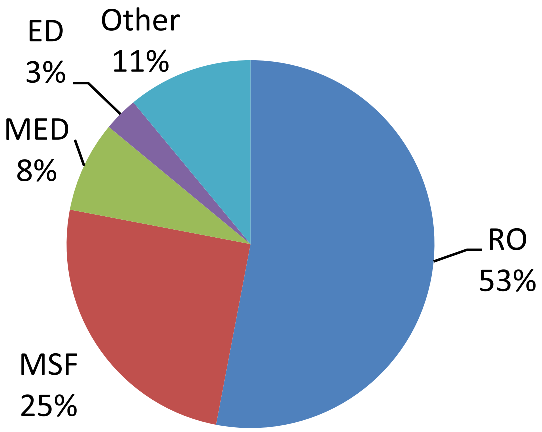

Fig. 2. Global desalination capacity by process [8,10].

Recently developed carbon nano-materials (CNMs) have a potential to decrease costs by increasing energy efficiency in desalination processes. The unique physical, chemical, and electronic properties of CNMs, which are due to their hybridization state (structural confirmation) [13], have made them a successful contributor toward research in drug delivery, electronics, structural materials, bio-imaging, bio-sensing, and energy conservation [14]. The single- and multi-walled carbon nanotubes (CNTs), fullerenes, and graphene are among the CNMs which are successfully applied to various environmental problems [15]. Recent researches indicate their potential to successfully contribute toward desalination technologies. CNMs are either applied directly or used to modify a material to improve its workability and efficiency. The objective of this paper is to provide a comprehensive review on CNMs with the potential to advance desalination technologies. The primary emphasis will be on the use of CNMs in RO, MD, CDI, and FO systems.

2. Nanotechnology

The term "nano" is used for structures and processes which carry, at least, one dimension between 1 and 100 nm. Nano science and technology deals with the preparation and application of structures at nanometer scale. Roughly, a nanometer is a size of 3–5 atoms stalked together and this is a new technology for the scientists working with bulk properties (>100 nm) or at atomic level (<1 nm). Nanotechnology originated due to advancement of probing and fabricating structures at nanometer level. So far, engineers have built the existing infrastructure while working with bulk properties of materials as against chemists and physicists who developed experimental as well as theoretical techniques for probing molecular and/or atomic structure of matter. Recently, experimental tools are devised which have helped to investigate the properties of materials at intermediate scale (between bulk and atomic) or nanoscale.

Nanoscale is significant because van der Waals interactions and surface tension play a prominent part when compared to that of gravity. Microscopic techniques, such as atomic force microscopy, tunneling microscopy, and transmission and scanning electron microscopy, have enhanced the interest of researchers in this newly emerging field. Development in nano science and technology is a result of new techniques for fabricating and testing nano structures having useful properties. These properties are, otherwise, unachievable.

Nanotechnology is important for desalination, because it is carried out at atomic and molecular levels, and desalination process (membrane and distillation) employs process properties at nanoscale as well. For instance, dissolution of salt in water can be expressed by the activity of individual atomic ions of salt with bulk matrix of water. Similarly, interaction of dissolved salts and water with membrane surfaces, mineral nucleation resulting in formation of scale on heat exchanger and/or membrane, ion transport in ion exchange membranes are the processes which occur at nanoscale because they involve larger aggregates of molecules. Neither individual molecules nor bulk scale investigations can characterize them. Membranes, especially, have interesting structures on the nanometer level.

3. Development in RO membranes

RO is one of the fastest growing desalination technologies because of its lower specific energy consumption. Energy consumption can be further decreased in RO due to the development of new membrane materials, scaling inhibitors, bio-fouling reducers, and energy recovery devices. Upon optimum modifications, RO can save up to 92 KWh energy for every cubic meter of freshwater produced when compared to conventional MSF [6]. Although, notable advances have been made in the development of RO membranes [16–19]; yet, future water challenges require more than mere optimization of polymeric membrane materials. Nanotechnology has enabled researchers to create size-selective, well-defined, nanostructured filtration membranes by providing them control over fabrication of nano materials. Unlike polymeric membranes, which carry flexible chains and are unable to form well-defined pores, nano-membranes with size-selective pores at nano meter scale are expected to allow separation of water molecules, while simultaneously preventing the salt ions with larger diameter due to hydration shells [16,17]. For example, hydrated sodium ion is 76 nm (diameter) which can be removed by sieving through a membrane having pores smaller than 76 nm and bigger than water molecule. Because tremendous amount of energy is required to remove ion from its solvation shell, 1,709 kJ/mol in case of sodium ion, therefore ions can be impeded by applying pressure little higher than osmotic pressure on the feed water [18,19]. Also, steric exclusion and van der Waals electrostatic interactions may contribute toward achieving desired selectivity. So far, CNTs, graphene, and zeolites are fabricated into macroscopic arrays to carry out desalination [20–24]. The following sections will describe the progress in understanding the application of nano materials to get optimum performance of RO membranes for more efficient desalination systems.

3.1. Carbon nano tubes

CNTs are rolled-up graphene sheets consisting of carbon atoms [25] and exhibit exceptional mechanical strength, high electrical and thermal conductance, and unique electronic properties. These properties make CNT an attractive candidate for large range of applications including, but not limited to, structural materials, energy storage devices, adsorbents, semiconductors, and other electronics [14,26]. CNTs of diameter down to 1.6 nm can be prepared successfully by applying recently developed synthetic procedures [27]. This control over synthesis opened a gateway to the CNT membranes which can be applied in RO systems [22]. Molecular dynamics experiments and models suggest that water flux through CNTs can be substantially higher than approximated continuum hydrodynamics [28]. Also, molecular dynamics simulations provide mechanistic understandings of this rapid water transport. Firstly, it was discovered that an ice shell with water chain structure is formed due to confinement of water molecules in CNTs exhibiting stronger interactions between water molecules compared to those between water molecules and walls of CNT [29]. Later, other research revealed that not only hydrophobicity but also atomistic smoothness is also required for rapid transport of water through CNTs [30]. Recently, curvature of CNT is held responsible for friction coefficient between CNT wall and water molecules [31] as represented in Fig. 3. However, molecular dynamics simulations revealed that interaction energy landscape of CNT with water molecules is modulated by its curvature; thereby, decreasing CNT diameter decreases friction leading to its total extinction at 0.5 nm. Also, simulations indicated that outer surfaces of CNTs are more prone to water friction compared to inner surfaces.

There are some experimental verifications for the 4–5 orders increase of liquid flux upon applying CNTs. An interesting feature of this increased flux is

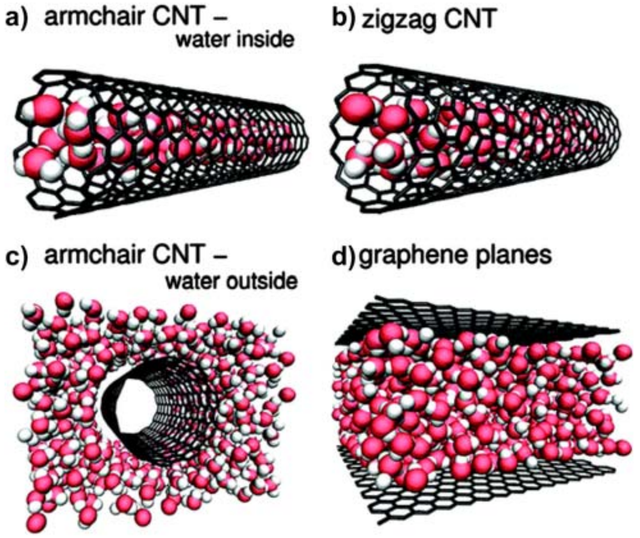

Fig. 3. Flow of water through CNT: (a) armchair, (b) zigzag, (c) outside armchair, and (d) between graphene sheets [31].

its independence of fluid viscosity [32]. This study was performed on water, ethanol, hexane, decane, and isopropane. These solvents were passed through 7 nm CNTs (under 1 bar pressure) and continuum hydrodynamics was applied to estimate flux using vertically aligned CNTs with polystyrene. The similar results were obtained when 1.6 nm CNTs were used [23]. But, in these studies, the diameter of CNTs used was large enough to sieve salt ions. Therefore, high salt rejection, due to electrostatic repulsion, was achieved by using membranes which carry chemically modified CNTs. A salt rejection of 40–60% (KCl) and up to 100% ($K_3Fe(CN)_6$) was achieved by using carboxylic groups functionalized CNTs [22]. Comparatively higher rejection of $K_3Fe(CN)_6$ can be attributed to higher electrostatic repulsion between carboxylic group and trivalent anion. However, this salt rejection decreased upon increasing salt concentrations and reached almost zero at 10 mM concentration of KCl, probably, due to smaller electrostatic Debye screening length. Fig. 4 describes the rejection of ions reported by Fornasiero et al. [22] when six different salts, having similar equivalent concentration, were attempted to pass through same charged CNTs.

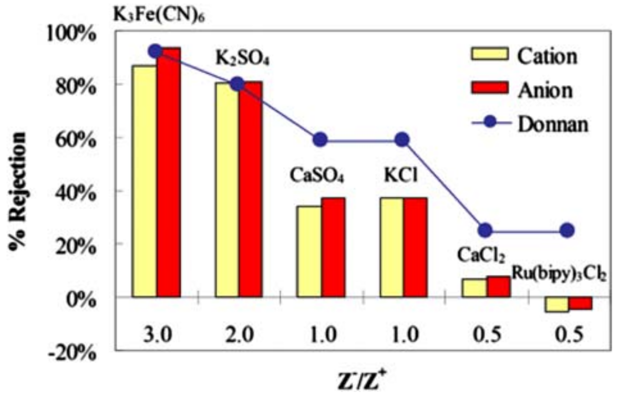

Fig. 4. Rejection of six salt solutions that have the same equivalent concentration but different ion valences by charged CNTs [22].

3.2. Graphene membranes

Graphene is an atomistic layer of graphite which consists of lattice of sp^2-bonded atoms arranged hexagonally [33]. It is not only attractive for electronic applications [33] due to its unique electronic properties, but also shows high breaking strength [34] and impermeability to small molecules including helium [35]. These characteristics of graphene enable it to construct extremely thin membrane with size tunable pores (for molecular sieving) allowing high flux. Also, it can be fabricated on large scale because there is an evidence of synthesizing 30 inch multilayer graphene sheets that have been transferred on roll-to-roll basis [36]. Researchers have recently studied the change in electrical properties of graphene by inducing defects [37]. Their experimental and simulation outcomes suggest that sub-nanometer pores can be generated and controlled by electron ion beam, oxidation, ion bombardment, or by doping [38–41]. It paved a way to explore the transport of molecules like gasses and ions through the pores in graphene membranes [42].

Again, molecular dynamics simulations helped in studying the transport of ions through 0.5 nm pores in graphene [43]. The graphene used in this study was terminated by either hydrogen or nitrogen. It was observed that pore in a graphene layer terminated by nitrogen allowed lithium, sodium, and potassium ions, whereas those terminated by hydrogen allowed chloride and bromide ions but did not let fluoride ion to pass. Unexpectedly, smaller ions showed lower passage rates when compared to larger ions. This might be due to strongly bound hydration shells of smaller ions. Also, like protein ion channels, charged terminal groups of pores helped water molecules to move out of hydration layer [43]. In another research, a comparison is made between water molecules transport through CNTs having 0.75–2.75 nm diameter and 2–10 nm length against a graphene membrane of similar diameter pores [42] as represented in Fig. 5. The observed flux through graphene was almost double to that of CNTs. The entrance regions of pores were the major contributor toward resistance. These studies show that graphene membranes might be better than polymeric RO membranes for water desalination in

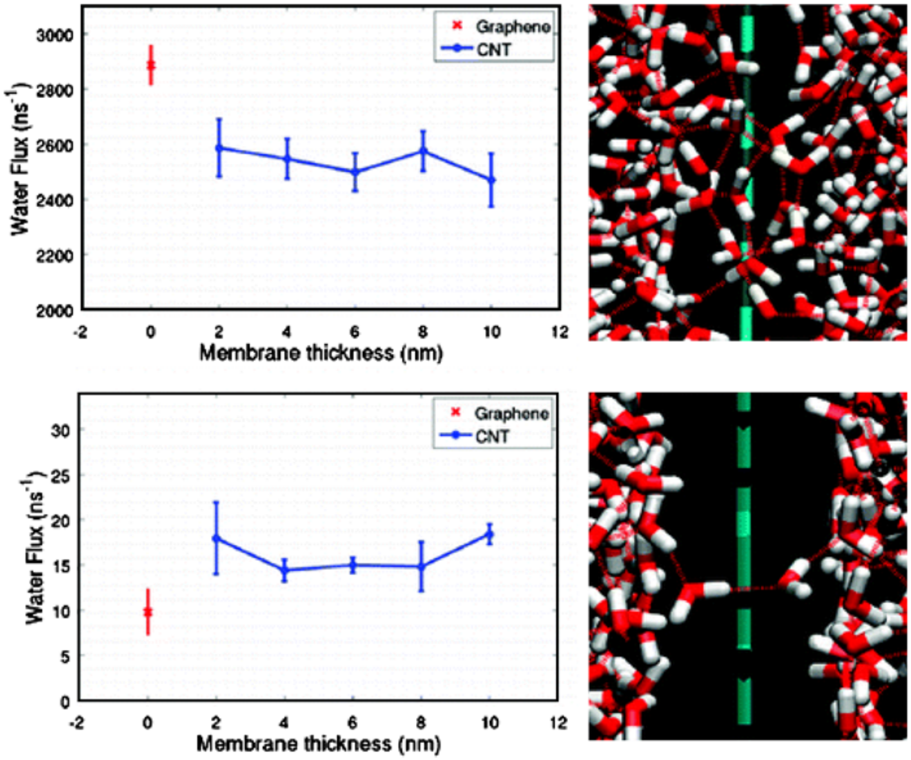

Fig. 5. Effect of membrane thickness on water transport across graphene sheet [42].

terms of allowable flux. However, there is a need of experimental measurements of salt rejection and water transport to verify these expectations [44].

4. Capacitive de-ionization

CDI is getting increased attention by researchers due to the availability of electrodes with high surface area for energy storage [45]. In CDI, the concentration of salts in feed water is reduced by applying voltage to the electrodes which causes ions to migrate and adsorb onto these electrodes as represented in Fig. 6. The desalinated water is then removed. Finally, the ions are desorbed by discharging electrodes. CDI may have more than one stages in order to achieve desired salt concentration in treated water [45]. During this process of adsorption onto electrodes, some of the electrical energy is stored due to the capacitance of electric double layer. When ions are released from the electric double layer, fraction of this energy is reacquired for further adsorption. This technique is especially useful for low salt concentration feed water like brackish water. Although CDI can work for high salinity water, yet, it becomes uneconomical because the capacitance of the electrodes limits the ion adsorption [46].

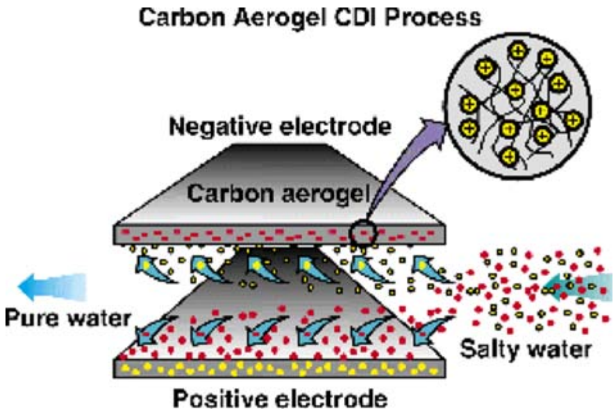

Fig. 6. Capacitive de-ionization process [47].

CDI can be an economical desalination technique if the following challenges are overcome: (1) insufficient electrical connectivity to the high surface area electrodes, (2) limitations on electrode materials, (3) geometries of electrodes leading to minimize the distance for ionic electro-migration, maximizing surface area, and (4) high energy consumption. Beside seeming simple process, the ion transport locality inside electrodes (in CDI) is yet to be fully understood. The classical electro-kinetic theory fails to explain CDI because of the application of high surface potential, complicated geometries of electrodes, long charging times, and small confined pores. However, some recent studies modified Poisson–Boltzman theory for the electric double layer (by taking steric effects of the ions into account) which lead to the better understanding of charge build-up in the electric double layer [8,48–51]. Also, some models are developed which couple electro-kinetic theory with ion diffusion, thereby, addressing dynamics of electrode charging [52,53]. But still, the development of high surface area electrode carrying good electrical conductivity with ability to control Faradaic reactions is a big challenge. Researchers have attempted to develop novel materials to increase the surface area and electrosorption of ions. These materials include activated carbon, activated carbon cloth, carbon aerogels, and CNTs and nano-fibers. A brief description of these materials is provided below.

The most studied of all the electrode material for CDI applications is activated carbon. Activated carbon is a highly porous form of carbon. It carries a very high surface area (~500–2,300 m^2/g). It is usually derived by the activation of pyrolysis of wood, coconut shell, and other carbonaceous materials [54]. Due to its large surface area, it is used in super capacitors and CDI [55–58]. A big problem in applying activated carbon for desalination is its hydrophobicity. The hydrophobic electrodes made from activated carbon require hydrophobic binder (like PTFE, polytetrafluroethylene) to maintain material consistency. Because, enhanced wettability of electrodes is required in CDI, therefore, efforts are made to decrease the hydrophobicity of activated carbon electrodes. One such effort was made by Lee et al. who increased the hydrophilicity of standard activated carbon/PTFE electrode by adding the ion-exchange resin. Addition of resin (24%) leads to the decrease of contact angle of electrode from 115 to 75°, resultantly, 35% increase in the desalination water production was achieved [58].

Along with activated carbon, activated carbon cloths also carry the high surface area (~1,500–2,500 m^2/g) but lack the PTFE binder. They are made up of fibers which are derived from phenolic resins [59,60]. So far, attempts are made to enhance CDI function by increasing electrosorption capacity of carbon cloths [61–66]. The electrosorption capacity of carbon cloths depends upon electrical connectivity, pore structure, surface area, and surface chemistry [61–66]. In one of such studies, three times increase in electrosorption capability of activated carbon cloth was observed by incorporating titania [62]. Another study depicts the successful increase in the efficiency of activated carbon cloth by surface modification through acid etching. The solutions of potassium hydroxide and nitric acid were used for this purpose, which increased the adsorption by increasing carbonyl, hydroxyl, and carboxyl functional groups on the surface of carbon [61]. For the same conditions of conductivity and electrical potential applied, nitric acid treated activated carbon cloth removed 16% and the one treated by potassium hydroxide removed 14% more salt compared to untreated ones. Researchers credited enhanced double layer electrosorption and Faradic reactions for this increased salt removal, which came into play due to the surface carboxyl groups added by nitric acid.

Unlike activated carbon and carbon cloths, carbon aerogels are prepared by sol–gel process. They are highly cross-linked networks of carbon particles which are covalently bonded together. The electrodes made from them can have a surface area of 400–1,200 m^2/g, pore size less than 100 nm, and porosities more than 50% [67–69]. One of the study reports the removal of 95% of NaCl (100 μS/cm) using these electrodes [70]. Besides large surface area, most of the pores in aerogels are less than 2 nm (micropores) and are, therefore, inaccessible to ions [71]. Due to the small pore size of these aerogels, the focus of researchers moved to new materials which carry larger pores. The newly developed CNMs especially CNTs and nanofibres, and ordered mesoporous carbons, can be potential candidates to enhance ion capture in CDI.

CNTs and nanofibre composites, fabricated by chemical vapor deposition, have shown effective ion capture [21,71–73]. CNTs and nanofiber composite electrode enhanced electrosorption, compared to standard activated carbon and activated carbon cloth electrodes, due to the presence of higher number of macropores and mesopores [72]. Macropores and mesopores are more suitable for CDI electrodes because they are not significantly affected by electric double layer unlike micropores which further decrease in diameter due to electric double layer. However, the specific surface of CNTs and nanofibre electrodes is found to be smaller than that of activated carbon.

Studies regarding CNT and nano-fiber composite electrodes revealed the significance of microstructure of electrode material for electro-sorption. Ordered mesoporous carbons can be synthesized by impregnating mesoporous silica (or similar materials) with carbon and then removing this base material. They carry pores ranging in diameter from 3 to 25 nm. These micropores were expected to have lower electrosorptive capacity; however, it was found that contrary is true [74]. Ordered mesoporous carbon having surface area 1,000–1,500 m^2/g exhibited increased ion capture even for a smaller capacitance [74]. Although the mechanism for this strange behavior is not fully understood; yet, it is thought to happen due to orderliness of material which aided the transport of ions. These observations may play a critical role in the design of future materials because it challenges the standard electrochemical testing techniques applied to obtain capacitance measurements of materials.

5. Membrane distillation

MD is another merging technique to desalinate saline water. It is a thermally driven process which is based on the principle of development of vapor pressure gradient across a hydrophobic membrane [75,76].

Direct contact membrane distillation (DCMD), Fig. 7, is the most popular configuration of MD. In DCMD, a barrier in the form of hydrophobic membrane separates cold freshwater and hot seawater or brackish water [78]. The driving force in DCMD is water vapors which permeate through the membrane, leaving behind liquid, hence creating a vapor partial pressure. It is observed that the temperature difference between the feed and permeate is responsible for flux through the membrane [79]. DCMD is rapidly getting attention of researchers around the globe due to its low energy consumption for water purification when compared to traditional technologies like RO and nanofiltration. However, there are few challenges in the way of commercializing the technology: (1) simultaneous increase in porosity and decrease in thickness of the membrane is required to achieve high vapor transport from hot side to the cold side [80,81] and (2) increase in the diameter of pores of the DCMD membrane is needed to suffice vapor transport while minimizing membrane hydrophilicity, so that liquid bridge between permeate and feed can be avoided [76,79–81]. So far, hydrophobic membranes such as poly vinyledene fluoride (PVDF) and polypropylene (PP) are studied for this purpose [82]; however, they are found difficult to process owing to their inherent ability of avoiding water. Hence, a need for the development of new membranes, to improve the process of MD, paved a way to explore newly developed CNMs especially CNTs.

The CNTs exhibit interesting properties to be used as a membrane material for MD. One of its highly studied applications is as a bucky paper [78,83–88]. Bucky paper is a paper-like structure of CNTs which is applied for MD after processing by vacuum filtration [83]. Recently, a successful immobilization of CNTs is carried out in polymer matrix as represented in Fig. 8. This new approach will assist water vapors to pass through CNTs while maintaining the resistance for water to flow through due to their inherent hydrophobicity. This results in enhanced adsorption and desorption rates [89].

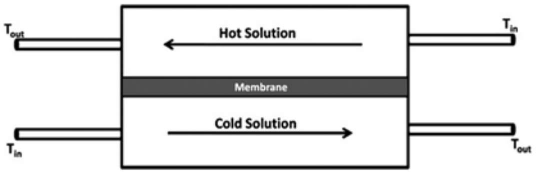

Fig. 7. Schematics of direct contact MD process [77].

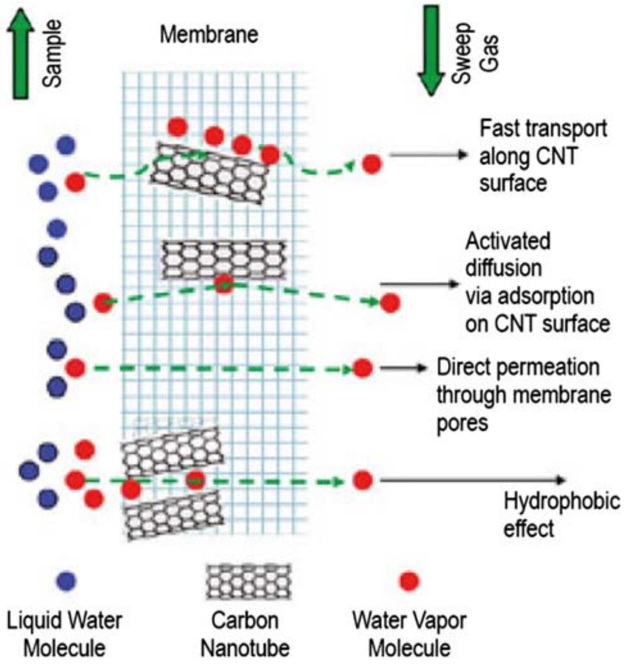

Fig. 8. MD mechanisms in the presence of CNTs [89].

6. Forward osmosis

CNTs are also a possible candidate for FO. The transport phenomena of water molecules and ions through the membranes incorporating the neutral "armchair" (R,R) CNTs with indices R = 6–11 in FO process are studied by means of the molecular dynamic simulation [90]. It was found that CNTs are very suitable for FO because CNT membrane is able to reject optimum salt while maintaining high flux as against the conventional membrane separation processes where trade-off between permeability and selectivity is encountered. It is, therefore, expected that CNTs will play a significant role in future FO membranes due to their unique mechanical properties, antifouling ability, and other characteristics along with high flux and high selectivity as documented [90].

7. Concluding remarks

The increasing demand for clean water, decrease of natural resources of fresh water, and the rise in energy costs are the main drivers for research in improvement of existing desalination technologies. The future desalinating technologies will require nano materials to enhance water permeation flux and reduces specific energy consumption as the existing desalination technologies reach maturation. The nanofabrication techniques provide opportunities to understand and develop nanomaterials with extraordinary structural and functional characteristics. This review emphasizes on the application of such carbon-based nanomaterials with a potential to impact future desalination technologies.

Although recent researches in energy recovery for RO membrane systems have significantly improved the performance of such systems, yet, newly developed CNMs carry a potential to further improve RO systems by contributing toward development of rigid and size selective membranes with higher permeability. However, deeper understanding of transport processes, optimization of pore sizes, and reduction in capital and operating costs of these CNMs containing membranes is required which needs more research in this area.

Other emerging desalination technology is CDI. The efficiency of CDI is primarily based on design and structure of electrodes. Therefore, the development of electrodes with enhanced surface area, controlled Faradic reactions, and improved conductivity has lead researchers to explore activated carbons, carbon aerogels, activated carbon cloths, and mesoporous carbons. Most recently, the carbon nanofibers and CNTs are giving encouraging results in this field. Similarly, MD (a phase change desalination process) requires stringent control over evaporation, condensation, and wetting to improve transportation of vapors. The emerging CNMs are assisting scientists and engineers in getting more hold on these processes. In case of FO, CNTs are expected to enhance flux without compromising on selectivity. Hence, it can be concluded that the development of CNMs and fundamental understanding of transport phenomena at nanoscale carry a big potential in contributing toward fulfilling the increased demand of clean water for future generations through affordable, efficient, and sustainable techniques.

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