Chapter 4
Developments of Carbon-Based Membrane Materials for Water Treatment

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Contents

Abstract Serious water contamination and freshwater shortage result in the urgent requirements of advanced technologies for water treatment. Membrane separation is an alternative technology to address the global water crisis. Hence the research for membrane materials with excellent properties is being undertaken vigorously. Recently, successful attempts have been made towards applying carbon-based membrane materials, such as carbon membranes, carbon nanotube membranes,

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carbon fiber membranes, activated carbon membranes, graphene-based membranes, etc. for achieving a high separation performance. The intrinsic properties of the carbon materials can potentially lead to enhancements in fouling mitigation, hydrophilicity, and permeate quality. This chapter provides a brief and comprehensive overview of the fabrication and synthesis mechanisms of the carbon-based membrane materials, characterization methods, and practical applications in water treatment. The major points are:

1. Carbon membranes, derived from phenolic resin and coal as precursors, have been widely used in water treatment, specifically utilizing the electrical conductivity of coal-based carbon membrane as the electrode and membrane filter simultaneously demonstrate great potential on water treatment.
2. Four types of carbon nanotube membranes are presented and indicate high separation performance due to the remarkable physicochemical properties of carbon nanotubes.
3. Carbon fiber membranes possess abundant functional groups on the surface, favoring high permeability in water treatment.
4. Activated carbon membranes are promising for organic matter removal owing to high surface area, micro–meso and macroscopic structure, and various chemical functional groups.
5. Graphene-based membranes as the novel carbon-based membrane materials with unique laminar pores are attracting more and more attentions.

Keywords Membrane · Carbon materials · Wastewater treatment · Water purification · Separation

4.1 Introduction

The industrial development and population growth have led to serious and sustainable challenge towards the water resources in the twenty-first century (Menachem and William 2011; Ma et al. 2017; Salgot and Folch 2018). The prediction from the United Nations indicates that half of the countries worldwide will be confronted with water shortage in the coming decades (Goh and Ismail 2018). The World Health Organization (WHO) also estimates that more than 1.2 billion people worldwide have gotten sick or died through drinking contaminated water, and the number is expected to significantly grow in the coming years (Montgomery and Elimelech 2007; Wilson et al. 2018). Hence, in order to reduce the hazards from water pollution to humankind, various technologies and industrial processes for water treatment or purification have been developed and applied rapidly in recent years (Zheng et al. 2015; Pintor et al. 2016; Hayat et al. 2017; Jiao et al. 2017).

Among them, membrane separation has been accepted as a promising and pervasive technology arising from its numerous advantages of no chemical additives requirement, low energy demand, easy operation, high separation selectivity, and good stability (Gin and Noble 2011; Li et al. 2016b; Thakur and Voicu 2016; Chowdhury et al. 2018; Lau et al. 2018). To date, membrane separation has been widely applied in industrial wastewater treatment and drinking water purification and desalinization (Pendergast and Hoek 2011; Singh and Hankins 2016; Parimal 2017; Zhang et al. 2018). As one of the dominated factors to determine membrane performance, membrane materials should be primarily concerned for exploring high-performance membranes.

Recently, carbon-based materials have been used to develop membranes with optimal structure and performance due to their excellent physicochemical properties (Goh et al. 2016; Thines et al. 2017; Anand et al. 2018; Wei et al. 2018). The carbon-based materials not only can improve the wetting ability and surface charges of the membranes but also introduce additional functions such as antimicrobial ability and photocatalytic and electrochemical reactions (Liu et al. 2011; Ong et al. 2018). According to previous works, several kinds of carbon-based membrane materials including carbon membranes, carbon nanotube membranes, carbon fiber membranes, activated carbon membranes, graphene-based membranes, etc. (Inagaki et al. 2014; Jiang et al. 2016; Lawler 2016; Vatanpour and Safarpour 2018) are described. This chapter aims to provide an overview on recent developments of carbon-based membrane materials for water treatment. A brief discussion of the existing challenges and their prospects are also considered.

4.2 Carbon Membranes

Carbon membranes, as novel porous inorganic membranes, are usually prepared by pyrolysis of carbonaceous materials, such as polyimide and its derivatives, polyacrylonitrile, poly(furfuryl alcohol), phenol–formaldehyde, coal, etc. In the past several decades, carbon membranes have demonstrated excellent gas separation performance (Hamm et al. 2017), however, only a few carbon membranes are applied on water treatment due to their high cost and complex preparation process. In the following parts, several kinds of carbon membranes used in water treatment will be introduced.

4.2.1 Phenolic Resin-Based Carbon Membranes

Phenolic resins have presented suitable features to be applied as the precursors of carbon membranes due to their low cost, thermosetting property, and high carbon yield (Muylaert et al. 2012). Several scholars have successfully prepared carbon membranes with phenolic resins for water treatment. Song et al. (2017) developed

carbon alumina mixed-matrix membranes by impregnating phenolic resin in porous alumina matrix via a vacuum-assisted method. Their results showed that carbon alumina mixed-matrix membranes with high water fluxes and salt rejections could be easily tailored. However, the carbon membrane, formed by dip coating a phenolic resin solution on an alumina substrate, could not exclude small molecules of glucose and sucrose. It only demonstrated high removal rates (80% and 100%, respectively) for 36 kda and 400 kda of polyvinylpyrrolidone polymers (Abd et al. 2017). Wu et al. (2016) prepared phenolic resin-based carbon membrane to treat oily wastewater. The oil concentration dramatically reduced from initial 200 mg/L in feed to below 10 mg/L in permeate, with the oil rejection rate of 95.3%. Zhao et al. (2018) prepared the original precursor membrane by compressing the mushy mixture composed of phenolic resin, hexamethylenetetramine, carboxymethylcellulose sodium, and distilled water. The results showed that these carbon membranes could effectively remove phenol and phosphoric acid from water. The maximum removal rates were 81.9% for phenol and 55.3% for phosphoric acid. In addition, the carbon membrane derived from phenolic resin was also effective to treat dye wastewater. Asymmetric tubular carbon membranes on an ultrafiltration substrate were prepared by thermosetting phenolic resin and carbon black (Tahri et al. 2016), and such carbon membranes could be applied efficiently to the treatment of industrial dyeing effluent. According to the above research, carbon membranes made from phenolic resin as raw material or part of raw material have been applied in many aspects of water treatment and showed their unique performance.

4.2.2 Coal-Based Carbon Membranes

Coal, as a kind of natural mixture composed of macromolecular cross-linked polymers and inorganic minerals, is a good candidate for preparing carbon membranes because of its low price and abundant deposit. In the past two decades, our group explored the preparation technology of carbon membranes derived from coal, which was shown in Fig. 4.1. The coal was ground into fine particles first, and then mixed with binder into a dough, which was extruded into a tube of 10 mm external diameter by a hydraulic extruder at 2.5–3.0 MPa. After drying at ambient atmosphere, the tubular membrane was carbonized in Ar up to 900 °C at the rate of 3 °C/min and held for 1 h. The final product was cooled to room temperature naturally. A series of systematic investigations on the controlled preparation of coal-based carbon membranes were carried out, and the pore structure, mechanical strength, and electrical conductivity of CBCMs were further optimized. As expected, the coal-based carbon membranes showed excellent water treatment performance (Song et al. 2006).

During treatment, the retention and accumulation of pollutants on the membrane surface and inside the membrane pores would give rise to serious membrane fouling. In order to improve the antifouling ability of coal-based carbon membranes, an electric field was exerted on the treatment system; our group utilized the electrical conductivity of coal-based carbon membranes and designed a coupling system

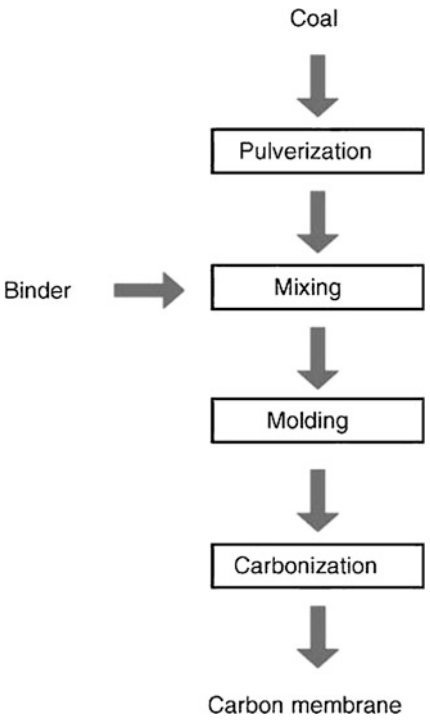

Fig. 4.1 Preparation process of coal-based carbon membranes. (Reprinted with permission of (Song et al. 2006))

which employs coal-based carbon membranes as the anode and Ti plate surrounding the membrane as the cathode. This system achieved significant improvement on removal efficiency and antifouling ability under an external electric field due to the electrochemical oxidation (Fig. 4.2). This system not only displayed excellent removal efficiency for organic pollutants (such as oil droplets) larger than the membrane pores (Li et al. 2016a) but also demonstrated great potential on those pollutants with a smaller molecule size than the membrane pore size including dyes, phenol, etc. (Yin et al. 2016); Tao et al. 2017b; Sun et al. 2018). Moreover, microorganisms such as microalgae and *Vibrio cholerae* were also effectively removed (Tao et al. 2017a). Compared with other membrane processes such as ultrafiltration, nanofiltration, and reverse osmosis, this technology possessed obvious advantages on processing capacity and energy consumption.

Although the coupling system has been proved to be effective for organic wastewater treatment, further potential for improvement in the removal efficiency and life span of the coupling system is often limited by the relatively low electrochemical activity of membrane electrode materials. Therefore, improving electrochemical activity of the membrane electrode material is a key to make a significant breakthrough in this field. Yang et al. (2011) presented the design of a novel electrocatalytic membrane reactor by loading electrocatalyst on carbon membrane (Fig. 4.3). In the research, TiO_2 as the electrocatalyst and hydrophilic agent was coated on the membrane surface by a sol–gel approach to enhance electron transfer

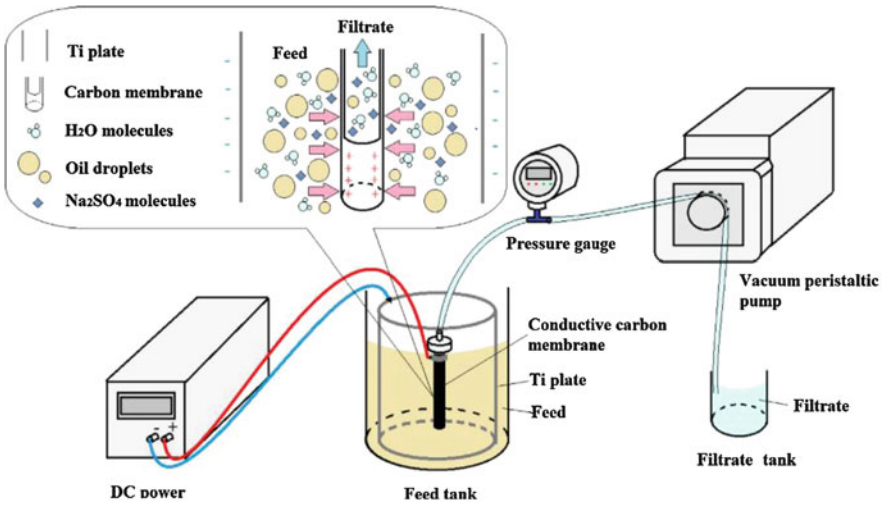

Fig. 4.2 Flow schematic diagram of carbonized membrane coupling with an electric field. (Reprinted with permission of (Li et al. 2016a))

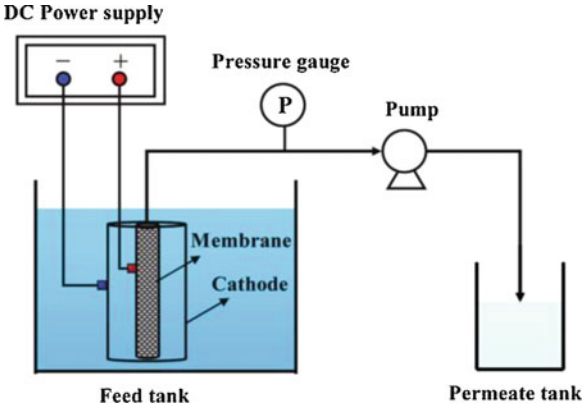

Fig. 4.3 Scheme of electrocatalytic membrane reactor. The figure shows an electrocatalytic membrane reactor with self-cleaning function for industrial water treatment. (Reprinted with permission of (Yang et al. 2011))

and improve membrane permeability. In this operation process, once the membrane anode was electrified, excitation of electrons in the conduction band took place at the TiO_2 surface. The obtained electrons and holes not only electrochemically decomposed H_2O into O_2 and H_2, inducing gas and liquid microflows to reduce concentration polarization and avoid membrane fouling, but also reacted with the adsorbed H_2O and O_2 at the TiO_2 surface to generate reactive intermediates, which could indirectly decompose the organic foulants into CO_2 and H_2O or biodegradable products, so as to realize the self-cleaning function of the electrocatalytic membrane. Similarly, Wang et al. (2014) also used an electrocatalytic membrane reactor constituted by TiO_2 loading carbon membrane to treat phenol wastewater. Besides, the Bi–SnO_2/C electrocatalytic membrane was fabricated via a simple electrochemical

reduction and hydrothermal method by Wang et al. (2018b). The Bi–SnO_2/C membrane could continuously remove and inactivate *E. coli* in water through flow-through mode. As a result, the sterilization efficiency reached more than 99.99% under the conditions of cell voltage of 4 V, flow rate of 1.4 mL/min, and *E. coli* initial concentration of 1.0×10^4 CFU/mL, owing to the synergistic effect of the membrane separation and electrocatalytic oxidation.

4.3 Carbon Nanotube Membranes

Carbon nanotubes (CNTs), as an important kind of carbon materials, have many remarkable electrical, thermal, mechanical, and optical properties, which make them be widely used in sensor, supercapacitor, lithium–ion battery, etc. (Ren et al. 2011; Gupta et al. 2013; Yu et al. 2014; Apul and Karanfil 2015; Patino et al. 2015). Generally, carbon nanotubes can be divided into single-walled carbon nanotubes and multi-walled carbon nanotubes (Fig. 4.4) (Ahn et al. 2012; Ihsanullah 2019). As we have known, carbon nanotubes were firstly discovered by Sumio Iijima (1991). Soon after, researchers observed ultrahigh water flow rates in carbon nanotubes, and this discovery produced great expectation that carbon nanotubes could be used as an ideal material for water treatment (Whitby and Quirk 2007; Lee et al. 2011; Ahn et al. 2012).

The concept of carbon nanotube membrane was introduced by Li and Richard (2000) when they studied the mass transfer phenomenon in single-walled carbon nanotubes. Recently, carbon nanotube membranes for water purification are getting more and more attention. According to the arrangement patterns of carbon nanotubes, carbon nanotube membranes are usually classified into vertically aligned carbon nanotubes (VA-CNT) membranes, horizontally aligned carbon nanotubes (HA-CNT) membranes, mixed-matrix carbon nanotube membranes, and electrochemical carbon nanotube membranes (as shown in Fig. 4.5).

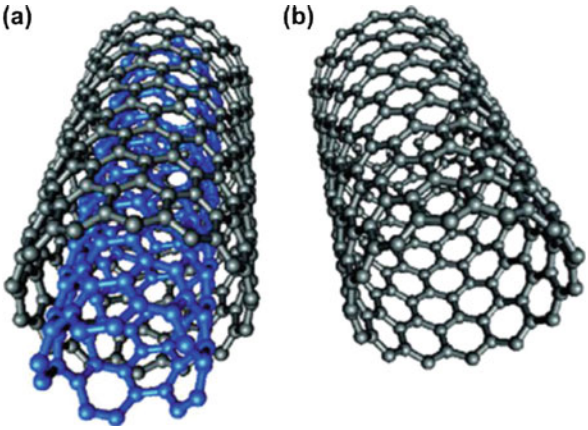

Fig. 4.4 The structure of multi-walled carbon nanotubes and single-walled carbon nanotubes. (Reprinted with permission of (Ihsanullah 2019))

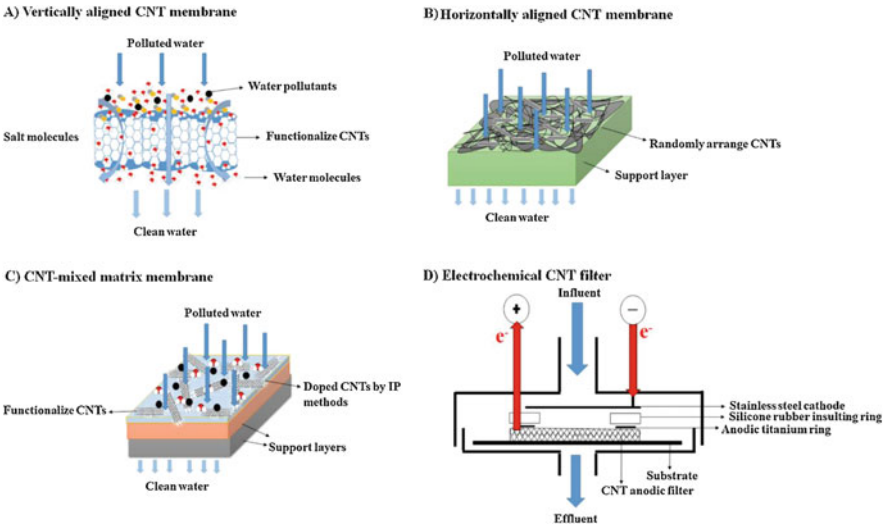

Fig. 4.5 Mechanism of water passing through the four types of carbon nanotube membranes: (**a**) vertically aligned carbon nanotube membrane, (**b**) horizontally aligned carbon nanotube membrane which is randomly arranged horizontally on a porous support layer, (**c**) mixed-matrix carbon nanotube membrane which is directly doped into the polymer membranes by interfacial polymerization or phase inversion, (**d**) electrochemical carbon nanotube membrane. (Reprinted with permission of (Ali et al. 2019))

4.3.1 Vertically Aligned Carbon Nanotube Membranes

Bruce et al. (2004) firstly constructed a multi-walled vertically aligned carbon nanotube membrane, and its typical preparation process was shown in Fig. 4.6 (Das et al. 2014), and the separation performance of vertically aligned carbon nanotube membranes was listed in Table 4.1. The work from Baek et al. (2014) showed the superiority of vertically aligned carbon nanotube membrane with the water permeation almost three times higher than a typical ultrafiltration membrane. Besides, the membrane prepared by Holt (2004) with silicon nitride (Si_3N_4)-filled carbon nanotube array obtained much higher water flux which was three times larger than that calculated by the Hagen–Poiseuille equation. This was mainly owing to the effect of the compact nanotube forest and short nanochannel length. In addition, some researchers prepared novel vertically aligned carbon nanotube membranes that possessed certain antimicrobial and antifouling capacities (Lee et al. 2015). A key challenge on preparing these kinds of membranes was to align the carbon nanotubes over a sufficiently large area for comprehensive water treatment (Ali et al. 2019). Instead of conventional preparation methods, Wu et al. (2014) utilized an electric field to obtain vertically aligned carbon nanotube membranes. Electro-casting allowed multi-walled carbon nanotubes to grow vertically and disperse more evenly. However, complex manufacturing techniques were still major obstacle to make these membranes suitable for large-scale applications (Ihsanullah 2019).

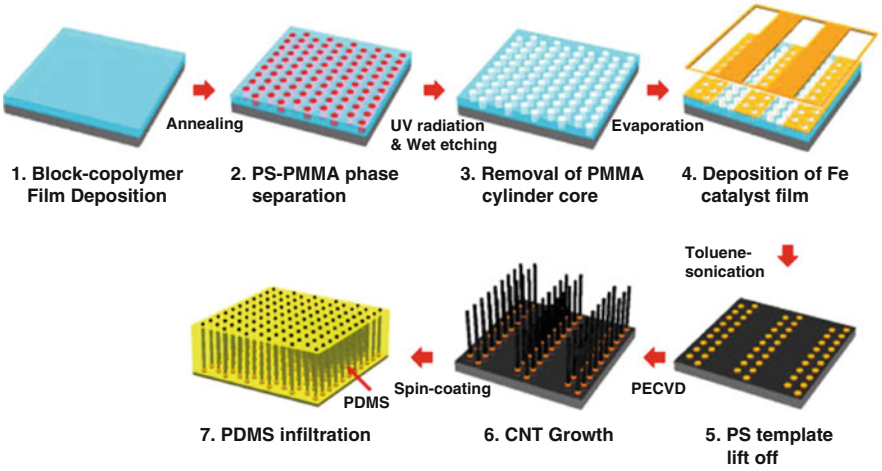

Fig. 4.6 Process flow for the fabrication of a vertically aligned carbon nanotube membrane using a block copolymer lithography method. (Reprinted with permission of (Ahn et al. 2012))

4.3.2 Horizontally Aligned Carbon Nanotube Membranes

In addition to vertically aligned pattern, carbon nanotubes can aggregate with each other by the van der Waals interactions to form horizontally aligned carbon nanotube membranes (Fig. 4.5B) (Ihsanullah 2019). This type of carbon nanotube membranes possesses several advantages such as a high specific surface area, large porous 3D network, etc. The most common methods for synthesizing horizontally aligned carbon nanotube membranes are electrospinning, vacuum filtration, and layer-by-layer deposition (Sears et al. 2010).

The preparation processes of horizontally aligned carbon nanotube membranes usually involve two steps: the functionalization of carbon nanotubes and vacuum filtration (Fig. 4.7). Firstly, the functionalized carbon nanotubes (horizontally aligned carbon nanotubes) are ultrasonically treated for uniformly dispersing in water or other solvents. Then, the dispersion is placed on the substrate membrane by vacuum filtration, after drying in an oven to remove the solvent (Lee et al. 2016a).

The related works on horizontally aligned carbon nanotube membranes are listed in Table 4.2. Due to the disordered arrangement of functionalized carbon nanotubes, the horizontally aligned carbon nanotube membranes can provide rich porous structure and large specific surface area (Sears et al. 2010), which makes the horizontally aligned carbon nanotube membranes possess high adsorption capacity to natural organic matter (Yang et al. 2013) and strong antimicrobial actions (Kang et al. 2007). Li et al. (2015) found that a "slanted carbon nanotube membrane" exhibited a higher water flux than a typical vertically aligned carbon nanotube membrane, because this kind of art structure could obviously lower the energy barrier for filling water into the carbon nanotubes. Brady Estevez et al. (2008) reported that the horizontally aligned single-walled carbon nanotube membrane

Table 4.1 Membrane performance of some vertically aligned carbon nanotube membranes

Membrane material	Membrane performance	Reference
CNT/polystyrene	The membrane flux of ruthenium bipyridine and methyl viologen was 9.57 (±0.91) and 21.05 (±2.32) nmol/h, respectively	Mainak et al. (2005)
CNT/stainless steel	The flux of diesel and water was 4692 kg/(m^2·h) (400 Pa) and 85.6 kg/(m^2·h) (1820 Pa) when the membrane was used to separate diesel–water mixture	Lee and Baik (2010)
CNT/polyethersulfone	The water flux was ~100 L/(m^2·h) at 60 Psi	Li et al. (2014)
CNT/PS/epoxy resin	The water flux was 1100 ± 130 L/m^2·h·bar (3 times higher than a commercial membrane). The VA-CNT membrane showed better biofouling resistance	Baek et al. (2014)
CNT/ polytetrafluoroethylene/ Si	The water flux was 30,000 L/m^2·h·bar (almost 12.5 times higher than the reported CNT membranes). The carbon nanotube walls of the membrane were proved to hinder the formation of biofilms and prevent bacterial adhesion	Lee et al. (2015)
CNT/Fe/Al_2O_3/Si	The BSA rejection increased from 71% to 90% with the modification of methacrylic acid. The pure water flux was 1000 ± 100 L/(m^2·h·bar)	Park et al. (2014)
CNT/Si wafer	The rejection rate of NaCl was 41.4%. The water flux was $1.31 \times 10^{-3} - 6.57 \times 10^{-2}$ L/(cm^2·day·MPa)	Matsumoto et al. (2017)
CNTs–TiO_2/Al_2O_3	The rejection rate of polyethylene glycol was 70% and the flux was 980 L/(m^2·h)	Zhao et al. (2013a)
Fe_3O_4/CNT	Membranes with a 10 and 1% iron oxide exhibited the best removal of 90 and 88% of SA after 3 h	Ihsanullah et al. (2016)
CNT–carbon fabrics	The hydrophobicity of the membrane increased; the wetted surface fraction and adhesion were lower. The separation efficiency of oil–water mixture was much higher	Hsieh et al. (2016)
PdO–CNT	The removal efficiency of atrazine was almost 100%	Vijwani et al. (2018)

displayed high removal rate for the virus MS_2 bacteriophage. Ihsanullah et al. (2015) synthesized a silver-doped carbon nanotube membrane and demonstrated good antibiofouling and antibacterial properties. Subsequently, they found that an iron oxide composite carbon nanotube membrane could present excellent antifouling property (Ihsanullah et al. 2016). Dumée et al. (2010) applied horizontally aligned carbon nanotube membranes to direct contact membrane distillation. Their work proved that horizontally aligned carbon nanotube membranes possessed high water flux and good desalination ability. After that, they modified high-purity carbon nanotubes by two chemical ways, and the resultant horizontally aligned carbon nanotube membrane had a larger contact angle (140° compared with 125°), which further improved the performance of the horizontally aligned carbon nanotube membrane (Dumée et al. 2011).

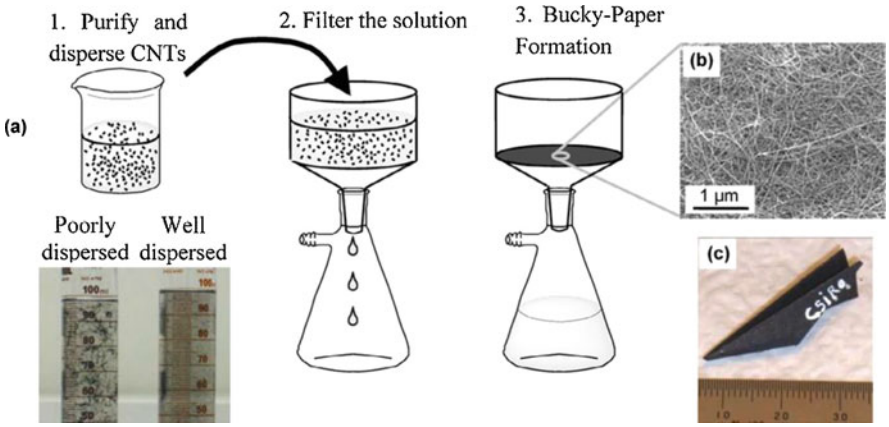

Fig. 4.7 Process flow for the fabrication of horizontally aligned carbon nanotube membrane. (**a**) Flow of manufacturing horizontally aligned carbon nanotube membrane. (**b**) SEM image of the membrane surface. (**c**) Fold it into a paper airplane to show its flexibility and mechanical robustness. (Reprinted with permission of (Sears et al. 2010))

Table 4.2 Application and membrane performance of some horizontally aligned carbon nanotube membranes

Membrane material	Membrane performance	Reference
CNT	The salt rejection was more than 95%. The water vapor flux was 4.5 ± 0.1 × 1012 kg/(m·s·Pa)	Dumée et al. (2011)
CNT	The salt rejection was more than 99%. Flux rate was ~12 kg/(m^2 h) at a water vapor partial pressure difference of 22.7 kPa	Dumée et al. (2010)
CNT/PP/PES/PS/PVDF	The salt rejection was 95%. The water vapor flux was $3.3\times10-^{12}$ kg/(m·s·Pa)	Dumée et al. (2012)
f-CNT	The rejection rate of humic acid was more than 93%	Yang et al. (2013)
CNT/PVDF	The rejection rate of *E. coli* was 94% (exhibited good antimicrobial capacity). The water flux was 13,800 L/m^2·h·bar and 6500 L/(m^2·h·bar) at SWNT loading of 0.3 mg/cm^2 and 0.8 mg/cm^2	Brady Estevez et al. (2008)
Cu–CNT/PVDF	The rejection rate of As(III) was above 90%. The pure water flux was 4639–4854 L/m^2·h·bar).	Luan et al. (2019)

However, carbon nanotubes usually tended to aggregate when they were dispersed in a polymer matrix or solvent. Therefore, it was difficult to prepare a uniform dispersion. For this reason, several surfactants such as Triton X-100, sodium lauryl sulfate, etc. were adopted to improve the dispersion of carbon nanotubes in aqueous solution (Wu et al. 2010c). Besides, another efficient method was chemical functionalization (Yang et al. 2013), which had been proved to increase the hydrophilicity and stability of carbon nanotube suspensions (Ansón-Casaos et al. 2010). For example, some researchers covalently grafted functional groups including

amines, fluorine, and sulfhydryl groups onto carbon nanotubes to help them disperse in horizontally aligned carbon nanotube membranes (Ansón-Casaos et al. 2010; Darryl et al. 2010).

4.3.3 Mixed-Matrix Carbon Nanotube Membranes

The main role of carbon nanotubes in mixed-matrix carbon nanotube membranes is to improve the performance of conventional polymer membrane (Ihsanullah 2019). Compared with the above two types of membranes, mixed-matrix membranes are easier to be commercialized for their simple preparation procedures. For preparing mixed-matrix carbon nanotube membranes, functional carbon nanotubes are generally added into polymeric membranes by several synthesis techniques (Ali et al. 2019; Ihsanullah 2019). The most common methods are phase inversion (Choi et al. 2006; Brunet et al. 2008; Majeed et al. 2012), interfacial polymerization (Shen et al. 2013; Kim et al. 2014), solution mixing (Ahmed et al. 2013), spray-assisted layer-by-layer (Liu et al. 2013), polymer grafting (Shawky et al. 2011), in situ polymerization (Zhao et al. 2014; Zarrabi et al. 2016), and in situ colloidal precipitation (Ho et al. 2017). The prepared membranes often exhibit excellent properties for reverse osmosis, ultrafiltration, and forward osmosis applications (Lee et al. 2016a). Some researches about the membrane performance of mixed-matrix nanotube membranes are listed in Table 4.3.

Mixed-matrix carbon nanotube membranes typically exhibited high removal efficiency and water flux. Zheng et al. (2017) prepared a novel sulfonated multi-walled carbon nanotube membrane by using the interfacial polymerization method. By adding 0.01% multi-walled carbon nanotubes, the membrane showed high salt rejection (96.8%) and water permeation (13.2 $L/(m^2 \cdot h \cdot bar)$). Moreover, a polysulfone membrane (Choi et al. 2006) and a polyether sulfone membrane (Celik et al. 2011b) doped with carbon nanotubes were more hydrophilic and demonstrated an enhanced antifouling ability because of the hydrophilic carboxylic groups of functionalized carbon nanotubes.

4.3.4 Electrochemical Carbon Nanotube Membranes

Electrochemical carbon nanotube membrane for wastewater treatment is a novel technique which combines electrochemical degradation with conventional membrane filtration to remove target contaminants (de Lannoy et al. 2012; Lalia et al. 2015; Ahmed et al. 2016; Elimelech and Boo 2017; Ho et al. 2018; Yi et al. 2018). In this process, the electrochemical carbon nanotube membranes are used both as a filter for contaminant sorption and an electrode for electrochemical degradation of aqueous pollutants (Ali et al. 2019).

Table 4.3 Application, membrane performance, and other conditions of mixed-matrix carbon nanotube membranes

Material	Synthesis technique	CNT amount (wt %)	Application	Membrane performance	Reference
MWNCTs/polysulfone (PSf)	Phase inversion	1.5	UF	The rejection rate of PEO was more than 99%. The flux was ~21 m^3/m^2·day at 4 bar	Choi et al. (2006)
MWCNTs/PSf	Phase inversion	4	UF	The flux increased from 24.6 ± 12.6 to 28 ± 10.7 L/(m^2·h) by the addition of 4 wt% CNTs	Brunet et al. (2008)
MWCNTs/PAN	Phase inversion	2	UF	The flux increased from ~41 to 53 L/(m^2·h) at 2 bar by the addition of 2 wt% CNTs	Majeed et al. (2012)
MWCNT/PSf (C/P)	Phase inversion	2	UF	The water flux was ~90 L/(m^2·h) at 60 Psi with the addition of 2% MWCNTs	Celik et al. (2011b)
MWCNT/PSf	Phase inversion	0–1	UF	The removal efficiency of Cr(VI)/Cr(II)was from 10.2% and 9.9% to 94.2% and 78.2%	Shah and Murthy (2013)
MWCNTs/PSf hollow fiber membrane	Phase inversion	0.1	UF	The flux increased from 36.1 ± 4.0 to 70.7 ± 1.8 L/(m^2·h) with the addition of 0.1% MWCNTs	Yin et al. (2013)
f-CNTs/PA	Interfacial polymerization	0–20	–	The flux increased by more than a factor of 4 with the addition of CNTs from 0 to 20%	Chan et al. (2013)
MWCNTs/PPSU	Phase inversion	0.5	UF	The pure water flux increased from 7.91 to 56.91 L/(m^2·h) at 345 kPa with addition of 0.5 wt % F-MWCNTs. The rejection rate of pepsin and trypsin decreased from 97 and 90% to ~90 and 84% with addition of 0.5 wt% f-MWCNTs.	Lawrence et al. (2012)
CNT/PA	Interfacial polymerization	0.2 g	RO	The rejection rate of NaCl was more than 95% (at 2000 ppm). The pure water flux increased from ~37 to 44 L/(m^2·h) at 15.5 bar of feed pressure with addition of 2 wt% MWCNTs. The water flux of PA-CNT membrane decreased by only 18.40%, compared with the PA membrane decreased by 32.80% after 48 h	Kim et al. (2014)

(continued)

Table 4.3 (continued)

Material	Synthesis technique	CNT amount (wt %)	Application	Membrane performance	Reference
MWCNTs/BPPO/ Triethanolamine (TEOA)	Phase inversion	5	UF	The rejection rate of egg albumin was 94%. The water flux increased from 197 to 487 L/(m^2·h) at 0.2 MPa	Wu et al. (2010a)
MWCNTs/polyester TFN	Interfacial polymerization	0.05	NF	The water flux through TFN membrane with 0.05% (w/v) MWNTs was 4.7 L/(m^2·h) . The salt separation capability of the membrane was improved	Wu et al. (2010b)
MWCNTs/MPDA	Interfacial polymerization	0.1	RO	The salt rejection rate of the membrane was 93.4%. The water flux increased from 11.1 to 13.6 L/(m^2·h)	Park et al. (2012)
MWCNT/polypropylene (PP) or PES	Interfacial polymerization	0.01–0.06	NF	The degradation rate of Brilliant Blue was more than 96%	Roy et al. (2011)
MWCNT/PSU	Phase inversion	1–20	MF	Pure water flux of MWCNT/PSU membranes was 1200 L/(m^2·h·bar)	Medina-Gonzalez and Remigy (2011)
MWNT/PA	Interfacial polymerization	0.005–0.2	RO	The water flux increased from 26 L/(m^2·h) to 71 L/(m^2·h)	Zhang et al. (2011)
MWNTs/PMMA	Interfacial polymerization	0.67	NF	The rejection rate of Na_2SO_4 was 99%. The water flux of the resultant membrane was $\sim 1.94 \times 10^{-3}$ $cm^3/(cm^2 \cdot s)$	Shen et al. (2013)
MWCNTs/polyester TFN	Interfacial polymerization	0–2.0 mg/ mL	NF	The water flux increased from 10.8 L/(m^2·h) to 21.2 L/(m^2·h) .	Wu et al. (2013)
f-MWCNT/PA/PSf	Interfacial polymerization	0.1	FO	The water flux was 95.7 L/(m^2·h) (nearly 160% higher)	Amini et al. (2013)
f-MWNTs/PA	Interfacial polymerization	0.1	RO	The water flux increased from 14.86 to 28.05 L/ (m^2·h) with the addition of 0.1% MWCNTs	Zhao et al. (2014)

DDA-MWNTs/PSf	Phase inversion	0.25	UF	The water flux of the resulting membrane was ~12 L/(m^2·h) at 1 bar.	Khalid et al. (2015)
NH2-functionalized MWCNT/PA	Interfacial polymerization	0–0.01	NF	The rejection rate of Na_2SO_4 and NaCl was 36.71% and 95.72% The flux increased from 48.6 (Na_2SO_4) and 48.1 L/(m^2·h) (NaCl) to 61.7 and 60.8 L/(m^2·h)	Zarrabi et al. (2016)
Sulfonated MWCNTs/ poly(piperazine amide)	Interfacial polymerization	0–0.02	NF/UF	The rejection rate of Na_2SO_4 was 96.8%. The water flux of the resulting membrane was 13.2 L/(m^2·h·bar) (1.6 times higher)	Zheng et al. (2017)
PMMA-modified MWNTs/PA	Interfacial polymerization	0.67–2.0 g/L	NF	The rejection rate of Na_2SO_4 was above 98% with the addition of 0.67 g/L PMMA-MWNTs, and the water flux was almost 1.5 times higher than the TFC membrane	Yu et al. (2013)
f-MWCNTs/PES	Layer-by-layer	1	UF	The irreversible fouling ratio of BSA was reduced from 49.3 ± 0.5 to 12.3 ± 2.9 after bilayer deposition of polyelectrolyte/MWCNTs	Liu et al. (2013)
Polycaprolactone (PCL)-MWCNTs/PES	Solution casting	3	–	The removal efficiency of Cd(II) increased from 8.7% to 27%. The water flux increased from 28 L/(m^2·h) to 61 L/(m^2·h)	Mansourpanah et al. (2011)
PVK/SWCNTs	Solution mixing	3	–	The membrane was tested by removing MS_2 bacteriophage virus (~2.5 logs)	Ahmed et al. (2013)
MWCNTs/aromatic PA	Polymer grafting	2.5–15 mg/g PA polymer	RO	The rejection rate of NaCl and HA increases to 3.17% and 1.67%. The permeability was decreased by 6.5%.	Shawky et al. (2011)
f-MWCNTs/GO/PVDF	In situ colloidal precipitation	0.001–0.1 g/L	UF	The removal efficiency of TDS, phosphorus, hardness, COD, chlorine, turbidity, color, and TSS was 1.51%, 6.55%, 21.79%, 75.5%, 76%, 81.94%, 86.3%, and 100%, respectively. The water flux were 43.99 , 52.62 , and 43.38 L/(m^2·h·bar) with the concentration of 0.1, 0.001, and 0.01 g/L	Ho et al. (2017)

(continued)

Table 4.3 (continued)

Material	Synthesis technique	CNT amount (wt %)	Application	Membrane performance	Reference
SBS/f-MWCNTs	Solution blending	0.01–0.1	NF	The PVDF/SBS-MWCNTs-SCN-Ag membrane had a tensile strength in the range of 12.6–20.1 MPa and a maximum decomposition temperature of 567–599 °C	Mehwish et al. (2015)
TiO_2-MWCNTs/PES	Phase inversion	0.1–1	NF	The pure water flux increased from ~3.71 to 5.66 kg/(m^2·h) at 5 bar with the addition of 1 wt% TiO_2-coated MWCNTs. The antifouling property of the PES membrane was decreased from 46.9% to 21.6% with addition of 1 wt% TiO_2 coated MWCNTs	Vatanpour et al. (2012)
f-MWCNTs/PES	Phase inversion precipitation	0.1	–	The water flux of the PCA-CNT membrane increased from ~10 to ~30 kg/(m^2·h) after 1 h with the addition of 0.1 wt% f-MWCNTs. The flux recovery ratio (FR) after passing whey solution increased from 44% to 95%. The membrane had a smooth and hydrophilic membrane surface	Daraei et al. (2013b)
f-MWCNTs/PES	Phase inversion	0.04–0.4	NF	The rejection rate of Na_2SO_4 had increased with the addition of 0.04 wt% MWCNTs. The pure water flux increased from ~5.5 to 9 kg/m^2·h at 4 bar with addition of 0.2 wt% MWCNTs	Vatanpour et al. (2011)
MWCNT/PVDF	Phase inversion	0–2	UF	The PVDF exhibited the highest protein adsorption (~70 mg/cm^2) with the addition of 2% MWNTHPAE content in casting solution. The pure water flux was to reach maximum when the MWNTHPAE/PVDF ratio was 1.5%. The flux	Zhao et al. (2012)

				recovery increased from 82% to 95.7% with the addition of MWCNTs	
f-MWCNTs/PES	Phase inversion	0–2	UF	The rejection rate of BSA increased from 81 to 88% with addition of 2% f-MWCNTs. The water flux was 184 L/(m^2·h) at 3 bar with the addition of 1 wt% f-MWCNTs	Rahimpour et al. (2012)
NH_2-MWCNTs/PES	Phase inversion	0–0.06	NF	The rejection rate of Na_2SO_4/$MgSO_4$/NaCl was 65%/45%/20% after 180 min of filtration. The water flux increased from 13.6 to 23.7 L/(m^2·h)	Vatanpour et al. (2014)
PVDF/GO/MWCNTs	Phase inversion	1	UF	The pure water flux was 251.73% higher than that of the original membrane when GO/OMWCNTs ratio was 5:5	Zhang et al. (2013)
MWCNTs/PANI/PES	In situ polymerization and phase inversion	0–2	UF	The water flux was1400 L/(m^2·h) (LMH)/bar (30 times higher). The NOM rejection rate (80%) was 4 times higher than that of the PES membrane	Lee et al. (2016b)
MWCNTs/TiO_2/PSf	Phase inversion	0–1	UF	The rejection rate of HA increased from 6% to 56% with the addition of 0.5% TiO_2–0.5% MWCNTs. The water flux increased from 10 L/(m^2·h) to 210 L/(m^2·h) with the addition of 1% MWCNTs	Esfahani et al. (2015)
MWCNTs/PVA	Pressure filtering deposition	0–20	UF	The rejection rate of the PEO was more than 90% with the addition of 5 wt% MWCNTs. The water flux increased from 1440 L/(m^2·h) by 20 w/% CNT concentration	de Lannoy et al. (2012)
MWCNTs/PVDF/ polydimethylsiloxane	Deposition and coating	0.05	MF	The rejection rate of Na_2SO_4 was ~80%. The water flux of the composite membrane was ~38 kg/(m^2·h) at 4 bar	Madaeni et al. (2013)

(continued)

Table 4.3 (continued)

Material	Synthesis technique	CNT amount (wt %)	Application	Membrane performance	Reference
MWCNT/PSf	–	0–10.55	UF	The water flux of the membrane increased from 2.5 to ~5.5 L/(m^2·h) with the addition of 6.94 wt % f-CNT	de Lannoy et al. (2013)
f-CNT/PSf	Phase inversion	0–0.5	UF	The pure water flux of the membrane increased from ~46 to 175 L/(m^2·h) at 100 kPa with the addition of 0.19% f-MWNTs. The adsorption of proteins was inhibited after the CNTs were added to the membrane	Qiu et al. (2009)
CNT/PES (C/P)	Phase inversion	0–4	UF	The highest pure water flux was 93 L/(m^2·h) with the addition of 0.5% CNT. The adsorption amount of BSA decreased from ~210 to ~75 μg/cm^2 at pH = 3 with the addition of 4% CNT	Celik et al. (2011a)
MWCNT/PVDF	Phase inversion	0–2	UF	The contact angle of the membrane decreased from 75.8° to 54.7° with the addition of 1 wt% f-MWCNTs. The pure water flux was 225 L/(m^2·h) (11 times higher) with the addition of 1 wt % f-MWCNTs. The rejection rate of BSA increased significantly (86.0%) with the addition of 0.5 wt% f-MWCNTs	Ma et al. (2013)
PVA/MWNTs/PAN	Electrospinning	10	UF	The membrane had a high water flux of about 270.1 L/(m^2·h) even at very low feeding pressure (0.1 MPa) with the addition of 10 wt% MWNTs. The same membrane also had a high separation rate (99.5%) of oil–water emulsion	You et al. (2013)

PVDF/MWCNTs	Phase inversion	1	UF	The PVDF/MWCNTs membrane had the highest water flux of about 620 L/(m^2·h) (114% higher). The rejection rate of BSA increased to about 31.8%.	Zhao et al. (2013b)
f-MWCNTs/ polyetherimide (PEI)/PA	Electrospinning	0.3	FO	The substrate porosity and the substrate tensile modulus of the membrane increased by 18% and 53%, and the structural parameter (S value) decreased by 30% with the addition of f-CNTs	Tian et al. (2015)
f-MWCNTs/chitosan/poly (vinyl) alcohol	Casting and evaporation	0–2	–	The amount of Cu(II) adsorbed doubled with the addition of 2 wt% MWCNTs (20.1 mg/g compared with 11.1 mg/g at 40 °C)	Salehi et al. (2012)
f-MWNTs/nano-silver/ PSf	Phase inversion and interfacial polymerization	0–5	UF	The addition of 5.0 wt% f-MWNTs in the support layer enhanced the pure water permeability of the n-TFN membrane by 23%	Kim et al. (2012)
f-MWCNTs/PES	Phase inversion	0–2	UF	COD and total phenol removal capacity of the prepared membrane was 72.6% and 89.5%, respectively. The pure water flux was 21.2 (kg/(m^2·h))	Zirehpour et al. (2014)
MWCNTs/PA and MWCNTs/PSf	Phase inversion and interfacial polymerization	5 and 10	MF/UF	PSU membrane with MWCNTs had a higher water flux (from 16.4 L/(m^2·h) at 2.3 bar to 18.4 L/(m^2·h) at 2.1 bar)	Kim et al. (2013)
PAA-modified MWCNTs (PAA-g-MWCNT)/PES	Phase inversion	0–0.1	NF	The resultant membrane had a high water flux of 40 kg/(m^2·h) at 0.4 MPa, excellent antifouling properties, and higher salt rejection	Daraei et al. (2013a)
f-MWCNTs/PES	Phase inversion	0–0.5	UF	The pure water flux increased from 24.28 L/(m^2·h) to 53.91 L/(m^2·h) with the addition of 0.5 wt% of f-MWCNTs. The hydrophilic property of PES/f-MWCNTs was improved by 18.7%. The rejection rate of the membrane was 27–30%, much higher than that of a PES membrane	Saranya et al. (2014)

(continued)

Table 4.3 (continued)

Material	Synthesis technique	CNT amount (wt %)	Application	Membrane performance	Reference
f-CNTs/PES	Phase inversion	0–0.5	–	The water flux increased from 260 to 375 and 450 L/(m^2·h) with the addition of 0.02 and 0.04 wt % f-CNTs	Phao et al. (2013)
f-MWCNTs/ polyvinylpyrrolidone (PVP)/PES	Phase inversion	0–0.5	UF	The removal efficiency of bovine serum albumin, pepsin, and trypsin was 93.4%, 74.7%, and 59.4%, respectively	Masoomaa et al. (2015)
PES/f-MWCNTs	Phase inversion	0.01	UF and hemodialysis	The water flux increased from 22.57 to 149.67 L/(m^2·h·bar) with the addition of 0.1 wt% of f-MWCNTs	Abidin et al. (2017)
PES membrane/ZnO–MWCNTs	Non-solvent-induced phase inversion	0–1	NF	The rejection rate of Direct Red 16 was more than 90%. The water flux was 16.7 kg/(m^2·h) with the addition of 0.5 wt% ZnO–MWCNTs	Zinadini et al. (2017)
Mixed isotactic polypropylene membrane/f-MWCNTs	Melt mixing and melt pressing	4	–	The water permeability of the membrane increases by a factor of ~35 with the addition of 4 wt% MWCNT-g-PP	Bounos et al. (2017)
PVDF/Fe_2O_3/MWCNTs	In situ polymerization	0.2	–	For degradation of cyclohexanoic acid (CHA) by membrane in the presence of H_2O_2, the removal rate reached 48% in 24 h. For HAs, removal in the presence of H_2O_2 was much higher than that without H_2O_2	Alpatova et al. (2015)
PA/MWCNTs RO membrane	Interfacial polymerization	0–0.01	RO	The saline solution fluxes of the membrane increased from 20.3 ~25.9 to 28.9 L/(m^2·h) by adding f-MWCNTs. The salt (NaCl) rejection rate was >96% by adding MWCNTs. The membranes with f-MWCNTs had better antifouling properties than the original membrane	Farahbakhsh et al. (2017)

MWCNTs/PES	Phase inversion	0–3	UF	The adsorption amount of BSA decreased from 58.96 μg/cm^2 to 41.63 μg/cm^2. The water flux increased from 5.18 L/(m^2·h) to 71.26 L/(m^2·h) with the addition of 1.5 wt% f-MWCNTs	Wang et al. (2015b)
PA/MWCNTs NF membranes	Non-solvent-induced phase inversion	0.001–0.01	NF	The membrane with 0.005 wt% f-MWCNT added had the largest water flux. The membrane was improved in the rejection rate of Na_2SO_4 by the addition of f-MWCNTs	Mahdavi et al. (2017)
MWCNTs/PSf TFN membrane	Solution mixing	0.01–0.05	RO	The membrane with 0.03% f-MWCNTs added to the PA layer reached the highest water permeability after 48 hours of treatment with 1.5 M H_2SO_4. The NaCl rejection rate was higher than 96%	Wan Azelee et al. (2018)
f-MWCNTs-PAN/PP	Electrostatic spraying	1	–	The removal rate of Indigo was 98.73%. The water flux was 3891.85 L/m^2·h at a low pressure of 0.1 MPa	Xu et al. (2017b)
PHB–CaAlg/CMWCNT	Electrospinning	–	–	The adsorption rate of Brilliant Blue was 98.20%. The water flux of the resultant membrane was 32.95 L/m^2·h	Guo et al. (2016)
PSF/PVP/gCNT	Phase inversion	0.2	UF	The separation efficiency of oil–water was nearly 100% and the flux reached 121 LMH	Santosh et al. (2018)
f-MWCNT/PES	Phase inversion	0.4	NF	The rejection rate of Rhodamine B, crystal violet, indigo carmine, and orange G was 99.23%, 98.43%, 87.12%, and 82.13%, respectively	Mohammad et al. (2018)
PVDF/CNT	Layer-by-layer and simultaneous	0.02%	–	CNT blend membrane showed better wettability, higher permeability, and better MB removal efficiency and had a more open structure	Mavukkandy et al. (2018)
PSf/pebax/f-MWCNTs	Solution casting and solvent evaporation	0–2	NF	The separation efficiency of oil–water was 99.79% with the addition of 2 wt% f-MWCNTs	Saadati and Pakizeh (2017)

(continued)

Table 4.3 (continued)

Material	Synthesis technique	CNT amount (wt %)	Application	Membrane performance	Reference
PVDF-CNT/PU/PVDF-CNT	Sequential electrospinning	0–1	UF	The separation efficiency of oil–water was more than 94%	Gu et al. (2018)
CNT-PA	Interfacial polymerization	0–0.01	RO	The rejection rate of BSA was more than 96%. The water flux increased from 20.3 to 28.9 L/(m^2·h) with the addition of CNTs	Farahbakhsh et al. (2017)
PES/CNT	Phase inversion	0.01–1	NF	The rejection rate of Na_2SO_4 was 87.25%. The water flux was 38.91 L/m^2 h by the addition of 0.1 wt% CNT	Wang et al. (2015a)
PES/CNT	Phase inversion	0–0.1	NF	Membrane blended with 0.1 wt% f-MWCNTs had a highest permeation flux as well as dye (acid orange 7) removal efficiency (99%)	Ghaemi et al. (2015)
PES-SWCNT	Phase inversion	0–0.5	–	The rejection rate of bisphenol A and nonylphenol was 56% and 76% (improve more than 60%)	Kaminska et al. (2015)

Table 4.4 Application of electrochemical carbon nanotube membranes

CNT membrane	Voltage (V)	Target contaminant	Removal efficiency	Reference
COOH-MWNT	2.0	Ibuprofen	~100%	Bakr and Rahaman (2016)
CNT-PTFE	8.0	Pb^{2+}	98.8%	Gao et al. (2017b)
CNT-PVA	7.0	Cr (VI)	>99%	Duan et al. (2017)
N-CNT	–	TOC/NH^{4+}	95.2%/97.7%	Zuo et al. (2016)
Fe-CNT	1.0	Metoprolol	97%	Yanez et al. (2017)

The electrochemical carbon nanotube membranes exhibited great potential on wastewater treatment due to high degradation efficiency, low energy consumption, and simple operation process (Motoc et al. 2013; Bakr and Rahaman 2016, 2017; Liu et al. 2017). Besides, by transferring electrons directly through the surface of the electrochemical carbon nanotube membrane electrode, the solute transfer restriction of the conventional batch electrochemical process was overcome. Therefore, this method was more advantageous than conventional batch electrolysis. Table 4.4 provides some works on electrochemical carbon nanotube membranes. For example, Wei et al. (2017b) prepared a novel carbon nanotube-based hollow fiber membrane with a sandwich-like structure. Low concentration of microcystin-LR (0.5 mg/L) was removed economically and efficiently (>99.8%) by simple switching with adsorption and desorption as well as electrochemical oxidation by these carbon nanotube ultrafiltration membranes.

4.4 Graphene-Based Membranes

Graphene, consisting of a compact accumulation of sp2 hybrid carbon atoms, was reported for the first time by Geim and Novoselov (2004). Since then, graphene and graphene-based materials have been extensively studied and used to synthesize various multifunctional materials. As we know, graphene can be obtained by chemical vapor deposition or chemical reduction of graphene oxide. Generally, it is easy to fabricate single-layered or several-layered graphene on some catalytic substrates via chemical vapor deposition. Compared with the tedious and expensive chemical vapor deposition, reducing graphene oxide is more favorable for scale production. Graphene oxide is usually prepared by oxidizing graphite through the famous Hummer's method, which has abundant oxygen-containing functional groups on its surface and edges. After chemical reduction by hydrogen iodide acid, hydrazine, or thermal treatment, the oxygen-containing groups are reduced to obtain reduced graphene oxide which possesses similar properties to graphene. To date, both graphene and graphene oxide have also been applied to construct novel

membranes with laminar pores. Besides, these materials are also used as blender to improve the hydrophilicity, surface charges, and antifouling ability of the polymeric membranes.

4.4.1 Support-Free Graphene Membranes

The ideal separation membrane should possess uniform pore size, ultrathin thickness, high mechanical strength, and excellent physicochemical properties to provide good permeability and selectivity. Graphene membrane may be a suitable candidate to meet such requirements. According to the theoretical calculation, the single-layered graphene membrane can completely desalinate brine water and seawater, showing great potential for water treatment (Cohen-Tanugi and Grossman 2012).

Previous research suggested that salt rejection was negatively correlated to improve pore size and applied pressure (Anand et al. 2018). Meanwhile, ionization of functional groups surrounding nanopores could influence desalination efficiency of single-layered graphene membrane (Chao et al. 2017). Therefore, single-layered graphene membranes could achieve highly permeable desalination by controlling the pore size and functional groups of nanopores (Cohen-Tanugi and Grossman 2012). To date, the nanopores in single-layered graphene membranes were usually produced by ion beam and electron beam exposure, ion bombardment, UV-induced oxidation etching, hydroxyl radical etching, oxygen plasma etching, etc. (Anand et al. 2018). O'Hern et al. (2014) reported their works on the controllable high-density subnanometer pores in single-layered graphene membranes which allowed the transport of salt but rejected larger organic molecules.

Compared with single-layered graphene membranes, Celebi et al. (2014) reported highly efficient mass transfer across physically perforated double-layered graphene membrane. Wei et al. (2017a) reported a four-layered graphene membrane with about 2 nm thickness, indicating outstanding permeability and selectivity. Cohen-Tanugi et al. (2016) also reported a reverse osmosis membrane stacked by multilayer nanoporous graphene for desalination by using classical molecular dynamic simulation. They found that double-layered nanoporous graphene membranes with the 3.0 Å of nanopore radius exhibited full salt rejection. Compared to the single-layered graphene membranes, the bilayer nanoporous graphene membranes showed excellent salt rejection. Recently, the effects of pressure and wall interaction on the water transport through multilayer nanoporous graphene membranes were carried out by molecular dynamic simulation (Shahbabaei et al. 2017). They found the water flux was mostly doubled in the multilayered hydrophilic pore membrane owing to strong hydrogen bonds. And then Chang et al. (2017) reported the nanofiltration properties of reduced graphene-based membrane with adjustable porous structure. Similarly, Yi (2013) prepared ultrathin (≈22–53 nm thick) graphene nanofiltration membranes on microporous substrates. The performance of such ultrathin graphene nanofiltration membranes was tested on a dead-end filtration device, and the pure water flux of ultrathin graphene nanofiltration membranes was high (21.8 L/

$m^2 \cdot h \cdot bar$). Furthermore, Kabiri et al. (2016) synthesized a thiol-functionalized graphene composite with a unique three-dimensional porous structure to remove mercury ions (Hg^{2+}) from water. The results indicated that the removal efficiency of the membrane reached almost 100% for low (4 mg/L) and high (120 mg/L) concentration of Hg^{2+}. Due to excellent permeability and selectivity, support-free graphene membranes exhibited great potential in selective ion transportation and separation.

4.4.2 Graphene Oxide Membranes

Recently, graphene oxide has attracted increasing attention on membrane preparation and modification due to its excellent hydrophilic properties (Choi et al. 2013). Graphene oxide is usually obtained by oxidizing graphite with a strong acid or oxidant. Graphene oxide is a reforming form of graphene in which oxygen and hydrogen atoms are bonded with carbon atoms (Hu and Mi 2013). Due to the presence of oxygen- and hydrogen-based functional groups, graphene oxide can be well dispersed in water and other organic solvents, which favors the preparation of graphene oxide-based membranes (Stankovich et al. 2007).

Sun et al. (2014a) used graphene oxide membranes to recover acids from iron-based electrolyte wastewater. The mechanism was that Fe^{3+} was blocked by graphene oxide membranes, while H^+ could migrate fast. Sun et al. (2014b) also studied ion mobility and interactions with graphene oxide membranes. They found that ion permeability exhibited the order of $Mg^{2+} > Na^+ > Cd^{2+} > Ba^{2+} = > Ca^{2+} > K^+ > Cu^{3+} > Fe^{3+}$. Various interactions between ions and graphene oxide sheets, such as chelation, static electricity, van der Waals forces, etc., were attributed to the selectivity of graphene oxide membranes. Figure 4.8 showed the schematic diagram of

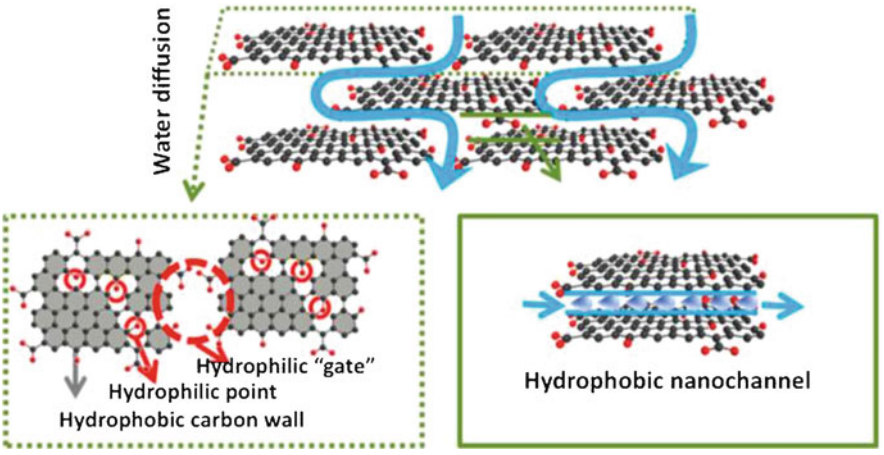

Fig. 4.8 Nanochannels in a graphene oxide membrane and hydrophilic pores for water flow in desalination. (Reprinted with permission of (Wang et al. 2016a))

graphene oxide membranes for water transport (Wang et al. 2016a). Water molecules firstly arrived in the hydrophilic sites in graphene oxide and then slipped through the hydrophobic nanochannel with low or no friction.

A dopamine-coated polysulfone membrane has been prepared to investigate the dependence of water flux and charge effect on separation. They revealed that the water flux was independent of the number of graphene oxide layers and salt exclusion but depended on interlayer spacing (Hu and Mi 2013). However, the volume of graphene oxide membrane would swell in the aqueous environment. Nair et al. (2012) studied the water mobility in nanochannels between graphene oxide tablets under different condition. They showed that the interlayer spacing between the original graphene oxide membrane region and the stacked graphene oxide membrane was about 0.6 nm in the dry conditions. Because of the diffusion of water molecules to graphene oxide layer, the increased interlayer spacing between graphene oxide membranes resulted in high mobility for water molecules. However, when the graphene oxide membrane was immerged in an ionic solution, the increased gap by the hydration cannot repel K^+ and Na^+ ions, making the membrane inappropriate for desalination applications (Joshi et al. 2014). Addressing to this issue, graphene oxide was functionalized with glycine and carboxylation for preparing membrane by pressure-assisted self-assembly to achieve high salt rejection efficiency (Yuan et al. 2017). Xu et al. (2017a) reported that the water flux and separation ability of graphene oxide membrane was related to the inner nanostructure of graphene oxide membrane. In addition to the interlayer spacing, it was found that the morphological characteristics of graphene oxide membranes, such as corrugation, could improve the separation performance (Qiu et al. 2011). Wang et al. (2012) presented that a graphene oxide/polyelectrolyte composite membrane had obvious nanofiltration performance in removing dyes, separating monovalent and divalent ions, and dehydrating solvent–water mixture. O'Hern et al. (2014) also verified the water purification and ion permeation (rather selective) properties of the graphene oxide membrane.

Similar to the study of graphene oxide membrane in ion transport, Chang et al. (2017) reported that carboxylation could increase the hydrophilicity of graphene oxide membrane, improving the efficiency of dye removal. Such improvement was potentially attributed to surface charge density. On the contrary, it was found that reduced preoxidized graphene membrane could increase the rejection efficiency of methyl orange dye to >90%. In addition, a graphene oxide hydrogel membrane was synthesized by Qin et al. (2012) via suspending the graphene oxide (graphene oxide) in water. This graphene oxide hydrogel exhibited pH responsiveness and good mechanical properties. Meanwhile, graphene oxide hydrogel had a good adsorption capacity for organic dye Rhodamine B and anionic chromate.

Graphene oxide membrane also possessed superior metal ion adsorption characteristics. The graphene oxide membranes, which were modified with hyperbranched polyethylenimine, were applied to obtain high permeability and rejection (>90%) of heavy metal ions (Zhang et al. 2015). The divalent metal ions,

such as Co^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+}, Pb^{2+}, etc., could be chemically adsorbed by graphene oxide membranes, and the membranes could be reused for up to ten cycles (Sitko et al. 2016).

Nowadays, graphene oxide membranes were also applied to oil–water separation. With vacuum-assisted filtration, Zhao et al. (2016) intercalated palygorskite nanorods into adjacent graphene oxide nanosheets and assembled graphene oxide nanosheets into laminate structures to prepare the freestanding graphene oxide membranes. Under various conditions (different concentration, pH, or oil species), the graphene oxide membranes showed excellent anti-oil performance in the separation process of water-containing oil emulsion.

4.4.3 Graphene Oxide Hybrid Membranes

Although graphene oxide membranes with a good desalination capability can be prepared by simple methods, these membranes could be trapped by the use of pressure-driven systems. Liu et al. (2015) found that the composite membrane prepared by adding graphene oxide to polysulfone displayed superior pressure-resisted ability, mechanical strength, and water permeability.

In order to increase water flux further, Dai et al. (2015) introduced a large quantity of nitrogen-containing and oxygen-containing groups into the surface of graphene oxide membrane and filled the interlayer space with polypropylene. The novel polypropylene-based composite membrane apparently improved the hydrophilicity and adsorption capacity. With the development of materials science, membranes consisted of polymeric materials, including nylon, aromatic polyamides, polyvinylidene fluoride, polysulfone, and polyethersulfone, as well as non-polymer materials, such as ceramics, metals, and composites, which have been readily fabricated and applied on the filtration of diverse solutions. Compared to pure polymer membranes, the polyamide membranes doped with graphene oxide showed higher water flux and desalination rate (Bano et al. 2015). The resultant increase in the permeate water flux was from 1.8 $L/(m^{2\cdot}\ h^{1})$ to 22 $L/(m^{2\cdot}\ h^{1})$, while salt rejection maintained at essentially above 80%. Similarly, research conducted by Lai et al. (2016) demonstrated that water flux and salt removal were improved by integrating graphene oxide in polyamide membrane. Moreover, Ali et al. (2016) prepared thin composite membranes embedded with graphene oxide to evaluate their desalination performance. They found that adding a small amount of graphene oxide (100 ppm) significantly improved water flux and mechanical stability and reduced membrane fouling. For salt solution with 2000 ppm NaCl, the launching flux at 1.5 MPa was 29.6 L/m^2, and the salt removal rate was 97%. Besides, Kochameshki et al. (2017) synthesized a polysulfone nanocomposite membrane modified with graphene grafted with diallyldimethylammonium chloride. The results showed that the water flux increased to about 450 $L/m^2{\cdot}h$, the antifouling performance was improved, and the heavy metal ion rejection rate increased to 86.68% (Cu^{2+}) and 88.68% (Cd^{2+}).

In addition, polyethylenimine membrane integrated with tannic acid-functionalized graphene oxide showed excellent ion separation performance against NaCl and $MgSO_4$ (Lim et al. 2017). A thin nanofiltration membrane was prepared by aggregating piperazine and trimesoyl chloride with reduced graphene oxide/TiO_2 composite, which demonstrated good separation performance and antifouling property in cross-flow filtration system due to the hydrophilicity of reduced graphene oxide (Safarpour et al. 2015b). Zhang et al. (2017c) synthesized a novel layered structure membrane which was prepared by coating graphene oxide sheets on the surface of electrospun aminated polyacrylonitrile (APAN) fibers, exhibiting ultrahigh flux (10,000 L/(m^2·h)), promising rejection (98%) and excellent antifouling performance for the separation of oil–water emulsions. Besides, Choi et al. (2013) also fabricated a dual-action barrier coating layer of graphene oxide on the surface of polyamide reverse osmosis membrane. The antifouling tests indicated that the graphene oxide coating layer can increase the surface hydrophilicity and decrease the surface roughness, which promoted the significantly improved antifouling performance against a protein foulant. Similarly, graphene oxide nanosheets were successfully doped across 200-nm-thick polyamide membranes by He et al. (2015). They observed the significant increase of water flux (80%) in the reverse osmosis membranes modified with graphene oxide nanosheets. Moreover, polyamide nanofiltration membranes modified with reduced graphene oxide–NH_2 were prepared by Li et al. (2017b) to enhance water flux and antifouling capability. There were some researchers reporting the improvement in the chlorine resistance of the polyamide membranes incorporated with graphene oxide (Safarpour et al. 2015a). In their opinion, the chemically stable graphene oxide plate embedded in the polyamide layer acted as a barrier layer, protecting the polyamide from chlorine erosion, as shown in Fig. 4.9 (Choi et al. 2013).

The researchers also identified that adding graphene to polymer membranes had positive influences on dye absorption. Polypyrrole-hydrolyzed polyacrylonitrile composite NF membrane doped with graphene oxide was prepared by Shao et al. (2014). It is found that the effectiveness of Rose Bengal dye rejection was approximately 99.0%, and the solvent permeability was enhanced. And the NF performance of graphene oxide mixed polyether sulfone membrane used for dyestuff (Direct red 16) removal was higher than that of polyethersulfone membrane (99% vs. 90%) (Zinadini et al. 2014). The NF membrane fabricated by multilayered deposition of graphene oxide on a polysulfone support exhibited high water permeability and superior rejection (93–95%) of Rhodamine B dye (Qiu et al. 2011). In addition, a polyamide membrane assembled with carboxyl-functionalized graphene oxide showed 98.1% dye rejection rate of the New Coccine (Zhang et al. 2017b).

Due to superior separation characteristic, graphene oxide-doped polymer membranes were also applied on oil–water separation. Hu et al. (2015) successfully fabricated a novel graphene oxide hybrid membrane on commercial 19-channel ceramic by adopting a vacuum method. During the treatment, the water permeation fluxes of modified membranes were about 667 L/(m^2·h·bar) after 150-min operation,

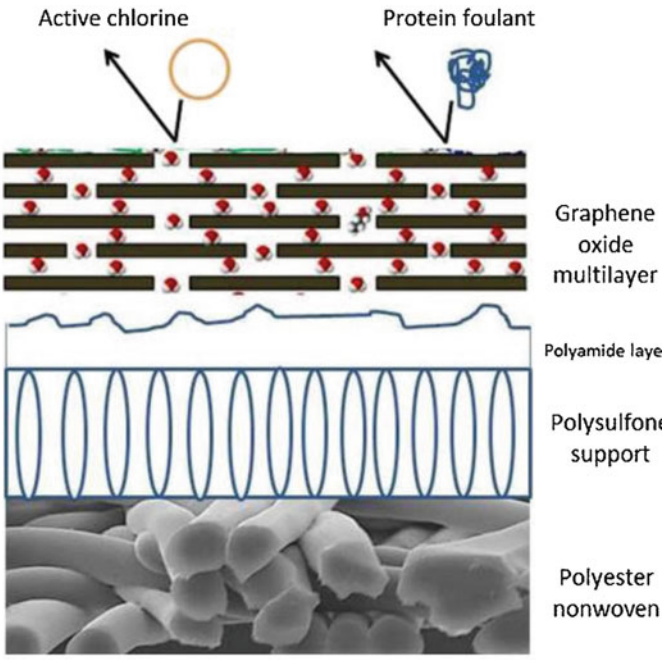

Fig. 4.9 Graphene oxide protective layer against foulants and active chlorine in the polyamide membrane. (Reprinted with permission of (Choi et al. 2013))

which was higher about 27.8% than that of the unmodified membrane (522 L/(m^2·h·bar)). These results showed that graphene oxide modification played a crucial role on improving oil–water separation performance. Similarly, in addition to the application of membrane in above wastewater treatment, the novel membranes were more widely applied to more intricate wastewater (Huang et al. 2015). Zinadini et al. (2015) synthesized three different hybrid membranes which were fabricated in three concentrations of 13, 15, and 17 wt% of polyethersulfone polymer. Polyethersulfone/graphene oxide membrane with 15 wt% of polyether sulfone and graphene oxide content of 0.5 wt% showed the most superior performances and was selected as optimal membrane for treatment of milk processing wastewater. Similarly, Sun et al. (2015) developed an antibiofouling membrane by in situ fabrication of graphene oxide–AgNPs onto cellulose acetate membranes. The presence of graphene oxide–AgNPs composite on the membrane caused an inactivation of 86% *Escherichia coli* after contacting with the membrane for 2 h. Compared to modifying graphene oxide with active substances, graphene oxide hybrid membranes by adding graphene oxide into polymer membranes achieve more significant advantages on improved water flux, mechanical stability, and fouling resistance. There is no doubt that graphene oxide hybrid membranes will provide us the new insight on the optimization of graphene-based membranes (Table 4.5).

Table 4.5 Application, membrane performance, and other conditions of mixed-matrix graphene oxide membranes

Membrane material	Synthesis technique	Application	Membrane performance	Reference
PES/GO/PAA	Solution casting	Remove synthetic melanoidin solution	54% color removal	Kiran et al. (2015)
Polycation/GO multilayer membrane	Self-assembly-assisted layer-by-layer deposition	Remove dye from water	The flux and retention rate could reach 6.42 kg/(m^2·h·bar) and 99.2%	Wang et al. (2016b)
MgSi@RGO/PAN composite membrane	Vacuum filtration and deposition	Desalination, wastewater treatment, separation, and purification	The membranes can effectively reject small molecules	Liang et al. (2016a)
PES-GO-4	Interfacial polymerization	Water or wastewater treatment applications	The PES-GO-4 membrane exhibited 2.6 times greater flux recovery than an unmodified PES-UF membrane	Efosa et al. (2016)
GO/APAN membrane	Electrospinning-assisted layer-by-layer assembly technique	Separation of oil–water emulsion	This membrane exhibited ultrahigh flux (~10,000 LMH), preferable rejection rate (≥98%), and remarkable antifouling performance	Zhang et al. (2017c)
Polysulfone–Fe_3O_4/GO mixed-matrix membrane	Immersion phase inversion	Water treatment during the backwashing procedure	The novel polysulfone–Fe_3O_4/GO mixed-matrix membrane was having 3 times higher permeate flux than the neat PSf membrane	Chai et al. (2016)
GO-ZnO membranes	Double-casting phase inversion (DCPI)	Wastewater reclamation	The novel membranes exhibited higher fluxes, with less fouling and high rejection rate of TOCs.	Mahlangua et al. (2016)
TA/GOQDs TFN membrane	Interfacial polymerization	Wastewater treatment, separation, and purification	The TA/GOQDs TFN membrane showed a pure water flux up to 23.33 L/(m^2·h) (0.2 MPa), and high dye rejection to Congo red (99.8%) and methylene blue (97.6%) was kept	Zhang et al. (2017a)

(continued)

Table 4.5 (continued)

Membrane material	Synthesis technique	Application	Membrane performance	Reference
3D PPy@GO membrane	One-step electrochemical co-deposition	Wastewater treatment	The 3D PPy@GO composite-coated electrodes showed excellent permselectivity of Pb^{2+} with a flux of 4.7 g/(m^2·h), a current efficiency of 51.9%, and excellent cycling stability	Gao et al. (2017a)
PVA/PAA/GO-COOH@PDA	Electrospinning technique	Wastewater treatment and dye removal	The PVA/PAA/GO-COOH@PDA composite materials showed efficient adsorption capacity towards the three model dyes. The composite membranes can be easily separated and regenerated from wastewater dye solution and demonstrated excellent reusability	Xing et al. (2017)
GPC ultrafiltration membrane	Drop-coating combined with vacuum filtration	Complex industrial wastewater streams	The membrane exhibited an excellent rejection coefficient of 99.2% for methylene blue and the permeation flux was 12 L/(m^2·h) at 0.1 bar	Wang et al. (2018a)
CG RO membranes	Embedding and melting method	Desalination	The RO membrane performance showed that the permeate flux of membrane increased from 1.67 L/(m^2·h) to 4.74 L/(m^2·h)	Chen et al. (2017)
PVA–GA composite membranes	Cross-linking and polymerization methods	Removing an industrial textile dye from wastewater	The nanofiltration membrane showed lowest fouling rate during removal of the industrial direct dye (flux recovery ratio, 96.60%; reversible fouling ratio, 23.82%; and irreversible fouling ratio, 3.39%)	Liu et al. (2018)

4.5 Carbon Fiber Membranes

Since Shimpei (1986) accidentally found that carbon fibers facilitated microbial attachment, and possessed excellent adsorption capacity for pollutants, the research works focused on carbon fibers for water treatment were widely carried out. It was believed that these advantages opened the "surprise door" for the application of carbon fibers (Xu and Luo 2012; Manawi et al. 2018). Especially, carbon fiber membranes, as one of the novel membrane materials, have been explored and applied in recent years (Xiao et al. 2016).

4.5.1 Support-Free Carbon Fiber Membranes

The support-free carbon fiber membranes are generally obtained by forming carbon fiber precursors into membrane shape and then stabilized and carbonized via thermal treatment. Beck et al. (2017) prepared carbon nanofiber membranes by electrospinning followed by carbonization (Fig. 4.10). The adsorption capacity, permeability, and adsorption kinetics of the carbon nanofiber membranes were about 10, 6, and 2 times larger than that of the traditional activated carbon

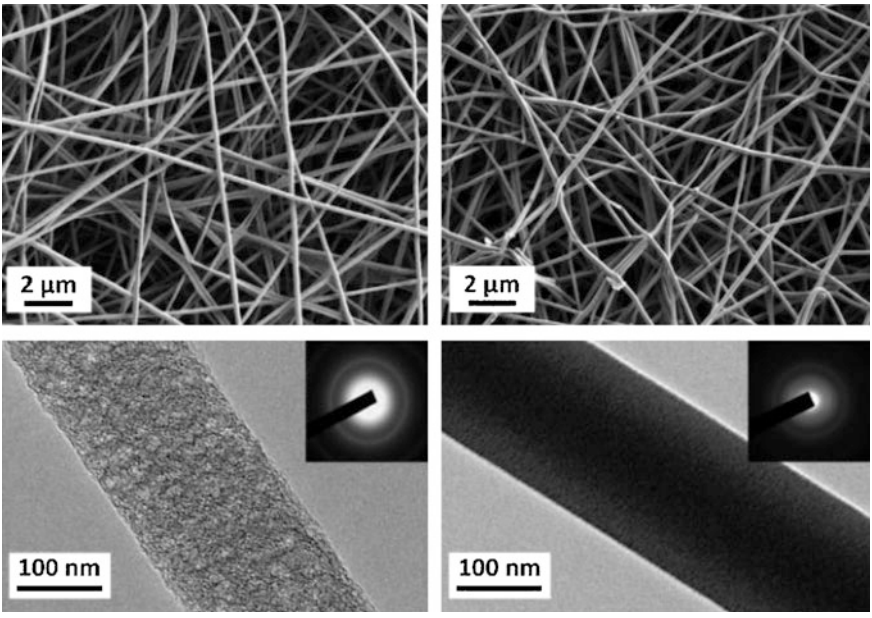

Fig. 4.10 SEM (top) and TEM (bottom) images of electrospun carbon nanofiber membranes prepared from the precursors of lignin/PVA (left) and PAN (right). The insets in the TEM images show the electron diffraction patterns. (Reprinted with permission of (Beck et al. 2017))

membrane, respectively. However, such carbon fiber membranes usually suffered from serious membrane fouling, limiting their application.

4.5.2 Carbon Fiber Hybrid Membranes

In order to expand the application of carbon fiber membrane in water treatment and improve the removal efficiency of pollutants, researchers have developed a variety of carbon fiber hybrid membranes, which combined the advantages of carbon fiber and membrane technology, improving its treatment efficiency.

Yang and Tsai (2008, 2009) prepared carbon fibers/carbon/alumina tubular composite membrane and applied it in a cross-flow electrocoagulation/electrofiltration module for Cu chemical mechanical polishing wastewater treatment. Under the optimal experimental conditions, the turbidity of the permeate was less than 1 NTU, and the removal rates of total solid content, copper, total organic carbon, and silicon were 72%, 92%, 81%, and 87%, respectively. Li et al. (2013a, b) reported their works on domestic sewage treatment using biological carbon fiber membrane. The biological carbon fiber membrane could effectively intercept sludge and most organic matter. Moreover, the bio-carbon fiber inside the membrane had a strong adsorption performance, which could further adsorb the organic matter across the membrane surface, thus ensuring a higher and more stable removal rate of organic matter.

Besides, Tai et al. (2014) developed a novel freestanding and flexible electrospun carbon–silica composite nanofibrous membrane. This composite membrane was more tough than the original carbon nanofibers when the SiO_2 concentration was 2.7 wt%. They found that after coating with silicone oil, the composite membrane became ultrahydrophobic and superoleophilic, which enabled the membrane to serve as an effective substrate for separating free oil from water. Yue et al. (2018) fabricated layered porous dynamic separation membranes containing primary and secondary nanostructures by in situ growth of ZnO nanowires on carbon fibers (Fig. 4.11). The

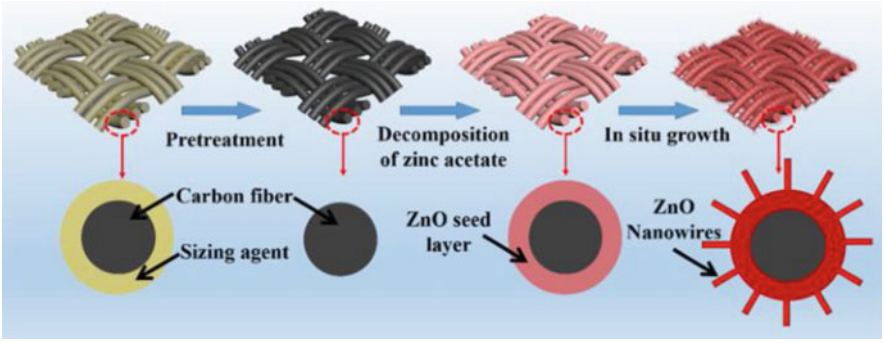

Fig. 4.11 Fabrication process of ZnO–carbon fiber dynamic membrane. (Reprinted with permission of (Yue et al. 2018))

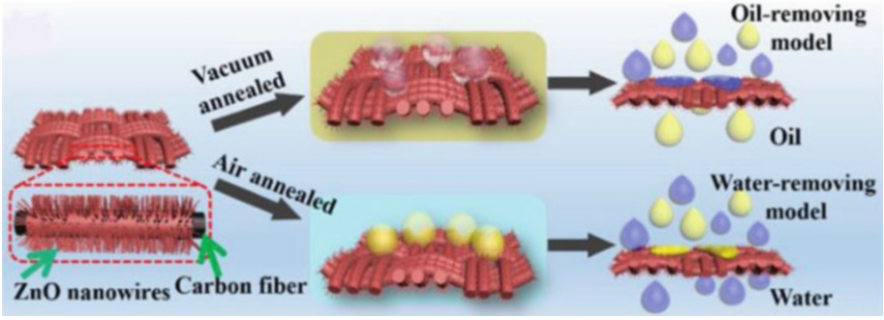

Fig. 4.12 The switchable wettability of ZnO–carbon fiber dynamic membrane when annealed in different atmosphere and the corresponding separation capacities of oil–water mixtures. (Reprinted with permission of (Yue et al. 2018))

membrane could switch wettability between high hydrophobicity and superhydrophilicity by simply annealing alternatively in vacuum and air environment (Fig. 4.12) and indicated more than 98% separation efficiency in deoiling and dewater modes. Han et al. (2017) prepared 3D structural Fe_2O_3–TiO_2@activated carbon fiber membranes by a modified electrospinning process followed by a thermal treatment. The membrane possessed high adsorption and visible light excitable photocatalytic properties and could be used to remove dyes and heavy metal ions.

4.5.3 The Composite Membranes Using Carbon Fiber Cloth as the Substrate

These composite membranes usually are obtained by loading various functional materials on carbon fiber cloth, which is adopted as the substrate. They can combine the advantages of functional materials and membrane technology. Meanwhile, the carbon fiber substrate has good mechanical properties and can reduce the loss of functional material in the process of water treatment.

Li et al. (2016c) successfully prepared a catalytic cathode membrane on the basis of low-cost carbon fiber cloth with Pd-reduced graphene oxide–$CoFe_2O_4$ catalyst (Fig. 4.13). The cathode membrane was used in microbial fuel cell/membrane bioreactor coupling system, exhibiting great potential on simulated wastewater treatment. Xiao et al. (2017) obtained carbon fiber/C_3N_4 cloth by a dip-coating and thermal condensation method with carbon fiber cloth as substrate (Fig. 4.14). The carbon fiber/C_3N_4 cloth possessed excellent flexibility and strong visible light absorption, which displayed good treatment performance for the degradation of flowing wastewater. To further improve the treatment efficiency, Shen et al. (2018) inserted TiO_2 between C_3N_4 and carbon fiber (Fig. 4.15). The carbon fiber/TiO_2/C_3N_4 cloth showed enhanced photocatalytic activity for degrading various organic pollutants in comparison with carbon fiber/C_3N_4 cloth.

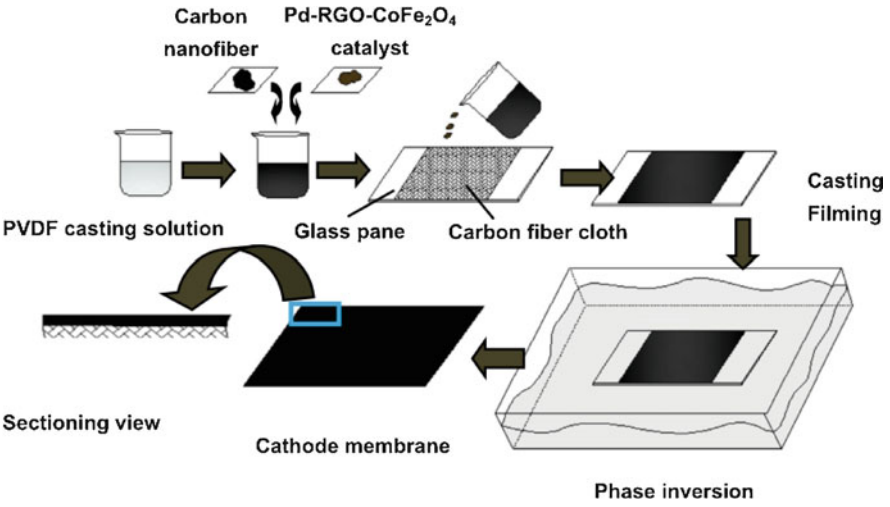

Fig. 4.13 The preparation process of cathode membrane. (Reprinted with permission of (Li et al. 2016c))

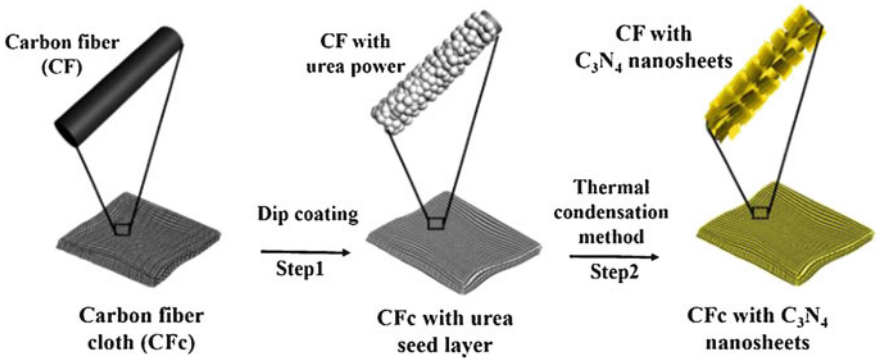

Fig. 4.14 Schematic illustration of the preparation process of carbon fiber/C_3N_4 cloth. (Reprinted with permission of (Xiao et al. 2017))

4.6 Activated Carbon Membranes

Activated carbon, as a unique multifunctional material with high surface area, micro–meso and macroscopic structure, and various chemical functional groups, is recognized worldwide as one of the most popular adsorbents in water treatment (Amit et al. 2013; Danish and Ahmad 2018). Up to now, activated carbon has been widely used in various industrial processes including food processing (Alvarez et al. 2011), chemical manufacturing (Jaria et al. 2018), pharmaceutical (Karelid et al. 2017), paper making (Ou Yang et al. 2013), etc. to remove water-soluble chemical

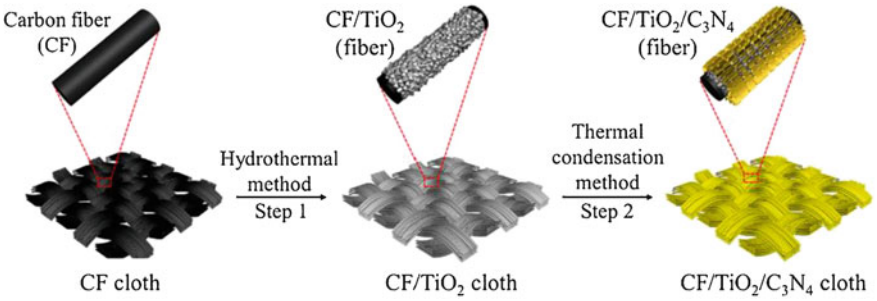

Fig. 4.15 Schematic illustration of the preparation of TiO_2/C_3N_4 heterojunctions on carbon fiber cloth. (Reprinted with permission of (Shen et al. 2018))

pollutants from inorganic and organic wastewater (Abdel-Nasser and El-Hendawy 2001; Mohammed 2011). Jacangelo (1995) found that activated carbon could adsorb organics to prevent the formation of membrane fouling in membrane separation processes. Several studies also demonstrated that membrane bioreactor achieved high removal efficiency for trace organic pollutants in synthetic and real wastewater by the use of granular activated carbon (Amaral et al. 2014; Jia et al. 2014). In this section, the membrane materials integrated with activated carbon, including activated carbon-coated membranes, support-free activated carbon membranes, and activated carbon mixed-matrix membranes for wastewater treatment, were described as follows.

4.6.1 Activated Carbon-Coated Membranes

Activated carbon could be coated on membranes to enhance membrane separation performance while removing contaminants from wastewater. Thiruvenkatachari et al. (2006) prepared activated carbon pre-coated microfiltration hollow fiber membrane using wood-based, coal-based, and coconut shell-based activated carbon for wastewater treatment (Fig. 4.16). After 8 h of operation, 63% of organic pollutants were removed by wood-based activated carbon-coated membrane, 57% by coal-based activated carbon-coated membrane, and 56% by coconut shell-based activated carbon-coated membrane, which were higher than that of non-pre-coated membrane. Simultaneously, the decrease of membrane flux was prevented effectively (less than 20% of initial flux). This work strongly confirmed that the membranes coated by activated carbon could significantly relieved membrane fouling, enhance membrane treatment performance, and improve membrane life. Amaral et al. (2016) developed microfiltration membranes coated by superfine powdered activated carbon for drinking water treatment. The coated membranes achieved excellent removal efficiency because superfine powdered activated carbon was more favorable for the adsorption of pollutants due to its smaller particle size compared with conventional activated carbon. Bae et al. (2007) designed activated carbon membrane with carbon whiskers for

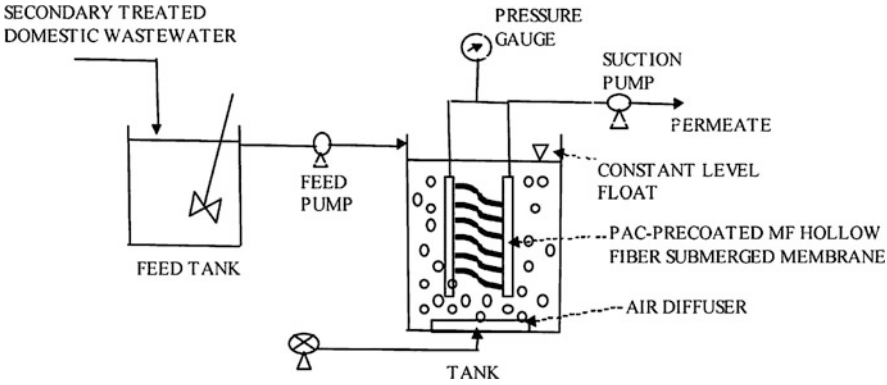

Fig. 4.16 Schematic of membrane hybrid system with pre-coated membrane. (Reprinted with permission of (Thiruvenkatachari et al. 2006))

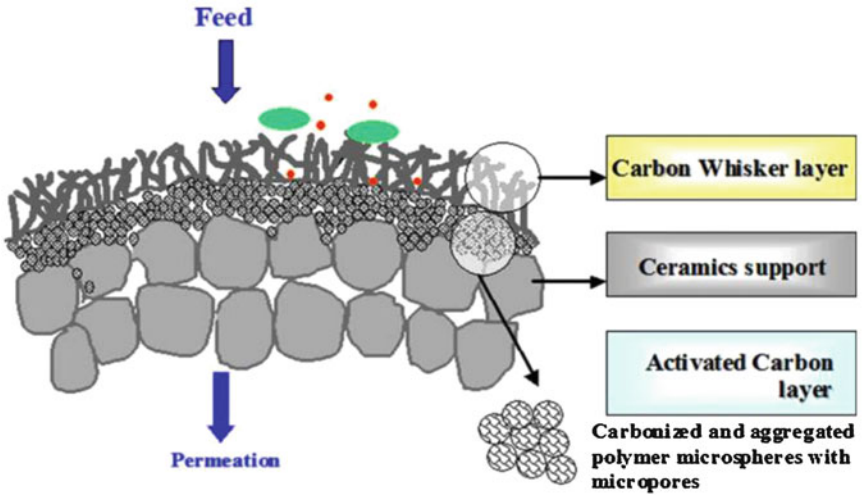

Fig. 4.17 Structure of an activated carbon membrane with carbon whiskers. (Reprinted with permission of (Bae et al. 2007))

wastewater and drinking water treatments. The carbon whiskers on the activated carbon membrane could significantly prevent the deposition and accumulation of particles, extending membrane lifetime (Fig. 4.17).

4.6.2 Support-Free Activated Carbon Membranes

Activated carbon membrane is a novel carbon-based membrane, which not only has excellent thermal stability and chemical stability of inorganic membrane materials

but also has excellent electrical conductivity and rich pore structure of carbon materials. Li et al. (2017a) designed and prepared a support-free activated carbon membrane by mixing activated carbon, binder, pore former, and conductive agent followed by compression modeling and carbonization. The activated carbon membrane realized the integration of the triple function of adsorption/electrocatalysis/membrane separation for deep water purification.

4.6.3 Activated Carbon Hybrid Membranes

In order to further improve membrane performance, activated carbon was also adopted as function material to be mixed in membrane matrix. Aghili et al. (2017) prepared a novel powdered activated carbon mixed-matrix membrane for cheese whey wastewater treatment. This membrane integrated a powdered activated carbon adsorption mechanism with the separation property of the polysulfone membrane, indicating high treatment efficiency for organic matter removal. Ahmad et al. (2018) fabricated high-performance hybrid ceramic/activated carbon symmetric membrane to purify oily wastewater (Fig. 4.18). The hybrid Al_2O_3/activated carbon membrane

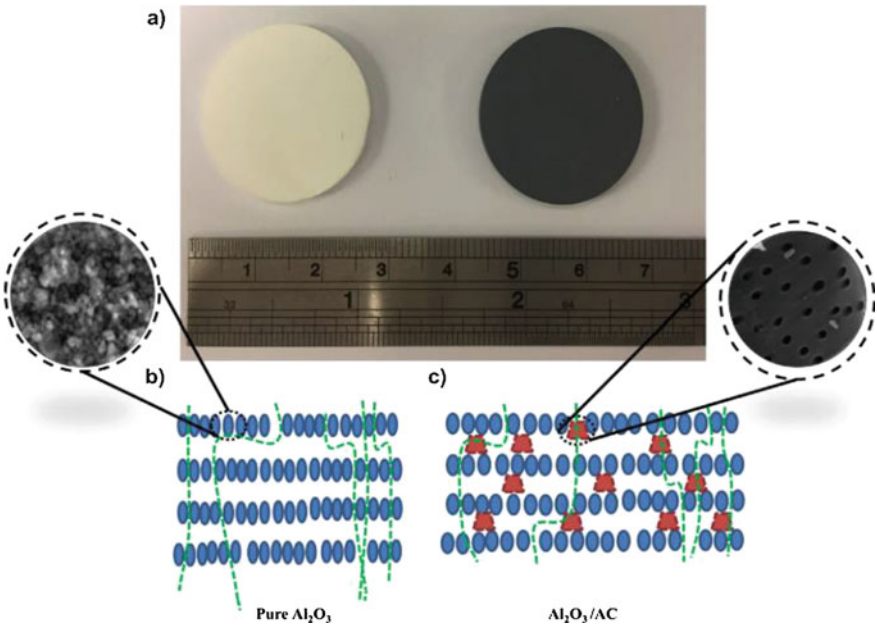

Fig. 4.18 Optical images of (**a**) Al_2O_3 membrane and Al_2O_3/activated carbon hybrid membrane. Schematic illustration of (**b**) Al_2O_3 and (**c**) Al_2O_3/activated carbon hybrid membranes. (The SEM image in (**b**) shows the particle size of the Al_2O_3 after sintering, while the SEM image in (**c**) shows the morphological structure of the activated carbon with highly porous structure and distribution of cylindrical-shaped pores.) (Reprinted with permission of (Ahmad et al. 2018))

possessed complex microchannel–nanochannel networks, which achieved two times higher porosity in comparison with Al_2O_3 membrane. As expected, the oil removal efficiency of the hybrid Al_2O_3/activated carbon membrane could reach 99.02%. On the whole, the development of a cost-effective membrane by doping a cheap material, such as activated carbon, could create a complementary structure, producing strong competitiveness in wastewater treatment.

4.7 Other Carbon Materials Incorporated Membrane

In addition to these carbon materials mentioned above, several other carbon materials such as asphalt were also be adopted to prepare membranes for water treatment. Liang et al. (2016b) used a tubular electrochemically reactive graphite membrane acting as cathode and evidenced the advantages of coupled advanced oxidation process (electro-Fenton reaction) for dynamic filtration. Liu et al. (2017) designed a novel b-cyclodextrin (β-CD)-functionalized g–C_3N_4 composite membrane with the integration of dual function of microfiltration and visible light-driven photocatalytic degradation. The membrane could remove the organic dye by adsorption, microfiltration, and photodegradation. Yvonne (2014) prepared a sulfonated asphalt sodium alginate hybrid membrane.

4.8 Conclusion and Future Prospects

Numerous studies have been performed in membrane technologies with diverse materials for highly efficient water treatment. Among them, carbon materials with outstanding properties have been proven with potential benefits to prepare carbon-based membranes and exhibit superiority over other membrane processes. To further enhance membrane separation performance and antifouling properties, several kinds of carbon-based membrane materials including carbon membranes, carbon nanotube membranes, carbon fiber membranes, activated carbon membranes, graphene-based membranes, etc. are explored for highly efficient water treatment. Various methods including surface modification, operation parameter optimization, and technologies combination are adopted to optimize membrane performance. All these attempts have been proved with fruitful results and make great progress in this field.

Although these carbon-based membrane materials have exhibited promising potential in the field of water treatment, further studies are still required to achieve the commercial application level. The concerned challenges are listed below:

1. More advanced membrane preparation technology should be developed to fabricate high-performance carbon-based membrane materials.
2. The electric assistance might speed up the corrosion of carbon-based membrane materials, shorten the lifetime, and cause secondary pollution. Therefore,

developing the modification technology of existing carbon materials and exploring novel carbon materials with great potential are important to pursue higher separation efficiency and better antifouling performance.
3. Besides electrochemical action, other innovative coupling processes should be further extended.
4. The vast majority of carbon-based membrane materials are carried out in laboratory scale, while much efforts should be paid before the pilot- and industrial-scale applications. In this process, the stability of carbon-based membrane materials needs to be further investigated during long-term operation.

Thus, these issues deserve more attention for membrane researchers. Although it would take a long time and quite great effort to resolve the remaining challenges, it is worth affirming that carbon-based membrane materials have promising potential in dealing with a large variety of industrial wastewater application in the future.

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