

Carbon‑based membrane materials and applications in water and wastewater treatment: a review

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Abstract

Water contamination and freshwater shortage are calling for advanced technologies of water recycling. High performance of membrane separation has been recently obtained using carbon-based membrane materials such as carbon, carbon nanotubes, carbon fber membranes, activated carbon and graphene. Properties of carbon materials improve fouling mitigation, hydrophilicity and permeate quality. Here, we review the fabrication of carbon-based membrane materials and applications in water treatment. The major points are: 1) carbon membranes derived from coal and phenolic resins have been widely used in water treatment. Coal-based carbon membranes used as both electrode and membrane flter display high potential owing to their electrical conductivity. 2) Four types of carbon nanotube membranes are presented, with focus on carbon nanotubes that show high separation performance. 3) Carbon fber membranes show high permeability due to abundant functional groups on the surface. 4) Activated carbon membranes are promising for organic matter removal owing to their high surface area, micro- and macroscopic structure, and various chemical functional groups. (5) Graphene-based membranes with unique laminar pores are very promising.

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

Introduction

The industrial development and population growth have led to serious and sustainable challenge toward the water resources in the twenty-frst century (Ma et al. [2017;](#page-15-0) Menachem and William [2011;](#page-15-1) Crini and Lichtfouse [2018](#page-13-0); Salgot and Folch [2018](#page-16-0)). The World Health Organization estimates that more than 1.2 billion people worldwide have gotten sick or died through drinking contaminated water, and the number is expected to signifcantly grow in the coming years (Maggie [2007](#page-15-2); Montgomery and Elimelech [2007;](#page-15-3) Wilson et al. [2018](#page-17-0)). Hence, in order to reduce the hazards from water pollution to humankind, various technologies and industrial processes for water treatment or purifcation have been developed and applied rapidly in recent years (Hayat

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 \boxtimes Chengwen Song chengwensong@dlmu.edu.cn et al. [2017;](#page-13-1) Jiao et al. [2017](#page-14-0); Pintor et al. [2016;](#page-16-1) Zheng et al. [2015](#page-18-0); Bouabidi et al. [2018](#page-12-0); Bello and Raman [2018](#page-12-1)).

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 (Chowdhury et al. [2018;](#page-13-2) Gin and Noble [2011;](#page-13-3) Lau et al. [2018](#page-14-1); Li et al. [2016a,](#page-14-2) [b](#page-14-3), [c](#page-14-4), [d;](#page-14-5) Madhura et al. [2017](#page-15-4); Rezakazemi et al. [2017;](#page-16-2) Thakur and Voicu [2016](#page-17-1)). 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 including coal, phenolic resin, carbon nanotube, carbon fber, activated carbon, graphene, etc. have been used to develop membranes with optimal structure and performance due to their excellent physicochemical properties (Anand et al. [2018;](#page-12-2) Goh et al. [2016](#page-13-4); Thines et al. [2017](#page-17-2); Madima et al. [2020;](#page-15-5) Wei et al. [2018](#page-17-3)). This review aims to provide an overview on recent developments on carbon-based membrane materials for water treatment. A brief discussion of the existing challenges and their prospects were also considered. This article is an abridged version of the chapter by Li et al. ([2020b](#page-15-6)).

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Carbon membranes

Carbon membranes, as novel porous inorganic membranes, are usually prepared by pyrolysis of carbonaceous materials. Due to their huge advantages including good resistance to high temperature and chemical solvent erosion, developed micron or nanometer porosity, high mechanical strength, easy preparation and high degree of cleanliness, they have shown attractive prospects on gas and liquid separations. The research on carbon membranes originated in the 1960s, which frstly focused on the adsorption and surface difusion process of gas through carbon membranes. Since then, Ash et al. ([1973](#page-12-3)) have made great contribution in this feld and a great deal of subsequent researches have been done (Ismail and David [2001](#page-14-6); Li et al. [2016a,](#page-14-2) [b,](#page-14-3) [c,](#page-14-4) [d;](#page-14-5) Qin et al. [2017](#page-16-3); Wei et al. [2007](#page-17-4)). Recently, with the development of membrane fabrication technology, microporous carbon membranes have been explored and successfully applied for water and wastewater treatments (Bauer et al. [1992](#page-12-4); Li et al. [2016a,](#page-14-2) [b,](#page-14-3) [c,](#page-14-4) [d](#page-14-5); Sun et al. [2018](#page-16-4); Tahri et al. [2013](#page-16-5)). Among them, the carbon membranes prepared with coal or phenolic resin were the mostly studied, thus introduced in the following part.

Coal‑based carbon membranes

Coal, as a kind of natural mixture composed of macromolecular cross-linked polymers and inorganic minerals, is a great material for preparing carbon membranes. In the past 2 decades, a series of systematic investigations on the controlled preparation of carbon membranes derived from coal were carried out by our research group. The pore structure, mechanical strength, physical and chemical properties and electrical conductivity of coal-based carbon membranes were further optimized. As expected, the coal-based carbon membranes showed excellent water treatment performance (Song et al. [2006](#page-16-6)). In addition, some other researchers also use coal as a precursor to prepare carbon membranes and use them in water treatment. Low-cost carbon membranes prepared from fy-ash of burning coal have been used for the clarifcation of centrifuged kiwifruit juice (Qin et al. [2015b](#page-16-7)). Tubular carbon microfltration membrane was prepared by mixing coal powder with phenolic resin solution and organic additives and was successfully applied to the retention of dye wastewater (Tahri et al. [2013\)](#page-16-5). However, during treatment process, 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 tackle this problem, our group utilized the electrical conductivity of coal-based carbon membranes and designed a coupling system which employed coal-based carbon membranes both as the flter and anode. Thus signifcant improvement on removal efficiency and antifouling ability were obtained due to the electrochemical oxidation synthetic efect (Fig. [1](#page-1-0)). This system not only displayed excellent removal efficiency for organic pollutants (such as oil droplets) larger than the membrane pores (Li et al. [2016a](#page-14-2), [b](#page-14-3), [c](#page-14-4), [d\)](#page-14-5), but also demonstrated great potential on those pollutants with a smaller molecule size than the membrane pore size including dyes, bisphenol A, phenol and antibiotics (Sun et al. [2018;](#page-16-4) Tao et al. [2017a](#page-16-8); Yin et al. [2016](#page-17-5)). Moreover, microorganisms such as microalgae and Vibrio cholerae were also effectively removed (Tao et al. [2017b\)](#page-17-6). Compared with other membrane processes such as ultrafltration, nanofltration and reverse osmosis, this technology possessed obvious advantages on processing capacity and energy consumption.

Although the coupling system has been proved to be efective 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 signifcant breakthrough in this feld. Yang et al. ([2011](#page-17-7)) presented the design of a novel electrocatalytic membrane reactor by loading $TiO₂$ electrocatalyst on carbon membrane surface by a sol–gel approach to enhance electron transfer and membrane permeability. In this operation process, once the membrane anode was electrifed, it not only electrochemically decomposed $H₂O$ into gas and liquid microflows to avoid membrane fouling, but also generated reactive intermediates which could indirectly decompose the organic foulants into $CO₂$ and $H₂O$ or biodegradable products to realize the self-cleaning function of the electrocatalytic membrane. Further, this $TiO₂/carbon$ membrane showed great performance on tetracycline removal with 100% tetracycline and 87.8% chemical oxygen demand (COD) removal rate achieved (Liu et al. 2016). Similarly, Sn–SnO₂ was coated on carbon membrane to enhance its treatment performance, The tetracycline removal rate of $Sb-SnO₂/carbon$ membrane could achieve 96.5% after 6 h operation compared to 72.8% for carbon membrane (Liu et al. [2017a,](#page-15-8) [b](#page-15-9)). Besides, the Bi–SnO₂/carbon electrocatalytic membrane was fabricated via a simple electrochemical reduction and hydrothermal method by Wang et al. (2018) (2018) (2018) . The Bi–SnO₂/carbon 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, fow rate of 1.4 mL/min and *E. coli* initial concentration of 1.0×10^4 CFU/mL, owing to the synergistic efect of the membrane separation and electrocatalytic oxidation. In order to further enhance the electrocatalytic activity of the membrane, a facile dynamic electrodeposition method was developed by Li et al. $(2020a)$ $(2020a)$, by which CuO nanoparticles were uniformly deposited on both the surface and pore walls of a coal-based carbon membrane. The prepared dynamic electrodeposited CuO/carbon membrane exhibited superior removal ability with Rhodamine B, COD and total organic carbon (TOC) removal efficiency of 99.96%, 71.82% and 64.29%, respectively, which were 18.7–20.1 and 1.5–1.8 times higher than that of the original carbon membrane and the conventional electrodeposited CuO/carbon membrane.

In addition, a novel integration system coupled coal-based carbon membrane with sulfate radicals-based advanced oxidation processes was proved to have great wastewater treatment potential. Under the optimal condition, the integrated system achieved 100% phenol removal efficiency which attributed to the co-existence of radical and nonradical mechanisms (Fan et al. [2019](#page-13-5)).

Phenolic resin‑based carbon membranes

Phenolic resins are applied as another precursors of carbon membranes due to their low cost, thermosetting property and high carbon yield (Muylaert et al. [2012\)](#page-15-11). Several scholars have successfully prepared phenolic resins-based carbon membrane for water and wastewater treatment. A method for preparing symmetric carbon membrane was presented by Wei et al. ([2012\)](#page-17-9). The carbon membrane was synthesized via sol–gel synthesis followed by super-critical drying with $CO₂$ at 50 °C and 10 MPa. Due to its mesoporous property, the pure water flux could achieve at 13.4 L m⁻² h^{-1} and the molecular weight cutoff was about 2000. Tubular carbon membranes on an ultrafltration substrate were prepared by thermosetting phenolic resin and carbon black (Tahri et al. 2016) and were applied efficiently to the treatment of industrial dyeing effluent. On the other hand, the fabrication and application of asymmetric tubular carbon membranes were studied. A simple approach for preparing carbon membranes was presented by Qin et al. [\(2015a\)](#page-16-10) with macroporous ceramic membrane as substrate and carbon layer loaded via sol–gel polymerization of resorcinol and formaldehyde. Owing to its uniform pores and high mesopore volume, the pure water fux of the obtained carbon membrane was as high as $167 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$. Song et al. [\(2017\)](#page-16-11) developed carbon alumina mixed matrix membranes by impregnating phenolic resin in porous alumina matrix via a vacuum-assisted method. The results showed that membranes with high water fuxes and salt rejections could be easily tailored. Based on this, the effects of different substrates and coating conditions on the formation of carbon/ ceramic substrate for desalination were studied (Song et al. [2018](#page-16-12)). Besides, the preparation and separation performance assessment of carbon membranes derived from phenolic resin by a vacuum-assisted method and carbonization in an inert atmosphere were shown (Abd et al. [2017](#page-12-5)). The study of phenolic resin-based carbon membrane for phenol and phosphoric acid removal by Zhao et al. ([2018](#page-18-1)) showed that the maximum removal rates could reach 81.9% for phenol and 55.3% for phosphoric acid. Due to the excellent electrical conductivity of carbon membranes, studies on coal-based carbon membranes above have proved that coupling electrocatalysis and membrane separation technology was one of the feasible ways to enhance the treatment performance. Similarly, phenolic resin-based carbon membrane was also used as electrocatalytic electrode for water and wastewater treatment. Study result showed that $TiO₂$ phenolic resin/ activated carbon carbon-based membranes had the superb electrochemical activity and catalytic degradation efficiency

with the COD and phenol removal rates of 86.5% and 94.6% (Hui et al. [2020](#page-14-7)).

Carbon nanotube membranes

Carbon nanotubes, as an important kind of carbon materials, with remarkable electrical, thermal, mechanical and optical properties, carbon nanotubes have been widely used in sensor, super-capacitor, lithium-ion battery, etc. (Apul and Karanfl [2015;](#page-12-6) Gupta et al. [2013](#page-13-6); Patino et al. [2015](#page-15-12); Ren et al. [2011](#page-16-13); Yu et al. [2014](#page-18-2)). Carbon nanotubes were frstly discovered by Ijima ([1991](#page-14-8)). Soon after, researchers observed ultrahigh water fow rates in carbon nanotubes and this discovery produced great expectation that carbon nanotubes could be used as an ideal material for water treatment (Ahn et al. [2012;](#page-12-7) Lee et al. [2011;](#page-14-9) Whitby and Quirk [2007](#page-17-10)). The concept of carbon nanotube membrane was introduced by Li and Richard [\(2000](#page-14-10)) when they studied the mass transfer phenomenon in single-walled carbon nanotubes. Recently, carbon nanotubes membranes are getting more and more attention in water treatment feld. Carbon

(A)Vertically aligned carbon nanotubes membrane

nanotubes membranes are usually classifed into vertically aligned carbon nanotubes membranes, horizontally aligned carbon nanotubes membranes, mixed matrix carbon nanotubes membranes and electrochemical carbon nanotubes membranes (as shown in Fig. [2\)](#page-3-0).

Vertically‑aligned carbon nanotubes membranes

The vertically aligned carbon nanotube membrane was frstly constructed by Bruce et al. [\(2004](#page-12-8)) with a high water fux. After that, the membranes prepared by Baek et al. ([2014](#page-12-9)) and Holt et al. [\(2004\)](#page-13-7) demonstrated that the water fux was most three times higher than a typical ultrafltration membrane. This was mainly owing to the efect of the compact nanotube forest and short nanochannel length. In addition, vertically aligned carbon nanotube membranes also exhibited certain antimicrobial and antifouling capacities (Ihsanullah et al. [2016](#page-14-11); Lee et al. [2015;](#page-14-12) Mainak et al. [2005](#page-15-13); Matsumoto et al. [2017;](#page-15-14) Park et al. [2014;](#page-15-15) Vijwani et al. [2018](#page-17-11); Zhao et al. [2013a](#page-18-3), [b\)](#page-18-4). Excellent oil removal efficiency was proved by using prepared membranes for oil–water mixture treatment (Hsieh et al. [2016;](#page-13-8) Lee and Baik [2010](#page-14-13)). A key

(B) Horizontally aligned carbon nanotubes membrane

(C)Carbon nanotubes mixed matrix membrane

Clean water

(D) Electrochemical carbon nanotubes filter

Fig. 2 Mechanism of water passing through the four types of carbon nanotubes membranes: **a** vertically aligned carbon nanotubes membrane, **b** horizontally aligned carbon nanotubes membrane which are randomly arranged horizontally on a porous support layer, **c** mixed

nanotubes

matrix carbon nanotubes membrane which are directly doped into the polymer membranes by interfacial polymerization or phase inversion, **d** electrochemical carbon nanotubes membrane

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\)](#page-12-10). Instead of conventional preparation methods, Wu et al. [\(2014](#page-17-12)) utilized an electric feld to obtain vertically aligned carbon nanotubes membranes. Electro-casting allowed multi-walled carbon nanotubes to grow vertically and disperse more evenly. However, complex manufacturing techniques were still major obstacle to make these membranes suitable for large-scale applications (Ihsanullah [2019\)](#page-14-14).

Horizontally‑aligned carbon nanotubes membranes

In addition to vertically aligned pattern, carbon nanotubes can aggregate with each other by Van der Waals interactions to form horizontally aligned carbon nanotubes membranes (Ihsanullah [2019\)](#page-14-14). Due to the disordered arrangement of functionalized carbon nanotubes, the horizontally aligned carbon nanotubes membranes can provide rich porous structure and large specifc surface area (Sears et al. [2010](#page-16-14)), which makes them possess high water fux, high adsorption capacity to natural organic matter (Yang et al. [2013](#page-17-13)) and strong antimicrobial actions (Ihsanullah et al. [2015;](#page-14-15) Kang et al. [2007\)](#page-14-16). Li et al. [\(2015\)](#page-14-17) found that a "slanted carbon nanotubes membrane" exhibited higher water fux than a typical vertically aligned carbon nanotubes membrane, as this kind of art structure could obviously lower the energy barrier for flling water into the carbon nanotubes. Brady et al. [\(2008\)](#page-12-11) reported that the horizontally aligned single-walled carbon nanotubes membrane displayed high removal rate for the virus MS₂ bacteriophage. Horizontally aligned carbon nanotubes membranes were also applied to direct contact membrane distillation, showing high water fux and good desalination ability (Dumée et al. [2010](#page-13-9)). Subsequently, the membrane was modifed by two chemical ways; the resultant membrane had a larger contact angle (140° compared with 125[°]) that further improved its treatment performance (Dumée et al. [2011](#page-13-10)).

However, carbon nanotubes usually tend to aggregate when they dispersed in a polymer matrix or solvent. Therefore, it was difficult to prepare a uniform dispersion. For this reason, several surfactants such as Triton-X100 and sodium lauryl sulfate were adopted to improve the dispersion of carbon nanotubes in aqueous solution (Wu et al. [2010a,](#page-17-14) [b](#page-17-15), [c](#page-17-16)). Besides, chemical functionalization was regarded as another efective way to increase the hydrophilicity and stability of carbon nanotubes suspensions (Yang et al. [2013](#page-17-13)). Some researchers covalently grafted functional groups including amines, fuorine and sulfhydryl groups onto carbon nanotubes to help them disperse in horizontally aligned carbon nanotubes membranes (Ansón-Casaos et al. [2010](#page-12-12); Darryl et al. [2010](#page-13-11)).

Mixed matrix‑carbon nanotubes membranes

Compared with the above two types of membranes, mixed matrix membranes are easier to be commercialized for their simple preparation procedures. For preparing mixed matrixcarbon nanotubes membranes, functional carbon nanotubes are generally added into polymeric membranes by several synthesis techniques (Ali et al. [2019;](#page-12-10) Ihsanullah [2019](#page-14-14)) including phase inversion (Brunet et al. [2008](#page-12-13); Choi et al. [2006](#page-13-12); Majeed et al. [2012](#page-15-16)), interfacial polymerization (Kim et al. [2014;](#page-14-18) Shen et al. [2013\)](#page-16-15), solution mixing (Ahmed et al. [2013\)](#page-12-14), spray-assisted layer-by-layer (Liu et al. [2013](#page-15-17)), polymer grafting (Shawky et al. [2011](#page-16-16)), polymerization (Zarrabi et al. [2016;](#page-18-5) Zhao et al. [2014](#page-18-6)) and colloidal precipitation (Ho et al. [2017\)](#page-13-13). The prepared membranes exhibit excellent properties for ultrafltration (Choi et al. [2006;](#page-13-12) Majeed et al. [2012](#page-15-16)), nanofltration (Shen et al. [2013](#page-16-15); Wu et al. [2010a\)](#page-17-14), microfltration (Madaeni et al. [2013;](#page-15-18) Medina-Gonzalez and Remigy [2011](#page-15-19)), reverse osmosis (Kim et al. [2014](#page-14-18)) and forward osmosis (Amini et al. [2013](#page-12-15)) applications. Researches about the membrane performance of mixed matrix nanotubes membranes are listed in Table [1](#page-5-0).

Electrochemical carbon nanotubes membranes

Electrochemical carbon nanotubes membrane for wastewater treatment is a novel technique, in which the membranes are used both as a flter for separation and electrodes for electrochemical degradation (Ahmed et al. [2016](#page-12-16); de Lannoy et al. [2012;](#page-13-14) Ho et al. [2018;](#page-13-15) Lalia et al. [2015;](#page-14-19) Yi et al. [2018\)](#page-17-17). Thus, they exhibit great potential on wastewater treatment due to high degradation efficiency, low energy consumption and simple operation process (Bakr and Rahaman [2016](#page-12-17), [2017;](#page-12-18) Liu et al. [2017a,](#page-15-8) [b](#page-15-9); Motoc et al. [2013](#page-15-20)). Besides, by transferring electrons directly through the surface of the membrane electrode, the solute transfer restriction of the conventional batch electrochemical process is overcome. A novel carbon nanotubes-based membrane with a sandwich-like structure was prepared by Wei et al. ([2017\)](#page-17-18). Low concentration of microcys- tin-LR (0.5 mg/L) was removed economically and efficiently $(>99.8\%)$. Besides, the selectivity of iron ions $(>98\%)$ was obtained by applying an electric feld to the membranes (Duan et al. [2017](#page-13-16); Gao et al. [2017\)](#page-13-17) and metoprolol degradation rate of 97% was reached by using a nanoparticulate zero-valent iron modifed multi-walled carbon nanotubes membrane under the applied voltage of 1 V (Yanez et al. [2017](#page-17-19)).

Graphene‑based membranes

Graphene, materials, consisting of a compact accumulation of sp2 hybrid carbon atoms, were reported for the frst time by Novoselov et al. ([2004](#page-15-21)). Since then, graphene and

 \overline{a}

Table 1

graphene oxide have been extensively applied to construct novel membranes with laminar pores. These materials have been also used as blender to improve the hydrophilicity, surface charges and antifouling ability of the polymeric membranes (Hebbar et al. [2017;](#page-13-23) Jilani et al. [2018](#page-14-22)).

Support‑free graphene membranes

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](#page-13-24)). Previous research suggested that highly permeable single-layered graphene membranes could achieve by controlling the pore size and functional groups (Cohen-Tanugi and Grossman [2012\)](#page-13-24). O'Hern et al. [\(2014\)](#page-15-26) reported their works on the controlled highdensity subnanometer pores in graphene membranes which allowed the transport of salt but rejected larger organic molecule. The efects of pressure and wall interaction on the water transport through graphene membranes were carried out by molecular dynamic simulation (Shahbabaei et al. [2017](#page-16-21)). It showed that the water fux was much high owing to strong hydrogen bonds. Besides, due to excellent selectivity, membranes exhibit great potential in selective ions transportation and separation. Kabiri et al. [\(2016\)](#page-14-23) synthesized a thiol-functionalized graphene membrane with a unique three-dimensional porous structure to remove mercury ions $(Hg²⁺)$ from water. The results indicated that the removal efficiency reached almost 100% for removing low (4 mg/L) and high (120 mg/L) concentration of Hg^{2+} .

Graphene oxide membranes

Graphene oxide is a reforming form of graphene in which oxygen and hydrogen atoms are bonded with carbon atoms (Hu and Mi [2013](#page-13-25)). It is usually obtained by oxidizing graphite with a strong acid or oxidant. 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\)](#page-16-22). Several researches of graphene oxide membranes preparation and its application have been made in recent decades. Graphene oxide nanosheets were coated on a highly porous nanofbrous mat directly using vacuum suction method by Wang et al. ([2016](#page-17-24)). The obtained graphene oxide layer exhibits "ideal" pathways (hydrophobic nanochannels) for water molecule (Fig. [3](#page-8-0)). As a result, water fux under an extremely low external pressure (0.1 bar) signifcantly increased and high removal performance of 100% rejection of Congo red and 56.7% rejection of $Na₂SO₄$ obtained. Besides, Nair et al. ([2012](#page-15-27)) studied the water mobility in nanochannels between graphene oxide tablets under diferent conditions. It showed that the layer **Fig. 3** Graphene oxide membrane for water transport. In the treatment process, water molecules frstly arrived at the hydrophilic "gate" (space between the two adjacent graphene oxide nanosheets or defects of graphene oxide nanosheets) and aggregated at this place. After passing through the "gate," water molecules slipped through the hydrophobic nanochannel at a high speed with low or no friction

spacing between the original graphene oxide membrane and the stacked graphene oxide membrane was about 0.6 nm in the dry condition. Because of the difusion of water molecules to graphene oxide layer, the increased interlayer spacing between graphene oxide membranes resulted in water molecules obtained high mobility. However, when the graphene oxide membrane was immersed in an ionic solution, the increased gap by the hydration of the sheet cannot repel K^+ and Na^+ ions and make the membrane inappropriate for desalination applications (Joshi et al. [2014](#page-14-25)). Addressing to this issue, graphene oxide (graphene oxide–COOH) was functionalized with glycine and carboxylation for preparing membrane to achieve high salt rejection efficiency (Yuan et al. [2017\)](#page-18-10). In addition to the layer spacing, it was found that the morphological characteristics of graphene oxide membranes, such as corrugation, could improve the separation performance (Qiu et al. [2011](#page-16-24)). Similar to the study of graphene oxide membrane in ions transport, Chang et al. [\(2017](#page-12-20)) reported that carboxylation could increase the hydrophilicity of graphene oxide membrane by which improved the efficiency of dye removal. Such improvement was potentially attributed to surface charge density. Nowadays, graphene oxide membranes were also applied for oil removal from wastewater. Zhao et al. [\(2016\)](#page-18-11) intercalated palygorskite nanorods into adjacent graphene oxide nanosheets before assembling into laminate structures, the prepared graphene oxide membranes showed excellent anti-oil performance in the separation process of water-containing oil emulsion.

Graphene oxide composite membranes

Although graphene oxide membranes with a good capability can be prepared by simple methods, these membranes would be trapped in the use of pressure-driven systems. Liu et al. ([2015\)](#page-15-29) found that the composite membrane prepared by adding graphene oxide to polysulfone displayed superior pressure-resisted ability, mechanical strength and water permeability. Similarly, research conducted by Lai et al. [\(2016](#page-14-26)) demonstrated that water fux and salt removal were improved by integrating graphene oxide in polyamide membrane. Kochameshki et al. [\(2017](#page-14-27)) synthesized a polysulfone nanocomposite membrane modifed with graphene grafted with diallyldimethylammonium chloride. The results showed that the water fux increased to about 450 L/m^2 h and the heavy metal ion rejection rate increased to 86.68% (Cu^{2+}) and 88.68% (Cd^{2+}). Besides, Choi et al. ([2013](#page-13-27)) fabricated a dual-action barrier coating layer of graphene oxide on the surface of polyamide reverse osmosis membrane. The antifouling tests indicated that the graphene oxide coating layer could increase the surface hydrophilicity and decrease the surface roughness, which promoted the antifouling performance against a protein foulant. There were some researchers reporting the improvement in the chlorine resistance of the polyamide membranes incorporated with graphene oxide (Safarpour et al. [2015](#page-16-25)). In their opinion, the chemically stable graphene oxide plate embedded in the polyamide layer and acted a barrier layer, protecting the polyamide from chlorine erosion (Choi et al. [2013](#page-13-27)). The researchers also identifed that the separation performance of graphene oxide mixed membrane used for dyes removal were all higher than that of their counterparts (Qiu et al. [2011\)](#page-16-24). Due to superior separation characteristic, graphene oxidedoped polymer membranes were also applied on oil–water separation (Zhang et al. [2017](#page-18-12)) and intricate wastewater treatment (Sun et al. [2015](#page-16-26)). Compared to modify graphene oxide with active substances, graphene oxide hybrid membranes by adding graphene oxide into polymer membranes achieves more signifcant advantages on improved water fux, mechanical stability and fouling resistance. There

is no doubt that graphene oxide hybrid membranes will provide us the new insight into the optimization graphenebased membranes.

Carbon fber membranes

Support‑free carbon fber membranes

Since Shimpei et al. [\(1986](#page-16-27)) accidentally found that carbon fbers facilitated microbial attachment and possessed excellent adsorption capacity for pollutants, the research works focused on carbon fbers for water treatment were widely carried out (Manawi et al. [2018;](#page-15-30) Ahmed et al. [2015;](#page-12-21) Xu and Luo [2012](#page-17-26)). The support-free carbon fber membranes are generally obtained by forming carbon fber precursors into membrane shape, and then stabilized and carbonized via thermal treatment. Rika et al. [\(2017\)](#page-16-28) prepared carbon nanofber membranes by electrospinning followed by carbonization. Results showed that the adsorption capacity, permeability and adsorption kinetics of the carbon nanofber membranes were about 10 times, 6 times and 2 times higher than that of the traditional activated carbon membrane, respectively. However, such carbon fber membranes usually sufered from serious membrane fouling, limiting their application.

Carbon fber hybrid membranes

In order to expand the application of carbon fber membrane in water treatment, researchers have developed a variety of carbon fber hybrid membranes, which combined the advantages of carbon fber and membrane technology. Yang and Tsai ([2008,](#page-17-27) [2009\)](#page-17-28) prepared carbon fbers/carbon/alumina tubular composite membrane and applied it in a crossfow electrocoagulation/electrofltration 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. $(2013a, b)$ $(2013a, b)$ $(2013a, b)$ reported their works on domestic sewage treatment using biological carbon fber membrane for efectively intercept sludge and most organic matter. Yue et al. [\(2018\)](#page-18-13) fabricated layered porous dynamic separation membranes containing primary and secondary nanostructures by in situ growth of ZnO nanowires on carbon fbers. The membrane could switch wettability between super-hydrophilicity and high hydrophobicity by simply annealing alternatively in air and vacuum environment (Fig. [4](#page-10-0)), and indicated more than 98% separation efficiency in dewater and deoiling modes.

The composite membranes using carbon fber cloth as the substrate

Taking advantage of the good mechanical properties of carbon fber cloth, various functional materials have been loaded on it to fabricate the composite membranes for waste-water treatment. Li et al. [\(2016a](#page-14-2), [b,](#page-14-3) [c,](#page-14-4) [d](#page-14-5)) successfully prepared a catalytic cathode membrane by loading Pd-reduced graphene oxide– $CoFe₂O₄$ catalyst on carbon fiber cloth and used it in microbial fuel cell/membrane bioreactor coupling system for wastewater treatment. Xiao et al. [\(2017](#page-17-29)) obtained carbon fiber/ C_3N_4 cloth by a dip-coating and thermal condensation method, the composite membrane possessed excellent fexibility and strong visible-light absorption for the degradation of fowing wastewater. To further improve the treatment efficiency, Shen et al. (2018) (2018) (2018) inserted TiO₂ between C_3N_4 and carbon fiber, the modified membrane showed enhanced photocatalytic activity for degrading various organic pollutants in comparison with carbon fiber/ C_3N_4 cloth.

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;](#page-12-22) Gopinath and Kadirvelu [2018](#page-13-28); Danish and Ahmad [2018](#page-13-29)). Adham et al. [\(2016\)](#page-12-23) successfully applied it into hollow fiber ultrafiltration system to investigate the infuence of carbon particle size on organic matter adsorption and wastewater treatment performance. Then, Jacangelo et al. ([1995](#page-14-30)) 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](#page-12-24); Jia et al. [2014](#page-14-31)).

Activated carbon‑coated membranes

Activated carbon can be coated on membranes by coating method and dipping method et al. to enhance membrane separation performance while removing contaminants from wastewater. Thiruvenkatachari et al. ([2006\)](#page-17-30) prepared activated carbon pre-coated microfltration membrane with wood-based, coal-based and coconut shellbased activated carbon for wastewater treatment. After 8 h 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 **Fig. 4** ZnO–carbon fber dynamic membrane for oil/ water mixtures treatment. The as-fabricated membrane is capable of switching into water-removing model with super-hydrophilicity via simply annealing under air environment and switching into oil-removing model with high hydrophobicity via simply annealing under vacuum environment

coconut shell-based activated carbon-coated membrane, which were all higher than that of non-pre-coated membrane. Simultaneously, the decrease in membrane fux was prevented efectively (less than 20% of initial fux). But in the absence of long-term experiments, this work was too early to elucidate the underlying mechanisms of the efects observed in it. Meanwhile, the optimal amount of particle activated carbon had not been determined, so the stability of the membrane remained to be investigated. Amaral et al. ([2016](#page-12-25)) developed microfltration membranes coated by superfne 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. However, comparing with the original membrane, the membrane coated by superfne powdered activated carbon caused fux loss, and the fux reduction caused by small particles were larger than the large particles. Bae et al. [\(2007\)](#page-12-26) designed activated carbon membrane with carbon whiskers for wastewater and drinking water treatments. The carbon whiskers on the activated carbon membrane could signifcantly prevent the deposition and accumulation of particles, extending membrane lifetime (Fig. [5](#page-10-1)). The adsorption capacity of activated carbon membrane with carbon whiskers for phenol was comparable to that of industrial activated carbon and it had better stability.

Support‑free activated carbon membranes

Support-free activated carbon membrane is a novel carbon-based membrane, which not only has excellent thermal stability and chemical stability of inorganic membrane

Fig. 5 Structure of an activated carbon membrane with carbon whiskers. Carbon whiskers on membrane surface were used for preventing the deposition and accumulation of particles. Activated carbon layer below carbon whiskers was assembled for the adsorption of dissolved organics

materials, but also owns excellent electrical conductivity and rich pore structure of carbon materials. Li et al. ([2017\)](#page-14-32) 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. It could adjust its multilevel channel structure, regulate its adsorption and membrane separation performance, and simultaneously support the catalyst on the carbon membrane substrate coupling with electrochemical technology. When the triple-functional carbon membrane material was used for deep purifcation of the source water and advanced treatment of the wastewater, the developed pore structure could efectively adsorb pollutants such as organic molecules, microorganisms and heavy metals. The membrane surfaces and pores were enriched. At the same time, combining with the function of membrane separation and electrocatalytic oxidation, the pollutants were separated and degraded to achieve the efect of deep purifcation. This greatly simplifed the advanced treatment process of water, and had broad the application prospects.

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](#page-12-27)). Ahmad et al. ([2018](#page-12-28)) fabricated high-performance hybrid ceramic/ activated carbon symmetric membrane to purify oily waste-water (Fig. [6\)](#page-11-0). The hybrid membrane possessed complex micro-nano-channel networks, which achieved two times higher porosity in comparison with Al_2O_3 membrane. As expected, the oil removal efficiency of the hybrid membrane could reach 99.02%. On the whole, the development of a cost-efective membrane by doping a cheap material, such as activated carbon, could create a complementary structure, producing strong competitiveness in wastewater treatment.

Conclusion

Carbon-based membrane materials including carbon membranes, carbon nanotube membranes, carbon fber membranes, activated carbon membranes, graphene-based membranes, etc. are explored for highly efficient water and wastewater treatment. Various methods including surface modifcation, 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 feld. Although these carbon-based membrane materials have exhibited promising potential in the feld 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, shortening the lifetime and cause secondary pollution. Therefore, developing the modifcation technology of existing carbon materials, exploring novel carbon materials with great potential are important to pursue higher separation efficiency and better antifouling performance.
- 3. 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-

Fig. 6 Al_2O_3 and Al_2O_3 /activated carbon hybrid membranes. Incorporating AC in the alumina matrix could pose porous structure of the membrane matrix, resulting in extra micro-channels for water to pass

through the membrane. Both the filtration and adsorption efficiencies were increased

based membrane materials needs to be further investigated during long-term operation.

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 water and wastewater application in the future.

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