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Review

Sustainable carbonaceous materials-based catalytic membranes for organic wastewater treatment: Progress and prospects

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ABSTRACT

Catalytic membrane, as a cutting-edge hybrid technology, is promising for organic wastewater treatment not only because of the excellent removal efficiency for various organic pollutants, but also because of the mitigation of membrane fouling. However, some challenges persist, including the relatively high fabrication costs of membranes, the high possibility of metal ions leaching from membrane structures, and the poor renewability of synthetic materials, which significantly restrict more widespread application. To address these issues, carbonaceous materials (e.g., biochar, activated carbon, carbon nanotubes, graphene, graphene oxide) are renewable and environmentally friendly materials that inherently have large surface areas, high porosity, and tuneable surface functional groups that can be employed as excellent alternatives in catalytic membranes. In this review, various methods (e.g., blending, in-situ growth, interfacial polymerization, and layer-by-layer assembly) for the fabrication of carbonaceous materials-based membranes are comprehensively summarized and discussed. Subsequently, the integration of catalytic membranes in different processes, whether individually (e.g., photocatalytic process, advanced oxidation process, and electrocatalytic process) or hybridized (e.g., photoelectrochemical process, photo-assisted advanced oxidation process), is assessed. In addition, various carbonaceous materials-based catalytic membranes implemented for the remediation of wastewater are critically discussed. Furthermore, the existing challenges are described, and further research recommendations are proposed. This review is expected to be beneficial for advancing the development of carbonaceous materials-based catalytic membranes for practical decontamination of organic wastewater.

1. Introduction

Increasing attention has been dedicated to innovating and augmenting water treatment due to the escalating water demands, particularly as the water infrastructure established in the 20th century nears expiration [1]. Correspondingly, various technologies have been developed, including biological methods [2], photodegradation process [3], electrochemical process [4], and membrane technologies [5]. Among them, membrane technologies are promising because of their simple operation, high efficiency, environmental friendliness, and ease of scale-up [6]. Nevertheless, membrane fouling remains the main technical bottleneck, not only decreasing water permeability but also increasing operational costs (e.g., membrane replacement, pre- and post-treatments) [7]. To address this, different approaches have been attempted, such as surface modification of the membrane (e.g., hydrophilicity [8], surface charge [9]), incorporation of nanostructured materials [10], as well as the fabrication of catalytic membranes [11,12].

Recently, catalytic membranes have gained prominence as excellent candidates for mitigating membrane fouling [13], and additionally are effective for immobilizing catalysts to solve the problem of needing to capture the nanomaterials employed [14]. Specifically, the catalytic membrane consists of catalysts embedded in the membrane matrix. During membrane-filtration, catalytic reactions generate active species to degrade the organic pollutants within the membrane and thus mitigate membrane fouling [15]. Currently, these reactions mainly involve advanced oxidation (e.g., persulfate-based system [16]), photocatalysis [17], electrochemical catalysis [18], and combined processes (e.g., photo/electro-catalysis [19]). Hitherto, numerous catalytic membranes involving catalysts such as CuO [20], Co₃O₄/carbon nanotubes [21], perovskite [22], covalent organic framework [23], and layered metal oxides [24] have been explored and demonstrated to be effective in alleviating membrane fouling during organic wastewater treatment. Although metal-based catalysts confer high catalytic efficiency for organic pollutants, the high synthesis costs, the high potential for

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secondary pollution (e.g., metal ions leaching), and the unsustainability of such synthetic materials hinder widespread implementation.

To exploit the benefits of catalytic membranes, renewable carbonaceous materials represent excellent alternatives due to their lower cost and lower environmental impact [25]. Carbonaceous materials can largely be classified into biochar, activated carbon, carbon nanotube, graphene and graphene oxide, fullerene, carbon fiber, graphite and carbon quantum dot [26]. Such materials have features that are advantageous for membrane processes, including relatively high specific surface areas, abundant surface functional groups, and tuneable properties that allow them to be used not only as catalysts but also for supporting metal-based materials [27]. For instance, the introduction of corn cob biochar [28], Fe/N co-doping biochar [29], and silver-graphene oxide [30] into the membrane structures have been proven to significantly improve the removal efficiency of pollutants from wastewater. In view of the excellent properties of carbonaceous materials-based catalytic membranes (e.g., high porosity, outstanding water permeability, anti-fouling, high removal efficiency of pollutants) and thus the potential for more widespread implementation, it is timely for a critical review, which motivated this study.

To date, extensive research has positioned catalytic membranes as promising for efficient organic wastewater treatment. A few reviews have summarized related aspects, such as photo-catalytic membrane reactors [31], metal-organic framework-based membranes for water purification [32], catalytic membrane-based oxidation-filtration systems [13], and membrane-based separation of organic pollutants from wastewater [33]. The current review represents a distinct and timely contribution to this knowledge base by focusing on the potential of carbonaceous materials-based catalytic membranes for organic wastewater treatment. In particular, carbonaceous materials as catalysts embedded in membranes have been demonstrated to be attractive in terms of highly tuneable properties, sustainability and environmental friendliness. In this review, the research trends of carbonaceous materials-based catalytic membranes in organic wastewater treatment are investigated via scientometric analysis. Subsequently, various methods (e.g., blending, *in-situ* growth, interfacial polymerization) for the fabrication of carbonaceous materials-based catalytic membranes are critically summarized. The applications of catalytic membranes (e.g., carbon nanotubes (CNTs), graphene, biochar) for the degradation of organic pollutants are discussed, and the underlying mechanisms are comprehensively overviewed. Finally, the future perspectives of this type of membrane for wastewater treatment are provided, and the recommendations for future research are proposed.

2. Scientometric analysis

The Web of Science core database was used to analyze the published articles related to the topic of carbonaceous materials-based catalytic membranes for organic wastewater treatment. To limit the scope for the review, a preliminary survey of the literature was conducted on September 25, 2024 using several sets of keywords. Table 1 shows that the number of articles between year 2000 and 2024 for each of the four sets of keywords is on the order of million, which is too excessive to capture in a review. The intersection of the four sets (i.e., using the 'AND' operator) gives 1,377 articles, which defines the boundary conditions for the current review and are further analyzed in Fig. 1. This review investigates the potential of carbonaceous materials-based catalytic membranes in organic wastewater treatment, including their preparation methods, application processes, advantages, and existing challenges, as well as providing insights into future developments.

Fig. 1a shows that the number of published articles and citations per year have continuously increased since 2000, while Fig. 1b indicates that the three top categories of the publications are multidisciplinary materials science, physical chemistry, and chemical engineering. Regarding dominant keywords in the publications, the top three are degradation, performance and removal (Fig. 1c), indicating focuses on

Table 1

Sets of the keywords used to retrieve articles from Web of Science (Statistics as of September 25, 2024).

	Keywords	Number of articles
#1	TI= (*cataly* or degrad* or decom* or oxid* or disinfect* or ozon* or persulfate or hydrogen peroxide or peroxydisulfate or Peroxomonosulfate or peracetic acid or *iodate or UV light or ultraviolet light or visible light or solar light)	1,430,848
#2	TI= (*char* or activated carbon or carbon nanotube* or graphene or graphene oxide* or fullerene* or carbon fiber* or graphite or carbon quantum dot* or reduced graphene oxide* or CNT* or * GO)	3,094,010
#3	TI= (membrane* or *film* or thin film* or filter*)	1,161,553
#4	TS= (*water* or Aqu* or effluent* or solution*)	8,297,449
	#4 AND #3 AND #2 AND #1	1,377

environmental remediation. By presenting the journals that cite these published articles, Fig. 1d confirms the focus on technological improvements through journals like Journal of American Chemistry Society, Advanced Materials, Applied Surface Science, Chemical Engineering Journal, Journal of Electroanalytical Chemistry, Carbon, as well as environmental theme through journals like Separation and purification technology, Journal of Membrane Science, Applied Catalysis B: Environment and Energy, and Environmental Science and Technology, International Journal of Hydrogen Energy.

3. Preparation methods for carbonaceous materials-based membrane

Optimal means for hybridizing the carbonaceous nanomaterials with the membrane structure is crucial for obtaining high-performance catalytic membranes for organic wastewater treatment applications. Here, the key methods of blending, *in-situ* growth and interfacial polymerization are overviewed. Finally, the pros and cons of the different fabrication methods are critically discussed.

3.1. Blending

The blending method, which has been developed recently, is one of the most widely applied approaches for the integration of carbonaceous nanomaterials into membrane structures due to the simplicity and superior results [34]. Typically, the carbonaceous nanomaterials are mixed with an organic solution to form a homogeneous solution via ultrasound [35], heating [36], or melting [37]. Subsequently, the casting solution is used for the fabrication of membranes through different techniques including dip-coating [38], spin-coating [39], spray-coating [40], phase inversion [41], etc.

Dip-coating is a facile method for the preparation of the membrane. As illustrated in Fig. 2a, the immersion of the substrate in the nanoparticle suspension coats it with layers of nanomaterials [42], with the thickness controllable by adjusting factors such as concentration, temperature, and coating time [43]. As an example, Guo et al. [44] prepared multiwalled carbon nanotubes (MWNTs) enhanced poly(styrene-alt-maleic anhydride)/fluorocarbon surfactant (PSMA/MWNTs/FS) membranes through dip-coating of PSMA/MWNTs/FS on stainless steel meshes for 30 s. Spin-coating is another established technique (Fig. 2b [45]), with advantages compared to dip-coating like more uniform dispersion of the nanoparticles, and thinner and smoother membranes [46]. This can be attributed to the synergistic effect of the forces (e.g., electrostatic, centrifugal, and viscous) generated by the spinning motion. For instance, Miao et al. [47] fabricated a carbon nanotube membrane by spin-coating the casting solution (consisting of polyvinylidene fluoride, dimethylformamide, and carbon nanotube) on the surface of the silicon wafer, followed by vacuum drying. In addition, spray-coating is also a widely used technique, which usually involves an external force

(such as compressed air) to press the precursor solution onto the substrate surface (Fig. 2c [42]). This technique has been proven to be useful for preparing carbonaceous materials-based membranes, such as polytetrafluoroethylene-CNTs composite membranes [48].

Another approach for the fabrication of carbonaceous-based membranes is phase inversion, which is a complex process that involves phase separation and polymer solidification [49]. In this process (Fig. 2d), the homogeneous polymer-carbonaceous materials casting solution is transformed into a solid phase by changing the temperature or exposure to another phase (e.g., liquid or vapour) [50]. Based on the phase transformation methods, this can be classified into non-solvent-induced phase separation (e.g., wet casting, immersion precipitation) [51], vapour induced phase conversion [52], and thermally induced phase separation [53]. For instance, Liu et al. [54] fabricated a graphene oxide-modified polyvinylidene fluoride (PVDF) ultrafiltration membrane through an immersed phase inversion process.

Electrostatic spinning, a facile and rapid membrane fabrication technique, has attracted much attention recently due to the resulting membrane exhibiting high stability and excellent separation performance in wastewater treatment applications [55]. As the solution is discharged onto the collector, the small droplets and nanoparticles are charged under a high electric field along with the collector (Fig. 2e [56]). While the droplets are become elongated on the collector to form a three-dimensional fiber network, the nanoparticles are uniformly dispersed onto the fiber membrane structure [57]. As an example, Du et al. [58] prepared a carbon nanotube (CNT)/nanofiber composite hollow fiber membrane with high porosity and low tortuosity through an electrostatic spinning process under the voltage of 9 ~ 21 kV and injection speed of 1 mL h⁻¹.

3.2. In-situ growth

As a direct growth technique, *in-situ* growth has been considered a simple and effective method for the fabrication of carbonaceous materials-based membranes. Typically, the nanomaterials are formed on

the surface of the membrane substrate by surface chemical reactions (e.g., hydrothermal reaction, catalytic reaction, reduction) [59]. Various parameters can be adjusted during the process to optimize the resulting membrane (e.g. thickness, porosity, permeability), including precursor concentration, growth time, and initiator ratio. In addition, the *in-situ* growth method can mitigate the agglomeration problem of nanomaterials during the membrane fabrication process [60]. For instance, a BC/geopolymer composite membrane with a large specific surface area, hierarchically porous structure, and abundant surface functional groups (e.g., —OH, C=O) was fabricated through an *in-situ* simultaneous carbonation and self-activation process at 600 °C for 2 h under an N₂ atmosphere [61]. As another example, Zhang et al. [62] prepared a graphene oxide/covalent organic framework membrane by *in-situ* growth of the nanocomposite on the polyacrylonitrile substrate using polyvinyl alcohol and poly(vinylamine) matrix as the cross-linker. As shown in Fig. 3a, Manoranjan et al. [63] also fabricated a single-walled carbon nanotube/covalent organic framework (SWCNT/COF) film by *in-situ* growth of COF on the surface of SWCNT substrate.

3.3. Interfacial polymerization

Interfacial polymerization (IP) is a well-developed technology that has been used worldwide for the fabrication of polymeric thin film composite (TFC) membranes. Typically, the porous substrates are first impregnated in a water solution containing an amine monomer (e.g., m-phenylenediamine (MPD) or piperazine (PIP)), after which the excess solution is removed [67]. This is followed by the introduction of an immiscible non-polar organic solvent with acid chloride monomers (e.g., trimesoyl chloride (TMC)), leading to a reaction-diffusion process occurring between the amine monomer and acid chloride monomer to form a polyamide membrane [68]. This type of membrane usually exhibits high water permeability and selectivity, as well as excellent durability in a broad range of pH (2-11) and temperature (0-45 °C), which underlie its widespread utilization in wastewater purification [69]. As illustrated in Fig. 3b, Zhang et al. [64] successfully prepared a

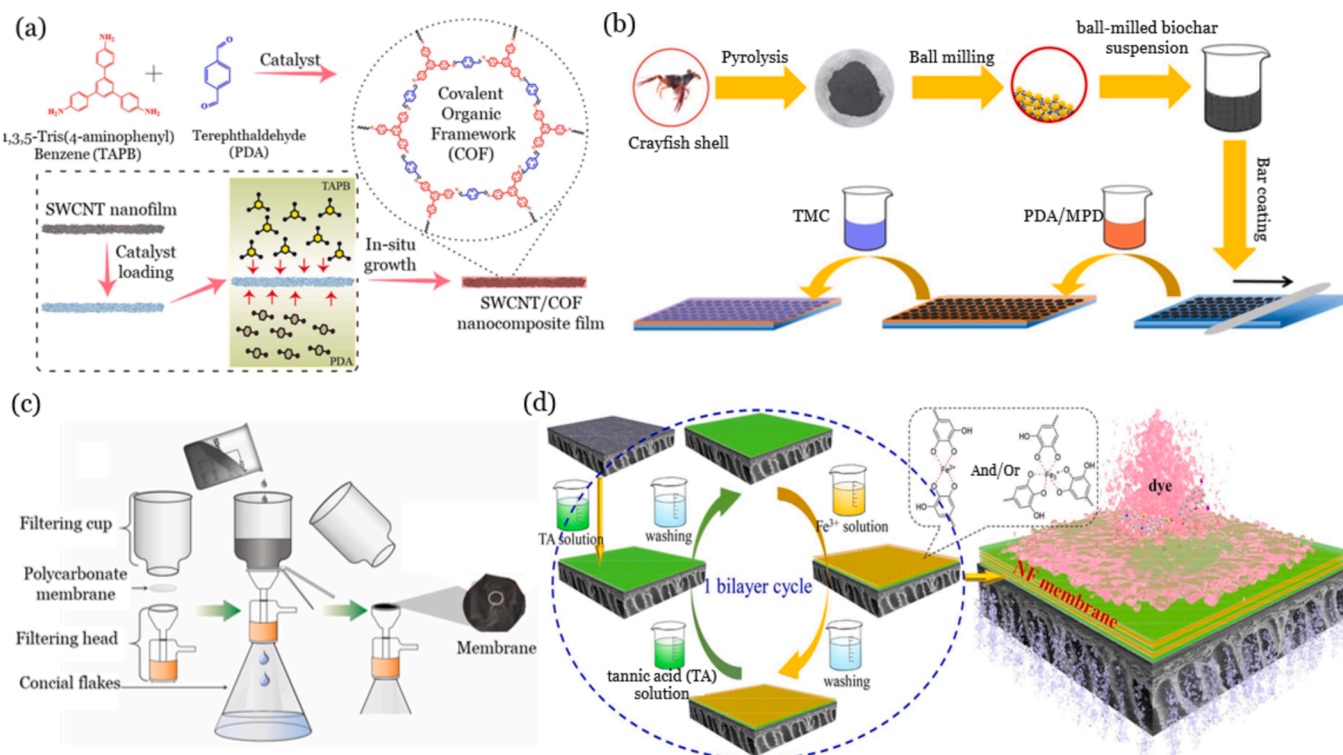


Fig. 3. Schematic illustration of various preparation methods: *in-situ* growth (a), adapted from Manoranjan et al. [63]; interfacial polymerization (b), adapted from Zhang et al. [64]; filtration process (c), adapted from Dehingia et al. [65]; layer-by-layer assembly (d), adapted from Xiao et al. [66].

polydopamine (PDA)/TFC membrane incorporated with crayfish shell biochar (BC) active interlayer through a conventional IP process. Specifically, the surface functional groups (e.g., OH) of BC interact with the hydroxyl groups of PDA molecules to form hydrogen bonds, which results in tightly bound BC interlayers and PDA layers in the membrane structures. As another example, Yang et al. [70] prepared a nanofiltration membrane composed of carbon quantum dots as the interlayer through an IP process. Firstly, polyethersulfone (PES) substrates were submerged in the carbon quantum dots suspension, followed by the polyethyleneimine solution for two minutes. Finally, the substrate was exposed to hexane solution containing 0.1 wt% TMC for 1 min, resulting in a polyamide layer on the surface of the carbon quantum dots modified PES substrate through the IP reaction between polyethyleneimine and TMC. A disadvantage of the conventional IP process is that the resulting TFC membrane typically exhibits nanoscale heterogeneity and wide pore size distribution due to the mismatch between the slow diffusion rate of amine monomers and the high reactivity of the monomers [71]. To address this, various assistive techniques, such as vacuum-assisted [72], spray-assisted [73], and electro-spray-assisted [74] IP processes, have been proven to be useful in improving membrane properties.

3.4. Other methods

Implementing filtration to immobilize carbonaceous materials onto the surface or inside of membrane structures is a facile and excellent alternative. Typically, the suspensions containing the nanomaterials are filtered through a porous membrane (Fig. 3c), thus attaching the nanomaterials to the surfaces through various effects (e.g., covalent bonding, hydrogen bonds, electrostatic interactions) [65]. For instance, Zheng et al. [75] successfully synthesized rGO nanosheets-based nanofiltration membranes via vacuum filtration of rGO suspensions through commercial PES membranes. In another study, Ding et al. [76] constructed CNTs/Mxene composite membranes via vacuum filtration through nylon membranes with a characteristic pore size of 0.22 μm .

Layer-by-layer assembly is another common technique for the fabrication of membranes. In general, the membrane can be fabricated through electrostatic force effects by alternately immersing a substrate into oppositely charged polyelectrolyte solutions [77]. As shown in

Fig. 3d, a novel layer-by-layer self-assembly method was developed for the fabrication of tannic acid (TA)- Fe^{3+} network based nanofiltration membrane [66]. In addition, this approach has been explored for the fabrication of carbonaceous materials-based membranes. For instance, a polyethyleneimine/graphene oxide membrane was constructed in this way [78]. Firstly, the positively charged polyethyleneimine solution interacted with the negatively charged polyamide substrate for 1 h, after which the substrate was rinsed with water to remove the excess solution. Subsequently, the substrate was introduced into the anionic GO solution to form the polyethyleneimine/graphene oxide membrane through electrostatic adhesion.

Furthermore, some novel assistive techniques have been explored for the fabrication of membranes with excellent properties. Plasma, which is an ionized gas that usually contains highly reactive substances such as ions, electrons, free radicals and neutral particles [79], has been applied for the modification of carbonaceous-based membranes to enhance hydrophilicity and confer surface functional groups. For instance, GO nanosheets with in-plane nanopores-based membranes (Fig. 4a [80]) and rGO-Ag nanofiltration membranes [81] were successfully fabricated through a plasma-assisted process. In addition, 3D printing is a promising additive process that has been utilized for the fabrication of carbonaceous materials-based membranes, such as polylactic acid-GO membranes (Fig. 4b) [82].

3.5. Summary and comparison

Various techniques (e.g., blending, *in-situ* growth, interfacial polymerization) have been developed for the fabrication of carbonaceous nanomaterials-based membranes, with each having some advantages and limitations, as discussed in Table 2. To address the limitations, the different techniques have been combined. For instance, a rGO/CNTs ceramic membrane was prepared through an *in-situ* oxidative polymerization-self-assembly combining chemical oxidation polymerization, impregnation reduction, and heating [83]. Also, Ma et al. [84] developed a novel strategy of embedding Prussian blue (PB) nanoparticles into reduced graphene oxide (rGO) lamellar membrane structure using *in-situ* growth and then thermal reduction processes (Fig. 4c). In another study, Kang et al. [85] confirmed that the graphene oxide

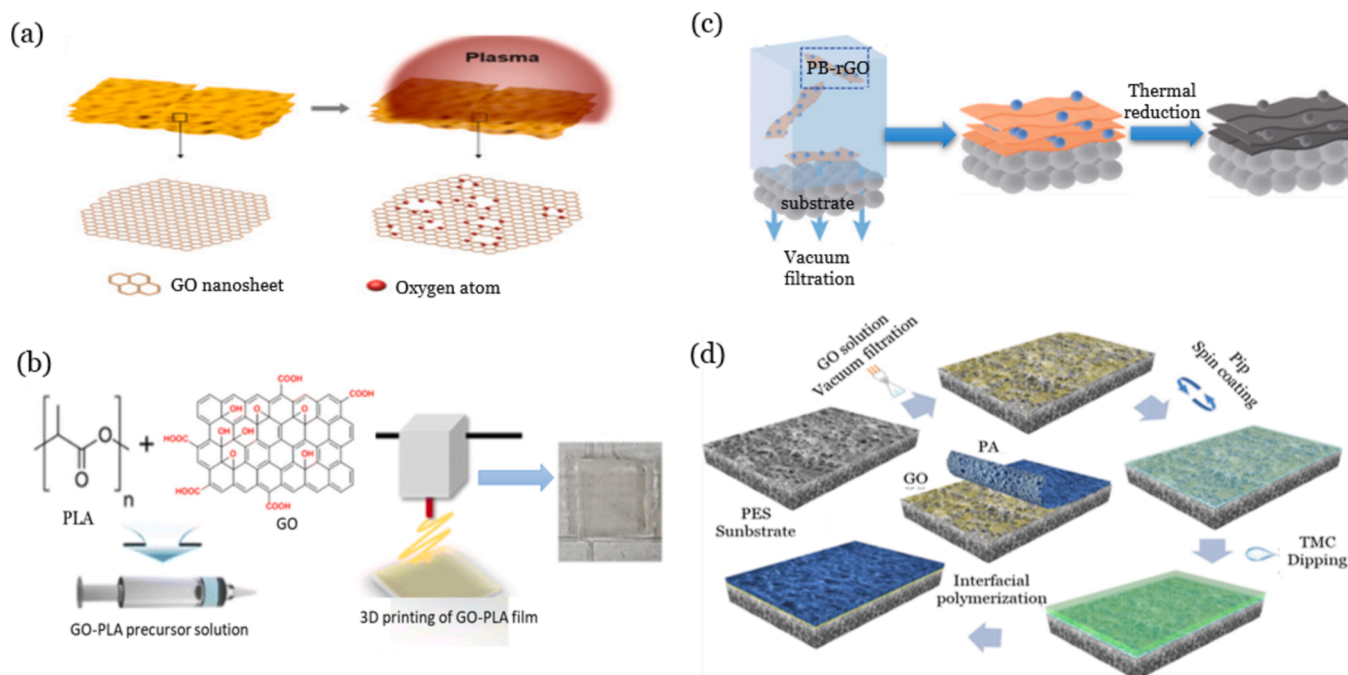


Fig. 4. Schematic illustration of fabrication steps: (a) plasma-assisted process for GO-based membrane (adapted from Chen et al. [80]); (b) 3D printing for poly(lactic acid)-GO membrane (adapted from Jang et al. [82]); (c) rGO membrane (adapted from Ma et al. [84]); (d) GO-polyamide membrane (adapted from Kang et al. [85]).

Table 2
Summary of preparation methods for carbonaceous materials-based membranes.

Methods	Advantages	Limitations	Ref.
Dip-coating	Simple operation, cost-effectiveness, and high versatility.	Limited uniformity and presence of defects.	[86]
Spin-coating	High uniformity, and precise thickness control.	Wastage of solvents, high requirements for the substrate.	[45]
Spray-coating	Suitable for large-area coating.	Limited uniformity, and spray materials may contain volatile organic compounds.	[42]
Phase inversion	Simple operation, tuneable porous structure and membrane properties.	Complex process, wastage of solvents.	[87]
Electrostatic spinning	Controllable process, high specific surface of the membrane.	Complex equipment, low yield of products.	[56]
<i>In-situ</i> growth	High stability and high purity, no transfer required.	Complex process.	[88]
Interfacial polymerization	Simple and efficient process, high membrane performance.	Difficult interface control and limited monomer materials.	[67]
Vacuum filtration	Simple, fast and efficient process.	Limited uniformity and pore size distribution accuracy.	[13]
Layer-by-layer assembly	Precise control, hierarchical structure, good uniformity.	Complex process, low efficiency	[89]

(GO) based membrane can be fabricated through vacuum filtration/spin-coating/dip-coating assisted IP process (Fig. 4d).

4. Catalytic membrane systems

Catalytic membranes play different roles in different wastewater decontamination processes. In this section, the catalytic membrane systems (e.g., static, dead-end, cross-flow) and their applications (e.g., photocatalytic, electrocatalytic, advanced oxidation) for organic wastewater treatment are discussed. In general, catalytic membrane systems can be operated three main ways, namely, static, dead-end, and cross-flow. As illustrated in Fig. 5a, the catalytic membrane was immobilized on a holder for the capture of light and subsequent generation of active species for the degradation of organic pollutants from wastewater [90]. In this process, the membrane structures act as substrates for catalysts immobilization and do not play any role in separation. The light source can be replaced by various oxidizing agents (e.g., persulfate, hydrogen peroxide) for converting to a catalytic membrane-oxidizing agent system. The catalytic membrane can directly activate oxidizing agents to decompose organic pollutants through the

production of active species, the type and rate of which are highly dependent on the type of catalyst and activation process. The dead-end filtration is a facile and simple process for the removal of pollutants. As depicted in Fig. 5b, an external force (normally provided by a gas bottle or pump) drives the liquid vertically through the membrane, which gets rid of the pollutants through a combination of different effects (e.g., rejection, degradation, adsorption) depending on the properties of the membrane [91]. The cross-flow system is another commonly used filtration process that has a lower fouling possibility compared to dead-end filtration. This is mainly due to the shear force of the horizontal flow of feed solution effectively reducing the accumulation of pollutants on the membrane surface (Fig. 5c) [92].

Furthermore, catalytic membranes can be applied in different processes either individually (e.g., photocatalytic, advanced oxidation, electrocatalytic, ozone-based, and thermal-based) or combined (e.g., photoelectrochemical, photo-assisted advanced oxidation). In Fig. 5d, the photocatalytic membrane can not only separate pollutants but also be activated by light for the generation of active species for decomposing pollutants [93]. As illustrated in Fig. 5e, the wastewater and oxidizing agents are pumped onto the membrane surface, and then the oxidizing

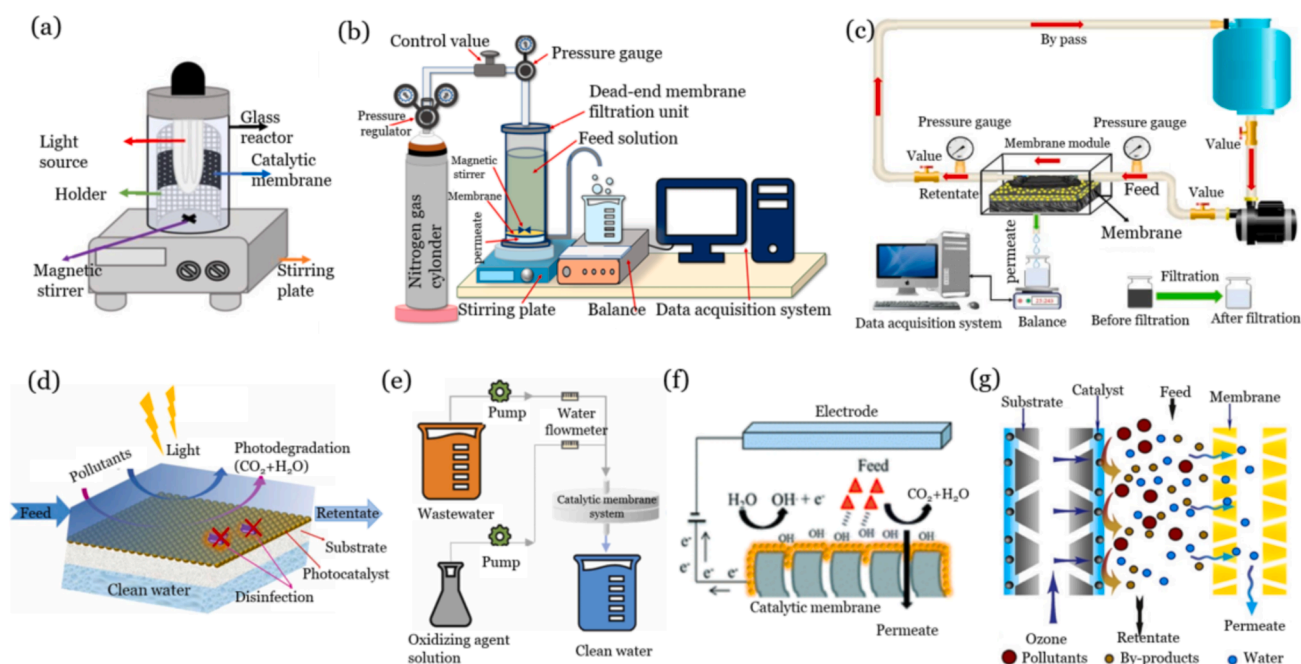


Fig. 5. Schematic diagrams of the modes of operation of the catalytic membrane systems: (a) static system (adapted from Singcharoen et al. [90]); (b) dead-end filtration system (adapted from Kamal et al. [91]); (c) cross-flow system filtration system (adapted from Hosseinzadeh et al. [92]); (d) photocatalytic membrane for the purification of wastewater (adapted from Chen et al. [93]); (e) catalytic membrane-oxidizing agents system (adapted from Guo et al. [94]); (f) electrochemical system (adapted from Kumari et al. [95]) and (g) catalytic membrane-ozone process (adapted from Li and Yeung [96]).

agent can be activated by the catalytic membrane to produce the active species. Hence, this membrane can remove pollutants from wastewater through the synergistic effects of separation and degradation [94]. In addition, the catalytic membrane can also be applied as an anode in the electrochemical system for the generation of hydroxyl radicals (Fig. 5f), which can also degrade pollutants from the membrane surface to alleviate membrane fouling [95]. Ozonation is an effective technology that also has been applied with the combination of catalytic membranes. As shown in Fig. 5g, the catalytic membrane can not only reject the pollutants but also promote the ozonation process by accelerating the generation of reactive hydroxyl radicals [96]. Moreover, hybridized systems based on the catalytic membrane (e.g., photo/electro-catalytic membrane [19], photothermal membrane [97]) have been also demonstrated to be efficient for organic wastewater treatment.

5. Carbonaceous materials-based catalytic membranes for organic wastewater decontamination

5.1. Carbon nanotubes-based catalytic membranes

CNTs have received enormous attention for wastewater treatment over the last few decades due to beneficial properties such as large specific surface area, outstanding electrical conductivities, high stability, and abundant and adjustable surface functional groups [98]. Accordingly, CNTs-modified catalytic membranes have been developed, which have superior pore structures, high specific surface area, good mechanical stability, high water permeability, and excellent removal efficiency for contaminants [99]. As an example, Liu et al. [100] observed that the nitrogen-doped multi-walled carbon nanotubes (N-MWCNTs) in the catalytic membrane structure could increase water permeability from $419 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ to $455 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$, which is

mainly due to the N-MWCNTs increasing the hydrophilicity (contact angle decreased from 68° to 60°), porosity (increased from 43 % to 59 %) and surface roughness (Fig. 6a and b) compared with the virgin membrane. It should be mentioned that, although the high surface roughness of the membrane can improve water permeability by increasing active sites and area, it also leads to rapid fouling of the membrane and thus compromising membrane performance. Fortunately, as shown in Fig. 6c, the abundant graphitic N of N-MWCNTs in the catalytic membrane acted as the active sites for activating PMS in a dead-end system [100], and the increased electrons and excellent conductivity of N-MWCNTs were also beneficial for the generation of active species (e.g., $\cdot\text{OH}$, $\text{SO}_4^{\cdot-}$, $^1\text{O}_2$) [101]. Hence, these species with strong oxidation ability degraded the contaminants into small molecules, which could easily pass through the membrane and alleviate membrane fouling. In addition, CNTs demonstrate high light-to-heat conversion efficiency and outstanding thermal conductivity of $6 \times 10^3 \text{ W M}^{-1} \text{ K}^{-1}$ [102], and thereby CNTs-based catalytic membranes can be applied in the thermal-based advanced oxidation process for wastewater treatment. As illustrated in Fig. 6d, CNTs-based membranes can act as light-to-heat conversion materials for the activation of peroxydisulfate in a flow-by system [103]. Evidently, the CNT membrane effectively harvested light to increase the temperature of the feed solution from 27.6°C to 86.5°C in 10 min (Fig. 6e). The electron paramagnetic resonance spectra (EPR, Fig. 6f) also confirmed that the *in-situ* generated active species ($\cdot\text{OH}$ and $\text{SO}_4^{\cdot-}$) in the medium were predominant for the degradation of various organic pollutants (e.g., methyl orange, phenol, carbamazepine) with a relatively high removal efficiency of above 90 %. This is mainly because the activation of peroxydisulfate is thermodynamically favourable. In general, peroxydisulfate starts to decompose and produce active species when the temperature is higher than 40°C , and the generation rate of active species increases with the increase of

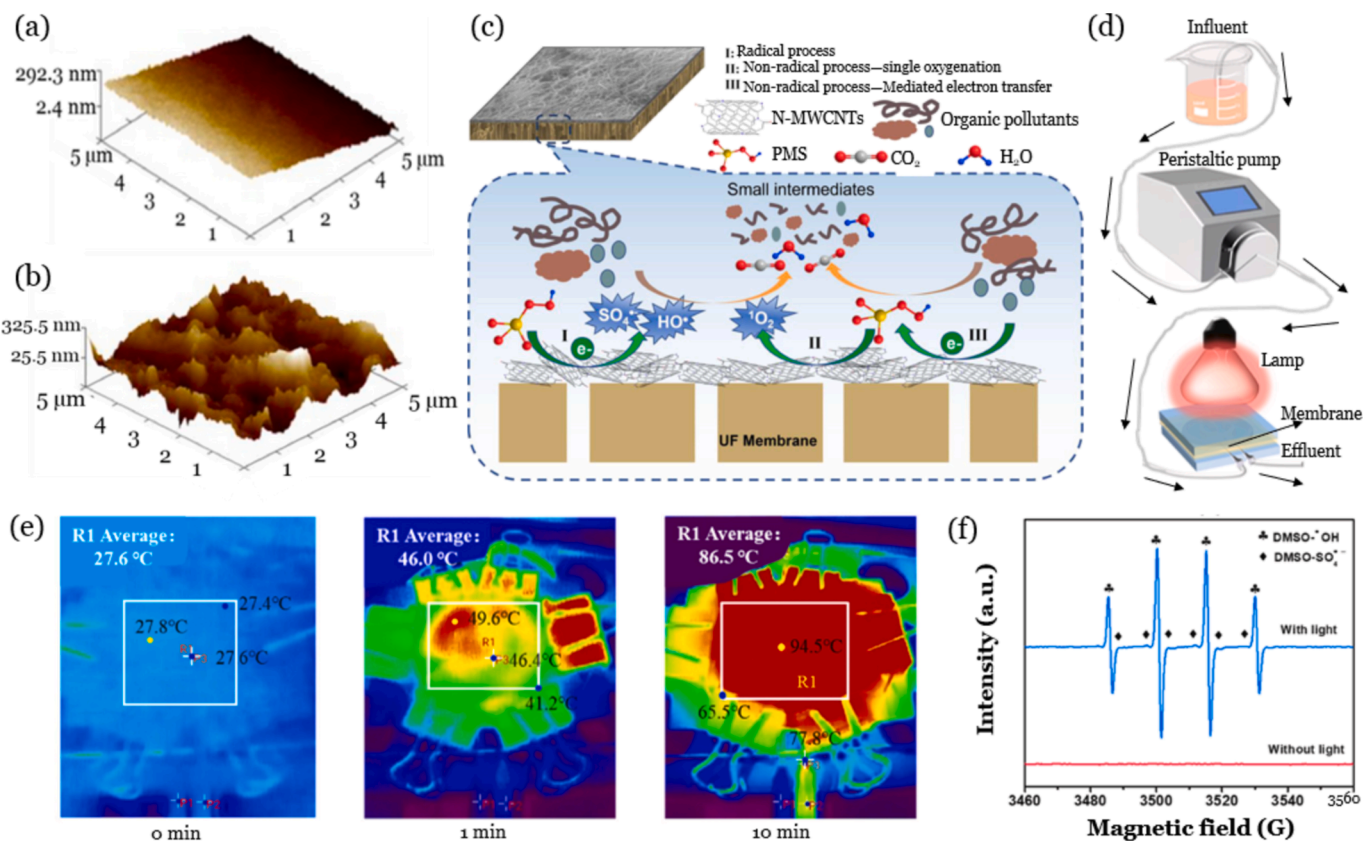


Fig. 6. Atomic force microscopy images of virgin membrane (a) and N-MWCNTs-modified membrane (b), possible mechanisms of self-cleaning N-MWCNTs-modified membrane (c), adapted from Liu et al. [100]; schematic illustration of the photothermal system (d), infrared thermal images of the reactor surface at different times (e), electron paramagnetic resonance spectra for DMPO- $\cdot\text{OH}$ and DMPO- $\text{SO}_4^{\cdot-}$ in the photothermal system, adapted from Yang et al. [103].

reaction temperature (within a certain range) [104].

Furthermore, CNTs-based membranes have great potential in electrochemical wastewater treatment because of the high conductivity, resulting in efficient simultaneous separation and degradation of the organic pollutants from wastewater [105]. Typically, the electrochemical reactions (e.g., oxidation, reduction, electrostatic repulsion or attraction) occur on the membrane surface under an additional electrical potential, which is conducive to the elimination of pollutants and mitigation of membrane fouling [106]. In a relevant study, Xu et al. [107] fabricated a polyaniline/MWCNT membrane as the anode in the electrochemical filtration system for the degradation of organic pollutants. As shown in Fig. 7a, the membrane demonstrated a low removal efficiency of less than 10 % for MB, methylene orange (MO), and phenol under the applied voltage of 1.0 V, when electrostatics govern the interactions between the membrane and pollutants. Under a higher applied voltage of 2.5 V, the membrane exhibited excellent removal efficiency for MB (95 %), MO (96 %), and phenol (96 %), which could be ascribed to the applied voltage being higher than the relative oxidation potential of MB (0.9 V), MO (0.6 V), and phenol (0.7 V) (Fig. 7b). In this case, electrochemical oxidation was responsible for the degradation of these contaminants. In addition to electrochemical oxidation, the CNTs-based membrane can also be used as a cathode to remove organic pollutants and mitigate membrane fouling. As an example, Si et al. [108] observed that using conductive stainless steel/CNTs membrane as the cathode and Ti mesh as the anode significantly alleviated fouling and corrosion of the membrane (Fig. 7c). This is mainly because of the strong repulsions between the negatively charged membranes and humic acid, and thus the pollutants cannot accumulate on the surface of the membrane. Moreover, the Ti mesh anode provided abundant electrons from the anode to the membrane to prevent the corrosion of the membrane.

Recently, the combination of electrically conductive membranes and advanced oxidation processes have been advanced for wastewater treatment applications because of their synergistic effects. For instance,

Xu et al. [109] fabricated a polypyrrole/CNTs PVDF membrane for the activation of peroxydisulfate to degrade organic pollutants in the electro-filtration system. This membrane displayed a high removal efficiency and high stability for carbamazepine (90 % after 20 cycles), while also exhibiting a high water flux recovery ratio of 95 %. The excellent performance is due to the electric field preventing membrane fouling though electrostatic repulsion, and the *in-situ* generated active species (e.g., $\cdot\text{OH}$, $\text{SO}_4^{\cdot-}$) by catalytic membrane/electrochemical effects degrading the organic contaminants on the surface of the membrane. Furthermore, ozonation, which is an extensively used wastewater treatment technique, has also been integrated with the electrocatalytic membrane to further enhance the water treatment efficiency. As illustrated in Fig. 7d and e, the electro-peroxone CNT/polytetrafluoroethylene membrane filtration system exhibited an outstanding removal efficiency for ibuprofen (97 %), which is 3.7, 2.0, and 1.6 times compared with the sole electrochemical filtration system, ozonation system, and ozonation-filtration system, respectively [110]. The $\cdot\text{OH}$ was proven as the main radical for degrading ibuprofen in the electro-peroxone membrane filtration system, and the synergistic effects of adsorption, electrochemical, ozonation, and catalytic processes were involved in pollutant removal.

Unfortunately, in real wastewater treatment applications, the performance of single CNTs catalytic membranes remain inadequate, at least in part due to membrane fouling. To address this, the combination of CNTs and metal-based nanomaterials is attractive. This can not only prevent the agglomeration of metal-based nanomaterials, but also retain the abundant surface functional groups and high porosity of CNTs [111]. Several studies have further explored CNTs/metal-based nanomaterial composite membranes for water treatment. As shown in Fig. 8a–c, Cheng et al. [21] observed that Co_3O_4 nanomaterials could attach uniformly on the surface of CNTs to prevent the agglomeration of magnetic Co_3O_4 nanoparticles. Subsequently, the Co_3O_4 @carbon nanotube composites were coated on the surface of the ceramic membrane through a

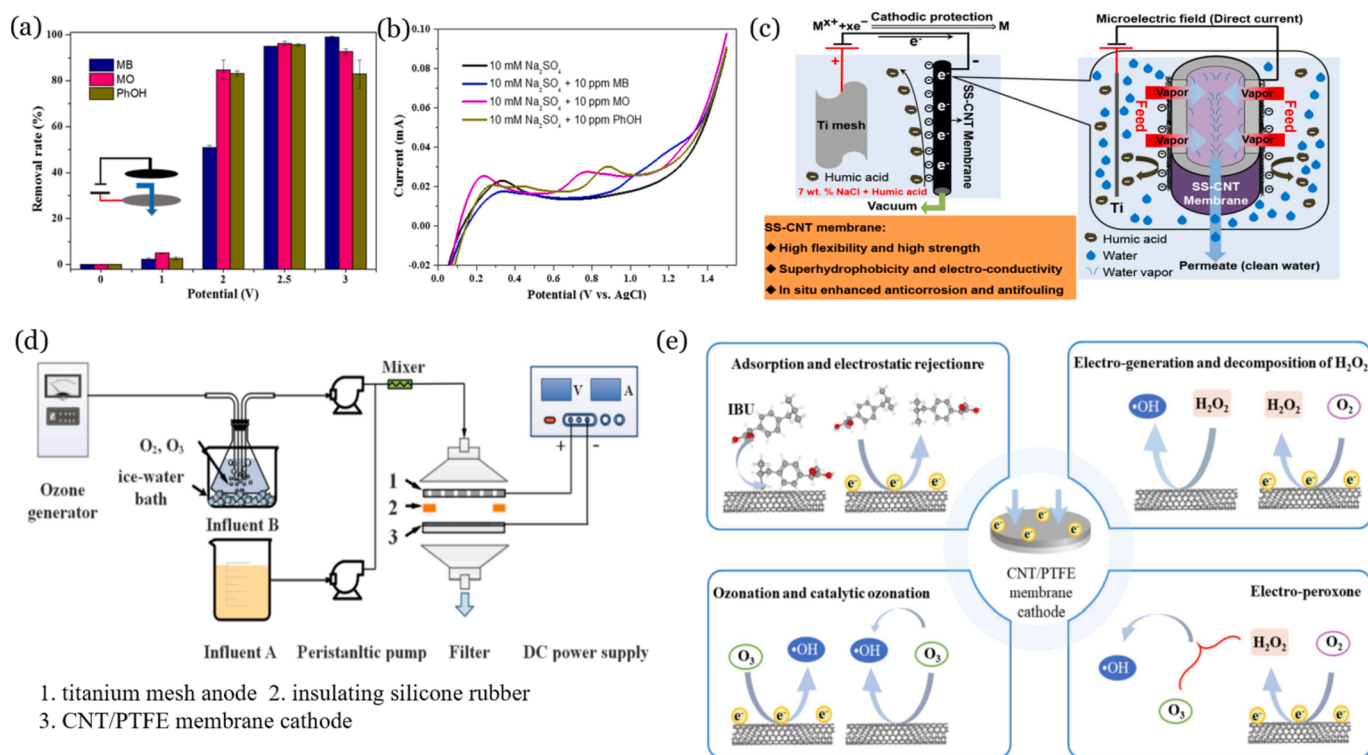


Fig. 7. Removal efficiency of pollutants under different applied potentials in the electrochemical membrane system (a), the relative oxidation potential of pollutants with the membrane as working electrode (b), reprinted with permission from Xu et al. [107]; schematic illustration of the conductive stainless steel/CNTs membrane in the electrochemical system (c), reprinted with permission from Si et al. [108]; schematic diagram of the electro-peroxone membrane filtration system (d), schematic illustration of major processes involved in the ibuprofen (IBU) removal by this system (e), reprinted with permission from Yang et al. [110].

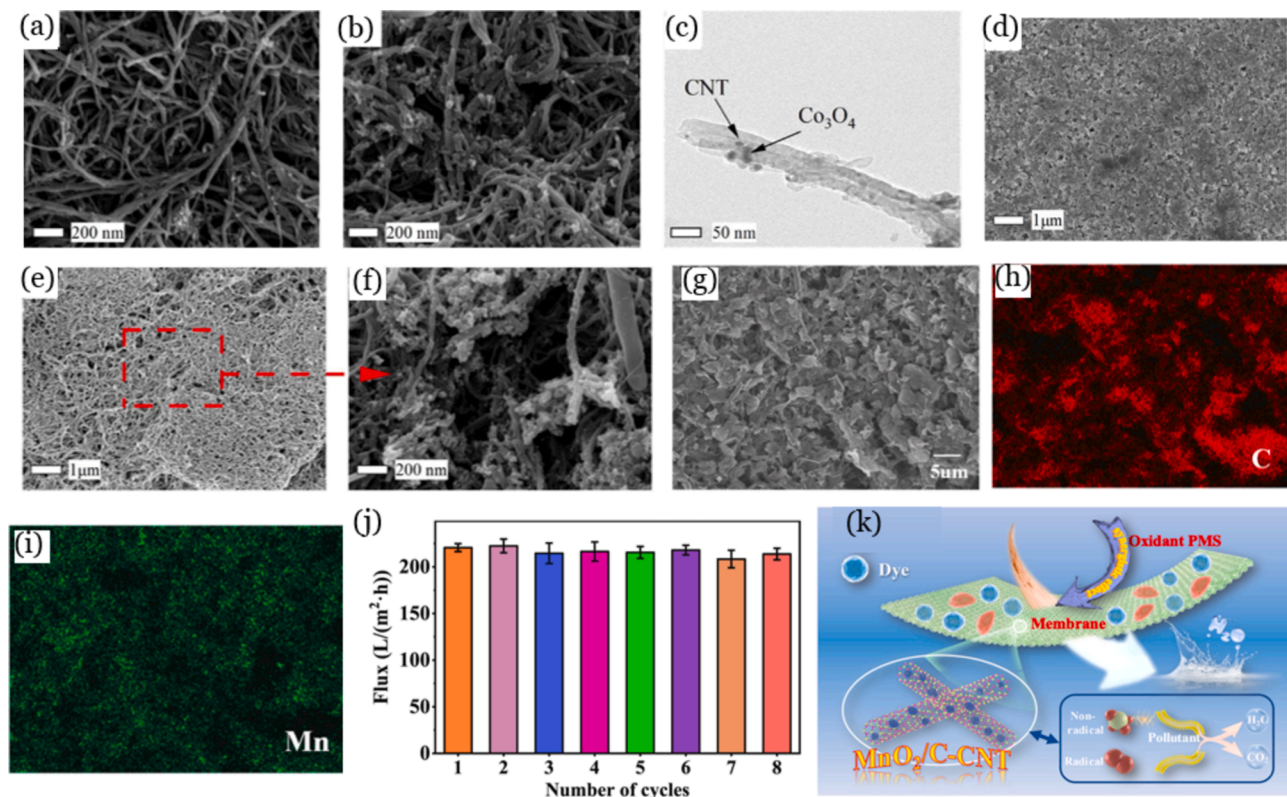


Fig. 8. SEM images of CNT(a), Co₃O₄@CNT (b), TEM image of Co₃O₄@CNT (c), SEM images of the virgin membrane (d), Co₃O₄@CNT-ceramic membrane (e-f), reprinted with permission from Cheng et al. [21]; SEM images of MnO₂/C-CNT@PVDF membrane (g), EDX mappings of C and Mn elements (h-i), water flux of the MnO₂/C-CNT@PVDF membrane for Rhodamine B (Rh B) degradation over eight cycles (j), and schematic illustration of the self-cleaning membrane (k), reprinted with permission from Chen et al. [112].

vacuum filtration process for fabricating the composite membrane (Fig. 8d–f). The membrane exhibited better antifouling performance compared to the pristine membrane in terms of decreasing the reversible and irreversible resistances by 17 % and 21 %, respectively. This is mainly attributed to the fact that the large specific surface areas of CNTs can both prevent the agglomeration of Co₃O₄ and also provide more active sites for the activation of PMS to give more active species (e.g., ·OH, O₂^{·-}, ¹O₂) in the medium. Hence, the generated active species can degrade the organic pollutants deposited on the membrane surface and pore structures, thereby improving the antifouling capability of the membrane. Similarly, Chen et al. [112] reported that MnO₂/CNT could uniformly distribute on the surface of the PVDF membrane, as confirmed by the SEM and EDX mapping results (Fig. 8g–i). The MnO₂/CNT catalytic membrane demonstrated a stable water flux after eight cycles in the presence of PMS solution (Fig. 8j), which is mainly due to MnO₂/CNT on the membrane surfaces quickly adsorbing pollutants, and efficiently activating PMS for the generation of active species to degrade the pollutants (Fig. 8k). Hence, this membrane exhibited excellent antifouling performance.

5.2. Graphene-based catalytic membranes

Graphene, which is a two-dimensional planar carbon allotrope consisting of a single layer of carbon atoms arranged in the honeycomb network, has some inherent beneficial properties such as large specific surface area, excellent electron mobility (200000 cm² V⁻¹ s⁻¹), outstanding stability, and high catalytic activity [113]. To date, graphene-based materials have proliferated because of advantages such as high flexibility, adjustable interlayer spacing and surface functional groups [114]. For instance, Liu et al. [54] fabricated a GO-modified PVDF ultrafiltration membrane with excellent mechanical properties

and hydrophilicity through an immersed phase inversion process. They also observed that the amount of GO in the membrane structure was crucial for membrane performance, because partial agglomeration of GO under higher concentrations resulting in rough membrane surfaces and defects, which reduce the mechanical properties and hydrophilicity of the membrane [115]. In addition, the oxygen-containing functional groups of GO on the membrane surface increases water transport resistance due to more extensive hydrogen bond generation between GO and water molecules [116]. Therefore, optimization of the content of GO in the membrane structure is essential to obtain membranes with high water permeability, high hydrophilicity, and high removal efficiency of pollutants. To reduce the oxygen-containing functional groups on the surface, reduced GO (rGO) have been used. As shown in Fig. 9a–f, Sheng et al. [117] observed that the packing compactness of the carbon mats increased with the increase on the reduction degree of GO in the rGO/CNTs membrane structure, with the decrease in average thickness from 5.16 μm to 4.41 μm resulting in higher water permeability. The excellent sulfamethoxazole removal efficiency of the rGO/CNTs membrane in a circulating filtration system was related to the abundant active sites (e.g., Lewis basic sites of GO-based materials, non-six-membered carbon rings) on the membrane efficiently activating persulfate to generate active species (e.g., ·OH, SO₄^{·-}) [118]. In addition, the CNTs and rGO in the membrane structure acted as electron mediators to accelerate the electron transport between persulfate and contaminants, promoting the production of active species in the medium. The increased active species in the medium more efficiently degraded organic pollutants in such systems [119].

More recently, non-metal-doped GO has also been applied to catalytic membranes to further improve the membrane properties and water treatment efficiency. For instance, a nitrogen-doped reduced GO (melamine as the nitrogen source) catalytic membrane was constructed for

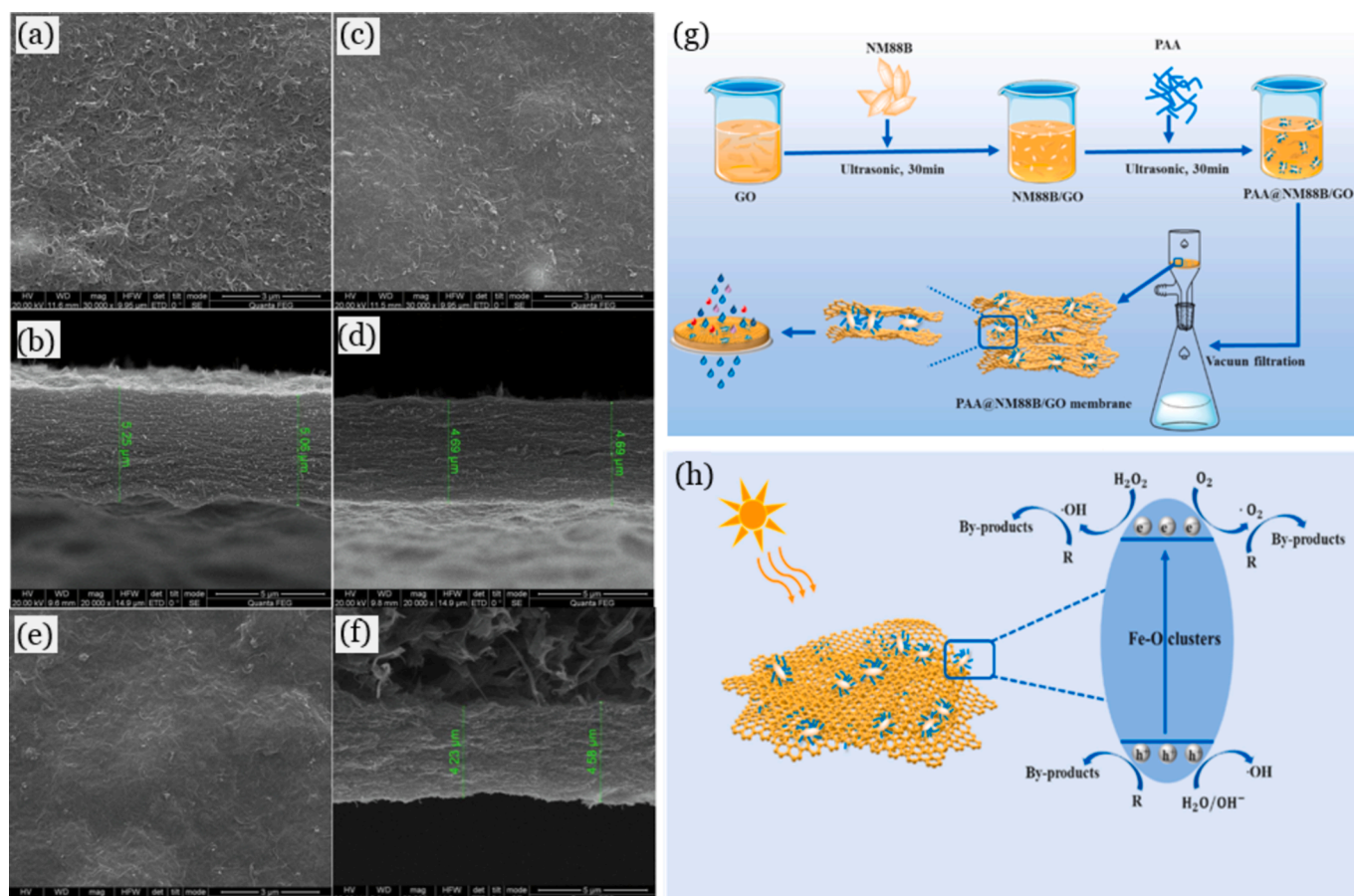


Fig. 9. The top-view and cross-sectional SEM images of rGO_{0h}/CNTs (a, b), rGO_{3h}/CNTs (c, d) and rGO_{8h}/CNTs (e, f), (8 h is the highest reduction degree), reprinted with permission from Sheng et al. [117]; schematic illustration of the fabrication of polyacrylic acid (PAA)@ NH₂-MIL-88B(Fe) (NM88B)/GO membrane (g), schematic diagram of self-cleaning mechanisms of the membrane under visible light(h), reprinted with permission from Gao et al. [123].

the activation of persulfate to simultaneously degrade three fluoroquinolone antibiotics (namely, ciprofloxacin, ofloxacin, enrofloxacin) in a continuous system [120]. The reasonable removal efficiencies (80 %, 54 %, and 91 % for ciprofloxacin, ofloxacin, and enrofloxacin, respectively) are tied to the activation of persulfate by catalytic membranes for generating active species. Song et al. [121] also observed that the nitrogen-doped reduced-GO (melamine as the nitrogen source) catalytic ceramic membrane/ozone system demonstrated a 3.9 times higher removal efficiency for p-chlorobenzoic-acid, as compared with the ceramic membrane/ozone system. This can be attributed to the abundant active sites on the nitrogen-doped reduced-graphene-oxide membrane efficiently anchoring ozone molecules and accelerating electron transport, which is beneficial for the generation of $\bullet\text{OH}$ radicals. In addition, it should be noted that, while nitrogen-doped GO-based membranes usually give higher degradation performance than that of GO-based membranes, the source of the nitrogen during preparation can significantly affect the properties of the materials. Pedrosa et al. [122] observed that nitrogen-doped GO with melamine as a nitrogen source had the highest nitrogen percentage (24 %), compared with using urea (13 %) and gaseous ammonia (11 %). Thus, in the presence of persulfate, the nitrogen-doped GO based on melamine demonstrated a higher 96 % degradation efficiency of phenol, compared to that of 40 % and 55 % for membranes prepared with respectively urea and gaseous ammonia. This can be attributed to the higher nitrogen content of GO providing more active sites for the activation of persulfate to generate more active species for the degradation of such pollutants.

Integrating catalysts into CNTs-based membranes can efficiently improve the removal of pollutants. Similarly, integrating catalysts into GO-based membrane structures has also been demonstrated as an

efficient method to improve the performance of the membranes. As an example, a polyacrylic acid@NH₂-MIL-88B(Fe)/GO membrane was fabricated through a facile vacuum filtration process (Fig. 9g), which gave high hydrophilicity, water permeability, and excellent degradation efficiency of 99 % for MB under visible light irradiation in the static system (20 mg/L MB, 50 μL 30 % H₂O₂, initial pH = 7) [123]. During the photo-Fenton process, the NH₂-MIL-88B(Fe)/GO of the membrane was activated for the generation of photoelectrons and holes, which could be further transformed into $\cdot\text{OH}$ (Fig. 9h). In addition, the iron ions of the materials could efficiently activate H₂O₂ to produce $\cdot\text{OH}$, and hence the holes and $\cdot\text{OH}$ with strong oxidation ability can directly decompose the organic pollutants on the surface of the membrane [124]. Moreover, other highly efficient catalysts such as Ag₂CO₃@UiO-66-NH₂ [125], Mn₃O₄/g-C₃N₄ [126], layered double hydroxide@g-C₃N₄ [127], and TiO₂ [128] have also been incorporated into GO-based membrane structures to improve the degradation efficiency of pollutants. Recently, some novel methods have been explored to further enhance the removal efficiency of contaminants in the GO-based membrane system. In particular, vacancy (e.g., oxygen vacancy, sulfur vacancy) is known as a lattice defect, which can produce excess charge carriers and accelerate the electron transfer of materials [129]. Hence, nanomaterials with abundant vacancies usually exhibit high efficiency in water treatment applications. In a related study, Ye et al. [130] synthesized novel Fe₂O₃@Co_{0.08}Fe_{1.92}@nitrogen-doped rGO@CNTs composites with enriched oxygen vacancies, following vacuum filtration was implemented to obtain the composite membrane. The membrane/PMS system displayed an excellent removal efficiency (separation and degradation) of around 100 % for sulfamethoxazole. It was also verified that the abundant oxygen vacancies of the composite improved electron transfer

to increase the carbon Fermi level, which enhances the degradation of organic pollutants. Based on the high organics removal efficiencies of CNTs-based catalytic membranes, a reasonable hypothesis is that the CNTs-GO composite membranes can further augment performance. Jiang et al. [131] also proved that carboxylated CNTs intercalated rGO-iron oxides membranes exhibited a high removal rate ($38.6 \text{ mg m}^{-2} \text{ h}^{-1}$) for florfenicol in the electro-Fenton membrane filtration system. They also reported that the incorporation of CNTs and rGO not only significantly enhanced the surface charge and electrical conductivity of the membrane, but also provided a large specific surface area for the immobilization of iron oxides, which were conducive to the generation of $\cdot\text{OH}$ to degrade contaminants.

Furthermore, GO has been extensively used in wastewater disinfection due to its excellent antibacterial properties, which can be classified into physical and chemical damages to the bacteria [132]. Physical damage refers to the direct physical contact between the pathogen and GO, with GO significantly damaging the bacterial wall to disrupt the metabolic processes. Chemical damage refers to oxidative stress due to the generation of active species [133]. GO-based catalytic membranes have been reported for wastewater disinfection. Vatanpour et al. [134] reported that boron nitride nanosheets/graphene oxide/silver polyethersulfone membranes exhibited excellent bactericidal performance for *E. coli*. The GO with a large specific surface area not only prevents the agglomeration of nanomaterials (e.g., Ag nanoparticles), but also efficiently adsorbs bacteria on their surfaces. The released Ag ions and reactive oxygen species efficiently destroyed the cellular respiration and permeability of bacteria, which killed the bacteria [135]. Additionally, the boron nitride nanosheets displayed high antibacterial properties by synergy with biopolymers. Hence, excellent antibacterial performance by membranes can be exploited for real wastewater disinfection applications.

5.3. Biochar and activated carbon-based catalytic membranes

In view of sustainability, biochar (BC), as a biomass waste – derived material, has attracted numerous interests for wastewater treatment applications, such as adsorption, precipitation, and degradation. This can be attributed to the inherent advantageous properties of BC-based materials, including cost-effectiveness, tunable surface functional groups and specific surface area, and high removal efficiency for organic pollutants [136]. Studies on BC-based materials used for the efficient catalytic degradation of various organic pollutants have been reported, including N, S co-doped corncob BC/PMS [137], manganese oxide-corn straw BC/periodate [138], sludge BC-Co-BiOCl/UV [139], pomelo BC/visible light [140], and Pd-BC electrode [141]. Other studies have also reported BC-based catalytic membranes for wastewater treatment applications. For instance, a wood BC-based membrane was applied for the activation of PMS to degrade sulfamethoxazole (100 %) in a static system [142], with the BC layer on the membrane surface acting as a catalyst to activate PMS for the decomposition of organic pollutants through a non-radical pathway. Huang et al. [61] also observed that the BC/geopolymer composite membrane demonstrated an excellent degradation efficiency for tetracycline (99 %) in the Fenton-like system. This can be attributed to the abundant defect structures of BC in the membrane activating H_2O_2 to generate $\cdot\text{OH}$ through electron-donation. The porous structures and large specific surface areas of BC in the membrane can accelerate the electron transfer during the degradation process, which is conducive to the degradation of contaminants. In addition, BC-based catalytic membranes have been used in the electrochemical system to efficiently eliminate organic pollutants. As an example, iron and nitrogen co-doped BC electro-Fenton catalytic membranes demonstrated excellent removal efficiency of 100 % for tetracycline and outstanding self-cleaning properties (i.e., water flux recovery of 100 %) [29]. This can be explained by the abundant oxygen and nitrogen-containing functional groups of BC in the membrane structure, which promotes the production of H_2O_2 and improve the Fe

(III)/Fe(II) cycle, thus facilitating the generation of active species (e.g., $\cdot\text{OH}$, $\text{O}_2^{\cdot-}$) and resulting in faster decomposition of organic pollutants on the membrane surface.

Activated carbon (AC), another carbon-rich material, usually possess high microporosity, large surface area, and a wide range of pore sizes, which makes it an efficient adsorbent and supporter for water treatment applications [143]. For instance, AC-supported Zn-MOF [144], AC-supported zero-valent iron [145], and AC@ Fe_2O_3 [146] have been proven to have excellent removal efficiency for various contaminants in wastewater treatment. There are limited reports on the fabrication of AC-based catalytic membranes for eliminating pollutants. Wang et al. [147] integrated granular AC and Co_3O_4 into the ceramic membrane structures for efficient activation of PMS to degrade bisphenol A. The results indicate that the AC in the membrane channels quickly adsorbed the organic pollutants into the membrane, while Co_3O_4 effectively decomposed PMS to generate active species (e.g., $\cdot\text{OH}$, $\text{SO}_4^{\cdot-}$) to degrade the contaminants. As another example, Li et al. [96] applied powdered AC@ iron oxide nanoparticles as the catalytic layer on the membrane surface for the removal of N, N-diethyl-*meta*-toluamide in the ozone/membrane system. The reasonable removal efficiency of 60 % for N, N-diethyl-*meta*-toluamide was related to the AC trapping the organic pollutants in the reaction region, while the iron oxides accelerated the transformation of ozone molecules into hydroxyl radicals, thereby enhancing the degradation of contaminants.

5.4. Other carbonaceous materials based catalytic membranes

Carbon fiber represents one of the most promising carbon-based materials, attracting significant attention due to remarkable properties like outstanding mechanical strength, high chemical and thermal stability, as well as excellent flexibility and electrical conductivity [148]. It has also been applied as a substrate for fabricating catalytic membranes to degrade organic pollutants. Zeng et al. [149] fabricated a Zr-MOF@ Fe_2O_3 -carbon fiber catalytic membrane with an excellent removal efficiency of 100 % for phosphate in the photo-Fenton system. The high regeneration efficiency (92 %) of the membrane was ascribed to the active species generated during the photo-Fenton process, which efficiently degraded contaminants in the membrane structures to mitigate membrane fouling.

Carbon dots are another type of carbonaceous material, characterized by carbon particles of less than 10 nm, typically with a graphitic structure, and high photoluminescence and antibacterial properties [150]. Carbon dots-based catalytic membranes have been fabricated for efficient antibacterial treatment of wastewater. Zou et al. [151] observed that the nitrogen-doped carbon dots thin-film composite forward osmosis membrane displayed a higher water permeability ($26 \text{ L m}^{-2} \text{ h}^{-1}$) compared with the pristine membrane ($18 \text{ L m}^{-2} \text{ h}^{-1}$), which was mainly due to the nitrogen-doped carbon dots increasing the membrane hydrophilicity. Additionally, the hybrid membrane exhibited high antibacterial efficacy under light irradiation, which was attributed to the nitrogen-doped carbon dots acting as oxidase mimics, producing superoxide anion radical and singlet oxygen with strong oxidation ability to mitigate membrane biofouling.

In addition, graphite, as a crystalline form of carbon, typically exhibits inherent properties such as being non-toxic and having a high specific surface area. It has been extensively used as electrode materials in wastewater treatment applications [152]. For instance, Jiang et al. [153] developed a novel UV-driven electro-Fenton catalytic membrane/filtration system using TiO_2 /graphite felt as photoanode, and polytetrafluoroethylene supporting graphene and ferric oxide as cathode for the elimination of florfenicol. The synergistic effects of separation, photocatalysis, and electrocatalysis contribute to efficient degradation of the pollutants. Specifically, the photogenerated electrons and holes were efficiently separated under the bias voltage conditions, resulting in more active species being generated in the medium to enhance the degradation of pollutants.

In summary, carbonaceous materials (e.g., carbon nanotubes, biochar) are sustainable and green materials that can be embedded into membrane structures for the decontamination of organic wastewater. The synergistic effects between membrane-filtration and the catalytic degradation significantly enhance the overall treatment efficiency of organic wastewater. Therefore, carbonaceous material-based catalytic membrane systems represent a promising technique for real-world organic wastewater treatment.

6. Conclusions and perspectives

As a frontier technology for the elimination of organic pollutants from contaminated water, catalytic membranes can not only mitigate membrane fouling but also prevent the leaching of nanomaterials during the wastewater treatment process. Within the past few years, various catalytic membranes (e.g., TiO₂, Co₃O₄, MnO₂) have been investigated to improve the removal efficiency of contaminants [13]. Among the catalysts embedded in membrane structures, renewable carbonaceous-based materials represent promising alternatives due to outstanding properties including tuneable specific surface area and porosity, abundant surface functional groups, high conductivity, high mechanical strength, high flexibility and environmentally friendliness. In view of the water scarcity crisis, the efficacy of such carbonaceous materials-based catalytic membranes in water treatment applications deserves attention. Therefore, the present review summarizes and discusses the methods (namely, blending, *in-situ* growth, interfacial polymerization) employed for the fabrication of carbonaceous materials-based catalytic membranes. Subsequently, the properties and performance of these catalytic materials (e.g., CNTs, GO, BC, AC) integrated into membranes to augment water treatment applications are critically investigated. Furthermore, the challenges and future perspectives are presented as follows.

- (1) In this review, different methods employed for the fabrication of carbonaceous materials-based membranes have been summarized and discussed in detail. However, most of the preparation methods are still in the laboratory stage, and some of the synthesis processes require expensive equipment (e.g., electrospinning machine), a lot of reagents, and complex reactions (e.g., interfacial polymerization). Hence, efforts should be directed towards green, sustainable, and facile methods for the fabrication of carbonaceous materials-based membranes. Additionally, the viability and feasibility of scale-up should be explored to promote their commercialization.
- (2) The catalytic membranes can be applied in advanced oxidation processes (e.g., for the activation of persulfate, PMS, H₂O₂), ozone/membrane systems, photocatalysis/membrane systems, electrocatalysis/membrane systems, and photoelectrocatalysis/membrane systems. All these have been demonstrated to exhibit high removal efficiency of pollutants in wastewater treatment applications. However, the costs (e.g., equipment, energy, environmental impact) have not been thoroughly evaluated. Life cycle assessment will be useful to examine the systems for real wastewater treatment applications. Among these systems, the catalytic membranes used for the activation of various oxidation agents and the membrane/photocatalysis systems appear to be cost-effective for water treatment applications. In particular, photocatalysis has been recognized as a promising technology for the elimination of contaminants from wastewater due to the reliance on a natural resource, and thus photocatalytic membranes using visible light or solar light for real wastewater treatment applications seem promising.
- (3) In the carbonaceous materials-based catalytic membrane systems, the main mechanism involves generating active species to degrade the organic pollutants and mitigate membrane fouling. The degradation mechanisms of pollutants by the catalytic

membrane have been well discussed by scavenging experiments and EPR, while few studies have been conducted on degradation products. Most reports claim that pollutants are degraded into small molecules that can then pass through the membrane, thereby reducing membrane fouling. However, whether these intermediates are harmful to the environment has not been studied. In this regard, the identification of degradation products (e.g., Liquid chromatography-mass spectrometry (LC-MS)) and performance of the corresponding toxicity analysis (e.g., ecotoxicity, model predictions) should be undertaken in future research.

- (4) Although the degradation by catalysts is the dominant mechanism for pollutant removal by catalytic membranes, the properties of the membranes also play a role. The embedded carbonaceous materials can affect the membrane properties, such as surface charge, hydrophilicity, mechanical properties, and surface roughness. The understanding of the changes to the membrane properties in the degradation process remains inadequate. Therefore, the investigation of the evolving membrane properties is beneficial and necessary for further understanding and enhancing pollutant removal by the carbonaceous materials-based catalytic membrane systems.
- (5) Currently, catalytic membranes are primarily used for the degradation of single organic pollutants at the laboratory scale. It should be noted that the composition of wastewater is complex, typically with varying water matrices (e.g., Cl⁻, HCO₃⁻, NO₃⁻, and humic acid), which can affect the generation of active species and correspondingly decrease the removal efficiency for organic pollutants. In addition, real wastewater usually contains various organic contaminants and inorganic ions, which can also hinder the membrane properties. Based on the different properties of the pollutants (e.g., surface charge, electro-rich functional groups, hydrophilicity), it is possible to design catalytic membranes customized for different contaminants. In this regard, the effects of various water matrices (e.g., ions, pollutants) on the catalytic membrane system should be explored in detail. The efficiency of catalytic membranes for real wastewater needs to be assessed comprehensively.
- (6) Membrane fouling remains a challenge that restricts more extensive applications. Although different carbonaceous materials-based catalytic membrane systems (e.g., advanced oxidation process, electrocatalysis, photocatalysis) have been critically discussed in this review as effective in alleviating membrane fouling, the practicability, stability and durability of the membranes must be carefully evaluated before their commercialization. As is well known, the self-cleaning ability of the membrane is highly dependent on the active species in the medium, and these species with strong oxidation ability may also destroy the membrane structures after a prolong period. At present, most research regarding this topic is carried out on a lab scale over short time periods, so it is not clear whether these systems are suitable for industrial applications. Hence, more research on membrane fouling and stability in scaled up systems and over extended times are needed.
- (7) Hybrid systems (e.g., photocatalysis/membrane systems, electrocatalysis/membrane systems) have been demonstrated to exhibit excellent degradation efficiency for various organic pollutants. The main underlying mechanisms have been linked to the synergistic effects of separation, and photocatalysis or electrocatalysis. However, the detailed mechanisms are unclear yet. For instance, the contribution of each factor (e.g., adsorption, degradation, separation) on the removal of pollutants have not been investigated in detail, which would be important to more precisely tailor such systems. In addition, the generation pathways of the active species in the medium and the effects of each active species (e.g., selectivity) on the degradation of pollutants

should be studied in depth for a more comprehensive understanding of the involved mechanisms in such systems.

- (8) Among all the carbonaceous materials discussed in this paper, the CNTs and GO-based catalytic membranes for organic wastewater treatment applications are most promising and thus getting most attention due to their high efficiency. However, CNTs and GO are expensive, which can increase the overall costs of water treatment. In this regard, cheap and green materials (e.g., biochar) can be considered. However, efficient modification methods should be explored to improve the removal efficiency. Also, using low-cost feedstocks or wastes for the fabrication of CNTs and GO can be a promising solution to reduce the overall costs. For instance, CNTs can be synthesized from biochar under microwave irradiation [154]. Hence, novel and efficient methods should be explored and developed for the fabrication of carbonaceous-based materials with excellent properties.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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